Void formation in composite prepregs – Effect of dissolved moisture

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Abstract: Void formation as a function of resin moisture content was investigated to better understand and control process defects in composite parts made from prepreg. In this study, uncured prepreg was conditioned at 70%, 80% and 90% relative humidity and at 35 °C. Conditioned prepreg was laid up into quasi-isotropic laminates and cured using vacuum bag only (VBO) processing (low-pressure), and autoclave processing. Moisture uptake in the resin was measured using coulometric Fischer titration. Void content was measured by image analysis of polished sections of cured laminates. Void fractions increased substantially with increasing moisture content in VBO processed laminates, while autoclave-processed parts remained void-free. Experimental results were consistent with trends predicted using a diffusion-based analytical model. The findings are discussed in the context of causes of voids in prepreg composites.

Key words: Out-of-autoclave; A. Carbon fibers; A. Laminate; B. Curing; B. Porosity/Voids

1. Introduction

Voids in composite structures are known to severely degrade mechanical performance [1], [2], [3], [4], [5] and [6]. Multiple studies have addressed the causes of void formation and mitigation

measures for parts produced by autoclave processing of prepregs [1], [2], [3], [5], [7], [8], [9] and [10]. However, a new generation of prepregs have been introduced that do not require autoclave processing, known as out-of-autoclave (OOA) or vacuum bag only (VBO) prepregs. These prepregs are formulated for processing without autoclaves at much lower pressures (3–6×), a factor that has great appeal in the aerospace industry. However, the introduction of this new class of VBO prepregs raises an important question about the extent to which our understanding of void formation mechanisms and mitigation measures will translate to VBO processing. Furthermore, there is not universal agreement regarding the source of voids in prepreg processed parts, and part manufacturers typically rely on high autoclave pressures to suppress voids.

A strong and growing need exists, particularly in the aerospace industry, to find lower cost alternatives to autoclave processing, such as vacuum assisted resin transfer molding (VARTM), pultrusion, and in the present case, vacuum bag only (VBO) processing of prepregs. The appeal of VBO processing over autoclave processing stems from the lower capital investment, elimination of the need for costly nitrogen gas, greater energy efficiency, and reduction of size constraints [11], [12], [13], [14] and [15]. However, before VBO processing can be utilized on structural components, part quality must be equivalent to parts produced by autoclave processing. Because of the high pressures applied by the autoclave during the prepreg cure cycle, species dissolved in resins generally remain in solution, and void-free parts can be produced consistently. However, a vacuum bag assembly involves a relatively small pressure difference, and thus production of void-free parts presents a greater challenge. The first step in addressing the challenge is to reach a deeper and improved understanding of the mechanisms by which voids form.

The mechanisms of void formation and growth in laminates produced from prepregs are not well-understood, and opposing views are held regarding the primary cause(s) of voids. Some researchers

contend that moisture dissolved in the resin is the primary source of voids [16], while others claim that entrapped air and volatiles are the leading causes [11] and [14]. Still others suggest that all three factors play a role [1], [2], [3], [4], [6], [8], [9] and [10], although there is no consensus regarding the relative impact of each factor. However, prepregs intended for OOA processing are designed and formulated to suppress void formation from volatiles and entrapped air. For example, VBO prepregs are manufactured by a hot-melt process [14], and thus contain negligible solvent content [8] and [17]. Furthermore, VBO prepregs are designed with engineered vacuum channels to facilitate air removal [12], [13], [14], [15] and [18]. Thus, in the present work, the effect of moisture content on void formation was examined to begin to clarify the causes of voids in composite laminates produced from prepregs.

2. Experimental procedure

The material used in this study was a carbon fiber/epoxy prepreg designed for VBO processing (MTM44-1/CF5804A, Advanced Composites Group, UK), featuring a woven 2×2 twill fabric (6k Tenax HTA fiber). Control materials were fabricated by laying up 16-ply quasi-isotropic laminates 203×292 mm in the as-received condition, followed by curing in an autoclave and in atmosphere, using a standard vacuum bag assembly. To determine the effects of moisture on void formation, prepreg plies were humidity-conditioned and laid up and cured using identical processing conditions. Humidity conditioning consisted of exposure for 24 h at 70%, 80% and 90% relative humidity and at 35 °C. Control panels were also fabricated from plies conditioned in an oven at 35 °C with no added humidity (RH = 30 ± 5%) to eliminate the possibility of heat effects as a source of voids. After conditioning, plies were laid up in a [0/±45/90]2s orientation and vacuum bagged as shown in Fig. 1.
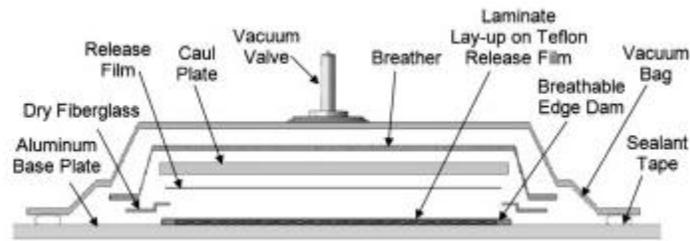
Figure 1: Vacuum bagging assembly.

