IN-SITU MONITORING OF AIR REMOVAL DURING VACUUM BAG-ONLY (VBO) PROCESSING

Wei Hu, Steven Nutt

M.C. Gill Composites Center, University of Southern California
3651 Watt Way, VHE-604
Los Angeles, CA, 90089

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ABSTRACT

Composites parts cured out-of-autoclave (OOA) sometimes exhibit bulk porosity, resulting in degraded mechanical properties. The primary origins of voids are dissolved volatiles, especially moisture, and air entrapped mechanically during the lay-up. However, the mechanisms of evolution and removal of voids are still not fully understood. In this study, we implement an *in-situ* monitoring technique to dynamically observe air removal during the cure process. We present a parametric study of void evolution in unidirectional prepregs during vacuum bag-only cure using the *in-situ* observation technique. Effects of process parameters, including room-temperature vacuum hold time, vacuum level, and resin moisture content, are evaluated in this study. Based on the observations of bubble migration, growth, and dissolution during cure, the effects of the key manufacturing parameters on void evolution are determined, yielding a clearer understanding of air removal mechanisms.

1 INTRODUCTION

In recent decades, the growth of the aerospace sector and increasing demand for composites in aircraft primary structures have motivated the shift from autoclave processing toward more cost- and time-efficient alternatives for composites manufacturing. Out-of-autoclave (OoA) prepreg processing is one such approach, which allows the manufacture of composite parts using vacuum bag-only (VBO) consolidation in conventional, unpressurized ovens. By eliminating the need for autoclaves, OoA prepreg processing can reduce acquisition and operating costs and enable the manufacture of larger parts at faster rates. However, since VBO processing involves a maximum consolidation pressure of only 0.1 MPa (1 atm), compared to up to 0.5-1 MPa (5-10 atm) in autoclaves, large composite parts can sometimes exhibit significant porosity, which degrades mechanical properties. Thus, strict science-based materials processes and protocols must be developed to avoid void formation during VBO processing.

Void formation in OoA prepreg processing has been extensively reported [1–4]. Primary void sources include air entrapped between prepreg plies during lay-up, and volatiles released from the resin during processing [5]. To facilitate air removal, OoA prepregs are partially impregnated by design, with dry fiber tows surrounded by resin-rich regions. To achieve low porosity, a room-temperature vacuum hold is used before cure to remove entrapped air. Then, laminates are cured at high temperature and, during heat-up, resin gradually infiltrates the dry fiber tows, resulting in a low-porosity part. If air is not removed before consolidation, it will remain in the composite as voids. Similarly, if resin does not fully saturate the fiber bed, the part will exhibit pervasive dry regions. For OoA prepregs, vacuum quality is important, as it is the sole driving force for air evacuation and fiber bed compaction, and strongly influences to resin infiltration. However, in practice, vacuum quality is often substandard due to an inadequate vacuum system or poor bagging [6]. The effects of reduced vacuum level on laminate quality have been studied [6,7], with results showing that lower levels have detrimental effects on void content.

Moisture is a primary volatile released during cure. Polymer resins absorb moisture from the ambient – for example, when exposed to uncontrolled environmental conditions during storage and lay-up. Grunenfelder et al. [8] compared the effects of relative humidity on porosity in both autoclave and out-of-autoclave processing. Prepreg plies were humidity-conditioned at different relative humidity levels before curing. The results showed that void content increased with increasing moisture content in VBO processed parts, while the autoclave-processed parts remained void-free. An explanation for this is that an increase of moisture content results in an increase in water vapor pressure in the voids, which leads to void growth if the water vapor pressure exceeds its surrounding resin pressure. The high pressure applied during autoclave cure can suppress moisture in resin, while VBO processing is more sensitive to moisture and relative humidity levels.

Despite this prior work, a detailed understanding of the precise mechanisms for void evolution (nucleation, growth, collapse, and/or migration) is limited. In the past, most parametric studies only described and compared final part quality, with analysis impeded, in most cases, by the difficulty of real-time monitoring. In previous work [9], we developed an *in-situ* monitoring method to investigate the formation and removal mechanisms of inter-ply gas-induced voids in unidirectional VBO prepreg. This is accomplished by incorporating a perforated resin film between a glass tool plate and a stack of prepreg plies. In this way, we intentionally introduce trapped air into the lay-up, accurately mimicking the conditions surrounding an internal void located inside a stack of prepreg plies. This method resulted in key insights into bubble migration, expansion, and removal during VBO cure. Here, we conduct parametric studies to study the effects of key processing parameters on gas transport and air removal using the same technique. The key objectives of the study are to establish correlations between process parameters and void evolution and to identify science-based void reduction approaches.

2 EXPERIMENTAL

2.1 Materials

Experiments were performed using a carbon fiber/epoxy prepreg formulated for vacuum bag-only cure. The prepreg consisted of a toughened epoxy (CYCOM 5320-1, Cytec Solvay, USA) and a unidirectional tape (IM7 12K, 145 g/m^2) with 33% resin content by weight. The recommended cure cycle for this material is 93°C (3 h) or 121°C (12 h). In this study, the lower temperature cycle was used, with an average ramp rate of 2°C/min.