The bagged assembly was then cured according to the temperature profile in Fig. 2. One set of laminates was cured under vacuum with an applied autoclave pressure of 5 atm, and one set under vacuum only. Compaction pressures were defined as the total pressure difference between the pressure within the vacuum bag and the pressure in the surrounding environment. The pressure for the vacuum bag only case was taken to be 1 atm, while the pressure in an autoclave (pressurized to 5 atm) was assumed to be a total of 6 atm.
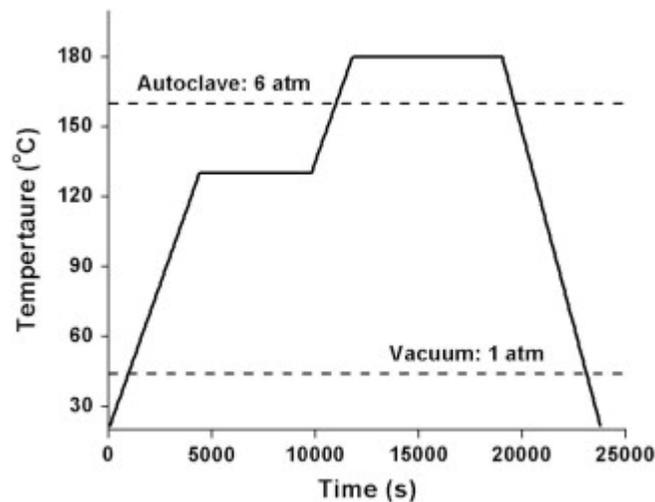

Figure 2: Cure schedule.

Image analysis was performed on polished sections of cured laminates to determine void volume fraction [1], [3], [4] and [5]. Sections were prepared from the center of cured laminates (where void content is likely to be largest) and examined using a digital stereo microscope (Keyence VHX-600).

Void volume fraction was determined by measuring the void area in the imaged cross section. The final value for volume percent of voids in each laminate was taken as an average from four samples, with each sample sectioned and polished four times to obtain a representative measurement. The thickness of the laminate at the center of each panel was also measured.

Neat resin films were humidity-conditioned and subsequently measured for weight loss by thermogravimetric analysis (TGA Q5000, TA Instruments) using a ramp rate of 15 °C/min. This weight loss was correlated with resin moisture content measured by Fischer titration using a coulmetric titrator (Mettler Toledo C20 with D0308 drying oven).

Experimental measurements of void content were compared to analytical predictions obtained using a diffusion-based void model, described in the following section.

3. Model framework

An analytical model was developed and employed to predict void formation as a function of resin moisture content. The model was based upon previous analysis of diffusion-based void growth by Kardos et al. [16]. Void growth during a thermoset cure process is time dependent, making diffusion-based analysis a natural choice. Because of the nature of VBO prepregs moisture in the resin was examined as a leading cause of voids.

The theoretical basis for the model stems from the assumption that voids grow via diffusion of water from the surrounding resin. The driving forces for this diffusion process are temperature and pressure, and diffusion can favor either void growth or dissolution, depending on the solubility of moisture in the resin and the concentration gradient [3], [9] and [10]. Void growth will occur when the pressure within a void exceeds the hydrostatic pressure in the surrounding resin. A void containing air will collapse under applied pressures, but when a void contains water, the water vapor

pressure will increase exponentially with temperature, causing the void to stabilize and grow [2], [7], [8] and [16]. Additionally, the diffusion coefficient of moisture in the resin will increase exponentially with temperature, resulting in accelerated void growth as temperature increases. Higher temperatures and lower applied pressures facilitate void growth, as both factors accelerate moisture diffusion through the resin.

To simplify the analysis, void growth was assumed to occur in a pseudo-homogeneous medium [16]. Because of this assumption, the situation resembles that of mass diffusion bubble-growth, a phenomenon that is well-understood [9], [10], [19] and [20]. Details of the assumptions of the analytical approach have been presented elsewhere [16]. The governing equations for the model define the void diameter d (mm) [2], [7], [16] and [19] and the growth driving force β [2], [7] and [16]:

$$d = 4\beta\sqrt{Dt} \qquad (1)$$

$$\beta = \frac{C_{bulk} - C_{void}}{\rho_g} \qquad (2)$$

In these equations, D (mm^2/h) is the diffusion coefficient of water in the resin, t (h) is time, Cbulk (g/mm^3) is the concentration of water in the bulk resin, Cvoid (g/mm^3) is the concentration of water at the surface of the void, and ρg (g/mm^3) is the gas density (in this case water vapor density). Examination of Eq. (2) shows that void growth will occur only if Cvoid < Cbulk. For this reason, the time and temperature for which Cvoid = Cbulk is used as a starting point for the analytical model [3] and [16].

The temperature-dependent diffusion coefficient of water used in the model calculations is given below [16].

$$D = 10.5\exp(-\frac{2817}{T}) \qquad (3)$$

Although the specific diffusivity for the selected prepreg resin is unknown, the value above (also used by Kardos et al. for a comparable epoxy system) is similar in order of magnitude to the diffusivity of water for a range of uncured epoxy resins [11] and [21].

The gas density within the void was taken to be the density of a pure water void, defined as [16]

$$\rho_g = \frac{M_{H_2O}\,p}{RT} \qquad (4)$$

where M_{H2O} (g/mol) is the molecular weight of water, p (atm) is the total pressure in the resin, which is assumed to follow the applied pressure during the cure cycle, and R (mm^3/molK) is the gas constant.

The concentration of water at the surface of the void depends on temperature and pressure and is given by [7] and [16]

$$C_{void} = 8.651 \times 10^{-17} \exp(\frac{9784}{T}) p^2 \qquad (5)$$

The final input parameter, Cbulk was determined experimentally.

4. Determination of model parameters

4.1. TGA and titration

To verify the validity of a moisture-based model, results of thermogravimetric analysis were compared to resin moisture content measured by Fischer titration. Within the margin of error, the values for wt% moisture in the resin were equivalent to the values for total sample weight loss during TGA (Table 1). The equivalence supports the assumption that escaped volatiles did not contribute appreciably to void growth.

Table 1: Values for total loss in sample wt% during TGA and wt% moisture in the sample determined by Fischer titration.

	Total wt% loss	wt% Moisture
As-received	0.26 ± 0.04	0.24 ± 0.03
35 °C 70% RH	0.61 ± 0.05	0.64 ± 0.03
35 °C 80% RH	0.78 ± 0.01	0.77 ± 0.04
35 °C 90% RH	0.95 ± 0.11	0.99 ± 0.05

4.2. Resin solubility

The value of Cbulk, the final input parameter required for the analytical model, was derived from resin solubility data. Moisture content in resin samples conditioned at selected relative humidity values was measured, and a parabolic solubility curve was fit to the data.

The parameter S in Fig. 3 is the coefficient of resin solubility obtained from the parabolic fit. Multiplying by the initial relative humidity (RHo)2 yields the solubility of the resin (wt% moisture/unit wt% resin). The coefficient for resin moisture content is defined as [16]

$$C = \frac{S}{100}\rho_r \qquad (6)$$

where ρr (g/mm³) is the density of uncured resin (1.25×10^{-3} for the selected system). Substituting the relevant values yields the equation for Cbulk

$$C_{bulk} = C(RH_0)^2 = 1.55 \times 10^{-9} RH_0{}^2 \qquad (7)$$
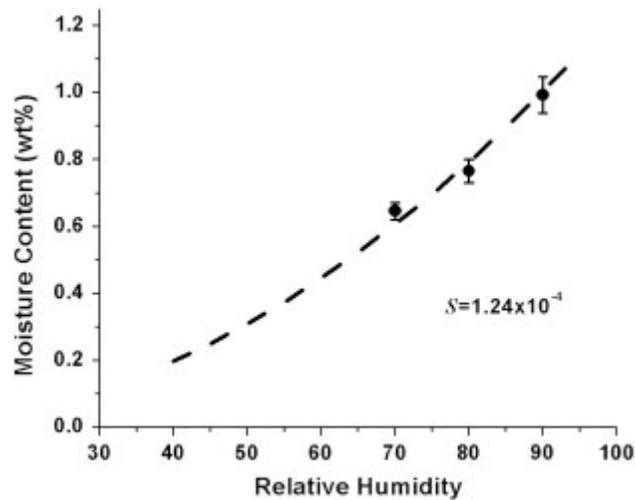
Figure 3: Moisture content as a function of relative humidity exposure with parabolic solubility fit.

4.3. Gelation

The resin gel point was determined to define an end-point for the analytical model. Resin flow ceases after gelation, and any voids remaining in the matrix are thus trapped [7], [9] and [14]. No void growth or dissolution occurs after the resin has gelled, making gelation an effective stopping point for a void formation model. To determine the gel point of the resin (MTM44-1), rheometry was performed on conditioned resin using a parallel plate rheometer (TA Instruments AR2000). Humidity conditioning altered the resin gel point, which showed a second order dependence on relative humidity. To increase the accuracy of the analytical model, this dependence was incorporated into the analysis. Fig. 4 shows the decrease in gel time with relative humidity exposure, while Fig. 5 shows characteristic rheology data for a humidity of 70%
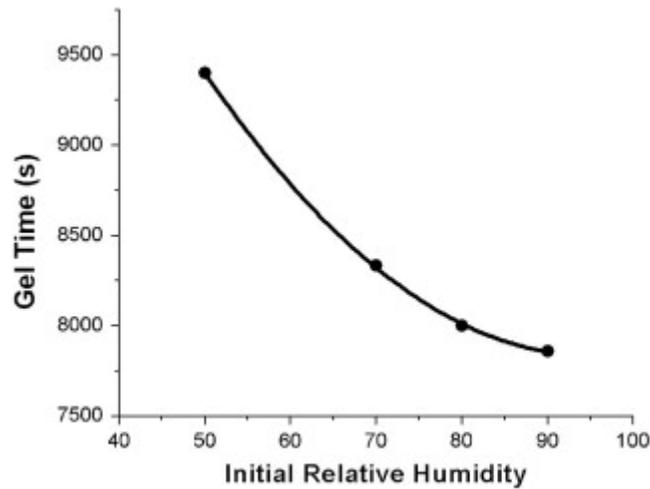
Figure 4: Gel time as a function of relative humidity exposure.
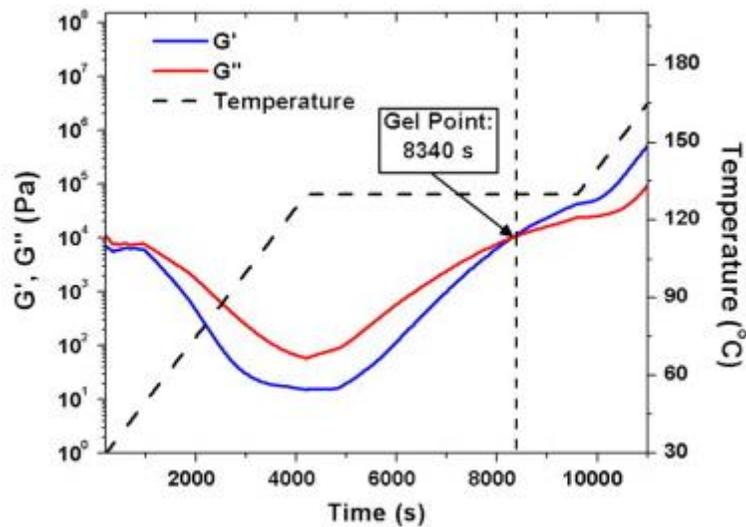
Figure 5: Rheological data (G' – storage modulus, and G'' – loss modulus) displaying resin gelation for a humidity exposure of 70%.

As shown in Fig. 5, the gel point was taken as the time when the curves of storage modulus (G′) and loss modulus (G″) intersect [22]. Using the second order fit to the gel point-humidity plot (shown in Fig. 4), the analytical model for void formation was truncated at the appropriate time for each humidity level to provide effective endpoints for void growth.

5. Results

5.1. Model prediction

Predictions for void growth were determined for VBO and autoclave processing using parameters determined for the selected system and different levels of moisture content (Fig. 6).
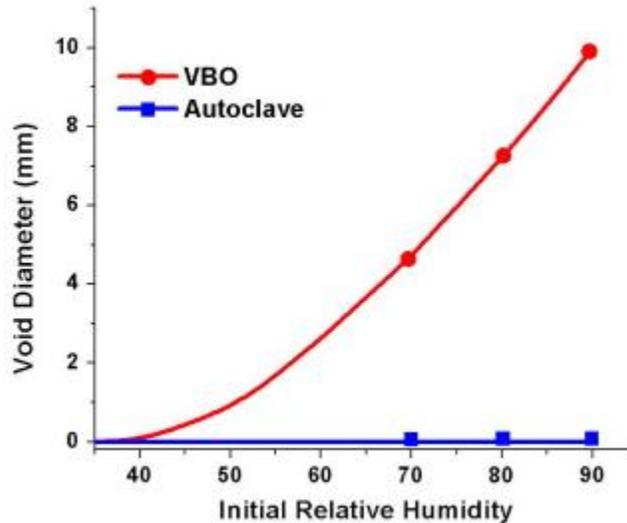
Figure 6: *Predicted void diameter for VBO and autoclave processing as a function of initial relative humidity exposure.*

The model-based calculations predicted an exponential dependence of void diameter on initial relative humidity exposure for VBO processed laminates. In contrast, no void formation was predicted for the case of autoclave processing, as the increased pressure prevents the $C_{void} < C_{bulk}$ condition from being met, eliminating void formation and growth.

5.2. Void content

To determine the accuracy of the model-based predictions, a series of experiments was performed. Panels were fabricated using prepreg conditioned at different relative humidities, then cured using VBO and autoclave techniques. Polished sections were then prepared and examined to determine void content. Characteristic images for each laminate are displayed in Fig. 7 and Fig. 8, along with

measured values of void volume fraction and laminate thickness. For the VBO laminates, micrographs were processed to enhance void visibility and measurement by image analysis. These micrographs (Fig. 7) show void content and morphology, with void content generally increasing with dissolved moisture content. The voids were typically 0.5–2 mm.
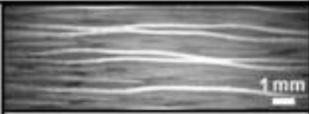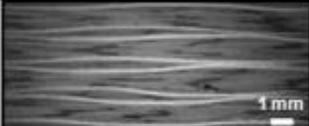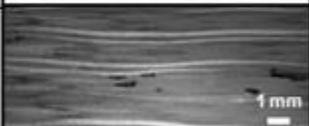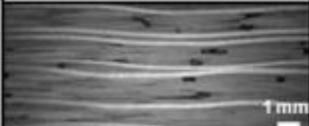

VBO Laminates		Void Content (Vol %)	Thickness (mm)
As Received		<<.1	5.08 ± .01
70% RH		.08 ± .08	5.13 ± .01
80% RH		1.00 ± .29	5.31 ± .05
90% RH		2.62 ± .48	5.48 ± .02

Figure 7: Micrographs of laminates conditioned under different initial relative humidites and cured using VBO processing. Original micrographs are displayed along with processed black and white images used for image analysis. Tabulated data presents void volume fraction and laminate thickness.
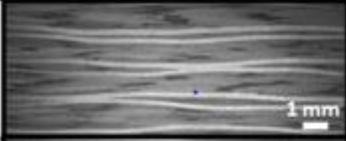
Figure 8: Micrographs of autoclave processed laminates conditioned under different initial relative humidities, and tabulated void volume fraction and laminate thickness data.

5.3. Measured vs. predicted

To compare the model prediction to the experimentally determined void content, the predicted void diameters were converted to void volume fractions. By modifying Eq. (1), the following expression for void volume fraction was derived [7]

$$Vol\% = \frac{V_v}{V_m} = \frac{\pi d^3}{6V_m} = \frac{\pi[4\beta(Dt)^{\frac{1}{2}}]^3}{6V_m} \tag{8}$$

where Vm (mm^3) is the unit matrix volume, which is used to scale the model results. This unit matrix volume represents the volume of resin required to produce an observed void volume fraction with a single void of a given diameter [7]. The value of Vm is expected to remain constant for a given resin system [7]. Using predicted void diameter values combined with measured values of void volume fraction (Vv), a characteristic Vm value for the resin was determined. The plot in Fig. 9 shows the

measured void fractions plotted as a function of initial humidity values, as well as the dependence

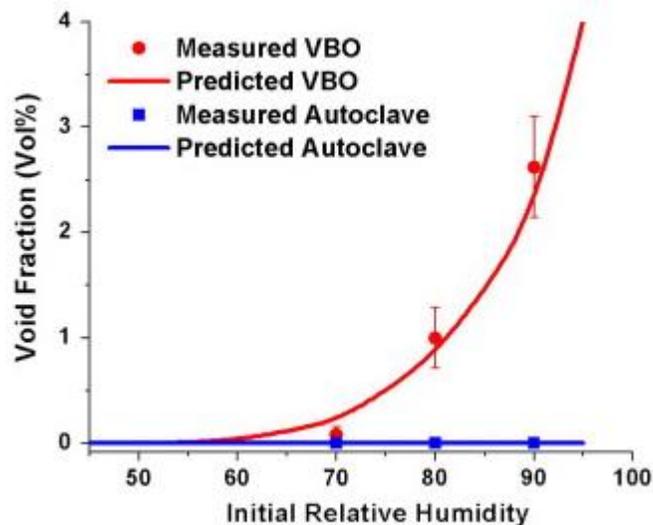predicted from the diffusion model.

Figure 9: Predicted and measured void volume fraction data.

6. Discussion

A fundamental question motivating the present work concerned the origin of voids in composite laminates

produced from prepregs. The nearly identical values of weight loss determined by TGA and Fischer titration

(Table 1) support the conclusion that in the present study, the weight loss can be attributed almost entirely to

moisture. Thus, any volatiles present in the resin system exist in negligible quantities, and the contribution of

volatiles to void formation can be neglected. Furthermore, the engineered vacuum channels present in the

prepreg, which are designed to provide pathways for evacuation of entrapped air, appear to be effective in

doing so, and there is no evidence that the voids observed here can be attributed to entrapped air. Elimination

of these two potential sources of voids leaves dissolved moisture as the only viable source, justifying the use

of a moisture-based model to predict void formation. Although the moisture content in the prepreg appears

relatively small when expressed as a weight percent, it represents a much larger mole percent and a significant

potential volume fraction of water vapor, indicating that dissolved moisture is a potential cause of void formation [3] and [8].

The measured void fraction dependence on moisture content is consistent with the model predictions, as shown in Fig. 9. Because the model predictions have been scaled, the measurements constitute validation of the predicted dependence of void content on moisture content (as opposed to the specific values). Note that the model calculations predict an absence of voids in autoclave-processed parts, and the prediction was corroborated by examination of polished sections. This finding has also been reported in a previous study examining void content in autoclave cured prepreg materials [18]. Autoclave pressures effectively force moisture to remain in solution, thereby preventing the formation and expansion of voids [9] and [14]. However, the safeguard supplied by high external pressures is absent in VBO processing, where compaction and curing occurs under atmospheric pressure, rendering laminates more susceptible to void formation from dissolved moisture.

To illustrate this issue, void pressure as a function of moisture content was examined. The following equation was derived by equating Eqs. (5) and (7) and solving for pressure [16]

$$p \geq 4.233 \times 10^3 \exp\left(-\frac{4892}{T}\right) RH_0 \qquad (9)$$

In this expression, the left hand side represents the applied compaction pressure, while the right hand side represents the water vapor pressure in a void as a function of temperature and relative humidity exposure. As described previously, void growth will take place only when Cvoid < Cbulk, so when the above inequality (Eq. (9)) is satisfied, void growth via moisture diffusion cannot occur [16].

To illustrate the dependence of void pressure on temperature during the cure cycle, a void pressure map was created (Fig. 10). Pressure dependence on temperature was plotted for two relative humidities, 100% and 45%. To prevent void formation and growth, the applied compaction pressure must exceed the water vapor pressure [3], [8] and [16]. The 100% RH curve represents an upper

bound on void pressure, while the 45% RH curve is included to display the maximum moisture

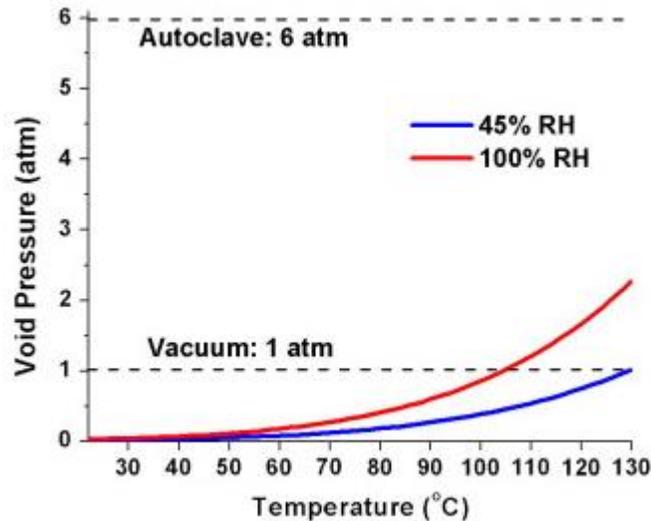content that can be successfully held in solution by vacuum pressures.

Figure 10: Void pressure as a function of initial relative humidity exposure.

This graph illustrates the effectiveness of autoclave pressures in suppressing void growth, as well as

the sensitivity of VBO processing to resin moisture content. A relative humidity of 45%, using the

solubility curve defined earlier, corresponds to a resin moisture content of approximately 0.25 wt%.

The resin in the as-received condition typically has a moisture content of 0.24 ± 0.03, a level slightly

greater than that which can be controlled by atmospheric pressure alone. After an out time of 24 h

in an uncontrolled lab environment (RH $= 50 \pm 5\%$), this level increased to 0.30 ± 0.01. Manufacture

of large parts often requires several days of out time for cutting and lay-up. Consequently, control

of the ambient humidity may be necessary to ensure part quality when manufacturing parts with

VBO processing.

Despite this concern, void-free laminates were consistently produced using VBO processing.

Laminates that were laid up and cured using the as-received prepreg exhibited void contents and

thicknesses identical to those cured in the autoclave. The findings demonstrate that void-free

laminates can be achieved by VBO processing using appropriate protocols. Additionally, laminates fabricated from VBO prepregs exposed to a temperature of 35 °C for 24 were void-free, confirming that dissolved moisture, not temperature exposure, was the source of voids in humidity conditioned laminates.

Note that although the diffusion-based model provides an accurate fit to the experimental data, there are limitations to the approach used here. The addition of moisture to epoxy resins affects material properties, often unavoidably, as in the case of gel time. Such properties include tack, resin reactivity, and resin viscosity [23] and [24], and all are likely to influence void formation. However, the goal of this work was to test the hypothesis that resin moisture content affects void formation. Additionally, our measurements were performed using small flat panels, and thus the contribution of entrapped air was neglected. With large and/or contoured parts, pathways for escape of entrapped air are generally longer and more tortuous. Factors such as sample geometry and part size are expected to affect gas removal and must be investigated to develop a more complete picture of void formation in VBO-processed composites.

7. Conclusions

Void formation was examined as a function of resin moisture content for both autoclave and VBO processed laminates. During autoclave processing, high pressures were sufficient to suppress void formation, while with vacuum bag only processing void volume fraction increased exponentially as a function of moisture content. The findings demonstrated that the production of void-free parts is possible with VBO processing, but the absence of high external pressures requires careful control of the lay-up environment to prevent excessive moisture uptake in the resin. A diffusion-based model of moisture-driven void formation was consistent with experimental data for small flat panels.

Both analysis and experimental measurements demonstrated that while autoclave pressures were sufficient to suppress void formation in the presence of dissolved moisture, low-pressure processing was more sensitive to moisture content, resulting in voids with high levels of dissolved moisture. Other manufacturing parameters are likely to give rise to similar effects, and low-pressure processing will require special care and attention to suppress void formation. One potential issue that must be considered is the extent to which vacuum bag processing can be successfully translated to the manufacture of larger parts. With the application of autoclave pressures, large parts generally can be processed without voids, largely because the high pressures suppress the evolution of dissolved species. However, the pressure differences associated with VBO processing are much lower, and thus careful control of process parameters will be more critical, particularly for production of complex parts. Examples of such parameters include site-specific vacuum levels and breathe-out distances. With large or complex parts maintaining full vacuum at all locations throughout the cure is often difficult. Additionally, for large part production, pathways for air and volatiles to escape must be maintained over longer distances. The longer the pathway to a vent (breathing edge), the more difficult it becomes for entrapped air to exit the part [18].

The present investigation of moisture effects suggests that in controlled lab environments, moisture can be eliminated as a significant source of voids in flat panels. This result sheds light on the controversy surrounding the role of moisture content vs. entrapped air as a leading source of voids, suggesting that small flat laminates produced from prepreg stored in a controlled environment will not develop voids as a result of moisture content. While these findings contribute to the general understanding of void formation, the role of other factors, most notably pressure and size effects, remain unclear. Systematically addressing additional factors affecting void formation will lead to a

better understanding of void formation in prepreg processed parts, and clarify the limits of VBO processing for composites manufacturing.

References:

1. L. Liu, B. Zhang, D. Wang, Z. Wu, *Effects of cure cycles on void content and mechanical properties of composite laminates*. Compos Struct, **73** (2006), pp. 303–309
2. L. Liu, B. Zhang, Z. Wu, D. Wang, *Effects of cure pressure induced voids on the mechanical strength of carbon/epoxy laminates*. J Mater Sci Technol, **21** (1) (2005), pp. 87–91
3. P. Olivier, J.P. Cottu, B. Ferret, *Effects of cure cycle pressure and voids on some mechanical properties of carbon/epoxy laminates*. Composites, **26** (7) (1995), pp. 509–515
4. H. Huang, R. Talreja, *Effects of void geometry on elastic properties of unidirectional fiber reinforced composites.* Compos Sci Technol, **65** (2005), pp. 1964–1981
5. J. Tang, W.I. Lee, G.S. Springer, *Effects of cure pressure on resin flow, voids, and mechanical properties.* J Compos Mater, **21** (1987), pp. 421–440
6. M.L. Costa, S.F.M. Almeida, M.C. Rezende, *The influence of porosity on the interlaminar shear strength of carbon/epoxy and carbon/bismaleimide fabric laminates*. Compos Sci Technol, **61** (2001), pp. 2101–2108
7. F.Y.C. Boey, S.W. Lye, *Void reduction in autoclave processing of thermoset composites part 1: high pressure effects on void reduction*. Composites, **23** (4) (1992), pp. 261–265
8. F.C. Campbell, A.R. Mallow, *Porosity in carbon fiber composites, an overview of causes*. J Adv Mater (1994), pp. 18–33
9. J.R. Wood, M.G. Bader, *Void control for polymer–matrix composites (1): theoretical and experimental methods for determining the growth and collapse of gas bubbles.* Compos Manuf, **5** (3) (1994), pp. 139–147
10. J.R. Wood, M.G. Bader, *Void control for polymer–matrix composites (2): experimental evaluation of a diffusion model for the growth and collapse of gas bubbles.* Compos Manuf, **5** (3) (1994), pp. 149–158
11. S.S. Tavares, V. Michaud, J.A.E. Manson, *Through thickness air permeability of prepregs during cure*. Composites: Part A, **40** (2009), pp. 1587–1596
12. S. Thomas, S.R. Nutt, *In situ estimation of through-thickness resin flow using ultrasound.* Compos Sci Technol, **68** (2008), pp. 3093–3098
13. S.S. Tavares, V. Michaud, J.A.E. Manson, *Assessment of semi-impregnated fabrics in honeycomb sandwich structures.* Composites: Part A, **41** (2010), pp. 8–15
14. Jackson K, Crabtree M, *Autoclave quality composites tooling for composite from vacuum bag only processing*. In: 47th International SAMPE symposium; 2002. p. 800–7.
15. Repecka L, Boyd J. *Vacuum-bag-only-curable prepregs that produce void-free parts*. In: 47th International SAMPE symposium; 2002. p. 1862–75.
16. J.L. Kardos, M.P. Dudukovic, R. Dave, *Void growth and resin transport during processing of thermosetting-matrix composites.* Adv Polym Sci, **80** (1986), pp. 102–123
17. M. Molyneux, *Prepreg, tape and fabric technology for advanced composites*. Composites, **14** (2) (1983), pp. 87–91

18.   Thorfinnson B, Biermann TF, *Measurement and control prepreg impregnation for elimination of porosity in composite parts*. In: Fabricating composites conference proceedings, Philadelphia, Pennsylvania; 1988. p. 534-1–534-14.

19.   L.E. Scriven, *On the dynamics of phase growth*. Chem Eng Sci, **10** (1/2) (1959), pp. 1–13

20.   R.S. Subramanian, M.C. Weinberg, *The role of convective transport in the dissolution or growth of a gas bubble*. J Chem Phys, **72** (12) (1980), pp. 6811–6813

21.   DeIasi R, Whiteside JB, *Effect of moisture on epoxy resins and composites*. Adv Compos Mater-Environ Effects. ASTM; 1978. p. 2–20.

22.   C.Y.M. Tung, P.J. Dynes, *Relationship between viscoelastic properties and gelation in thermosetting systems.* J Appl Polym Sci, **27** (1982), pp. 569–574

23.   K.J. Ahn, L. Peterson, J.C. Seferis, D. Nowacki, H.G. Zachmann, *Prepreg aging in relation to tack.* J Appl Polym Sci, **45** (1992), pp. 399–406

24.   Z.N. Sanjana, W.H. Schaefer, J.R. Ray, *Effect of aging and moisture on the reactivity of a graphite epoxy prepreg.* Polym Eng Sci, **21** (8) (1981), pp. 474–482