2.2 Test Matrix

Table 1 outlines the manufacturing parameters considered. A 24-hour vacuum hold at 25°C was performed before cure to investigate air evacuation at ambient conditions. The influence of vacuum quality was investigated by fabricating laminates using full vacuum (denoted as 99 kPa, and corresponding to an absolute bag pressure of ~2.3 kPa) and 80% vacuum (denoted as 80 kPa, and corresponding to an absolute bag pressure of ~21.3 kPa). The bag pressure was monitored throughout cure using a pressure sensor. Finally, prepregs were conditioned at different relative humidity levels to determine the effect of moisture content on void evolution. Prepreg plies were conditioned for 24 h within a humidity chamber filled with saturated K_2SO_4 solution. The saturated K_2SO_4 solution provided a constant relative humidity level of 99%, confirmed by a digital humidity sensor. Prepreg plies were also dehydrated for 24 h in a desiccator with a constant relative humidity of 1%. The moisture level of each sample was measured by Fischer titration using a coulometric titrator (Mettler Toledo C20 with D0308 drying oven). Void evolution during cure was investigated using *in-situ* monitoring for all conditions. Within each experiment, only one parameter was varied. A control panel was fabricated under full vacuum without room-temperature vacuum hold and humidity conditioning. Each laminate cured under different conditions was compared to the control panel.

Table 1 Parameters included in this study

Parameters	**Range**
Room temperature vacuum hold	0-24 h

Reduced vacuum	80 kPa, 99kPa
Humidity condition	1% RH, 99% RH for 24 h

2.3 In-situ monitoring method

Transparent glass tools allow real-time observation of air entrapment and void distribution during cure [10,11]. In this work, a custom-built experimental setup was used [9]. A perforated resin film with holes of controlled size and distribution was introduced into the lay-up to mimic the condition of air bubbles trapped between prepreg plies. The perforated resin film was laid up against the glass window of an oven, followed by four layers of prepreg plies and standard consumables, shown in Figure 1. In this way, air bubbles were intentionally introduced into the lay-up. The prepregs were then cured in a conventional air-circulating oven. Temperature was monitored by thermocouples, and time-lapse videos were recorded using a portable microscope.

To produce test panels, prepreg plies were cut to 127 mm × 127 mm and resin films 38 mm × 38 mm. The films were composed of the same resin as the prepreg (CYCOM 5320-1) with a thickness of ~50 µm. To create perforated resin films, holes were punched into the film manually using a coring tool with a diameter of 0.25 mm and spacing between holes of 2 mm (Fig. 1b).

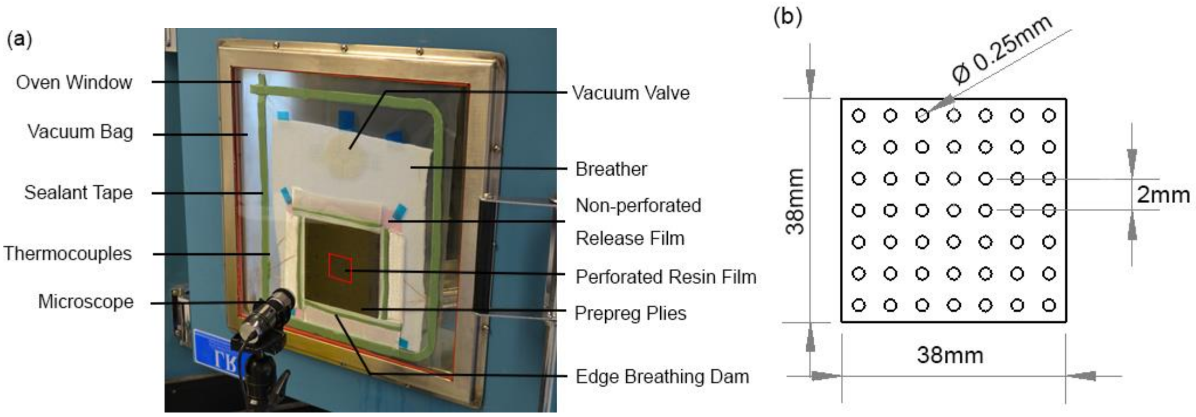

Fig 1. Schematic of (a) in-situ monitoring set-up (b) perforated resin film.

3 RESULTS AND DISCUSSION

3.1 Vacuum hold effects

Room-temperature vacuum hold is a potentially important step during OoA prepreg processing for removal of entrapped air. With conventional prepregs, hold times of 16 hours or more can be required for large composites parts. In this study, a 24-hour room-temperature vacuum hold was performed before cure. The evolution of air bubbles is shown in Fig. 2. Initially, air was trapped both in the artificial pores created using the coring tool (Fig. 2a circled in red), and between the perforated resin film and the first prepreg ply (white regions in Fig. 2a). After the vacuum hold, the naturally trapped air pockets mostly disappeared, while the artificial pores did not change appreciably in position and size (Fig. 2b).

Fig. 3 shows the void content as a function of time. During the first two hours, the total void content decreased from ~13% to ~7%. After that, air evacuation slowed down and a final void content of ~4% was achieved after 24 h. These observations indicate that inter-ply air evacuation in unidirectional prepregs is a slow process. At room temperature, as the resin viscosity is relatively high and fiber tows is partially dry, the naturally trapped air can occasionally get access to the dry fiber pathways via resin-starved regions (Fig. 2c) on the prepreg surface, while the larger artificial air bubbles remain stationary.

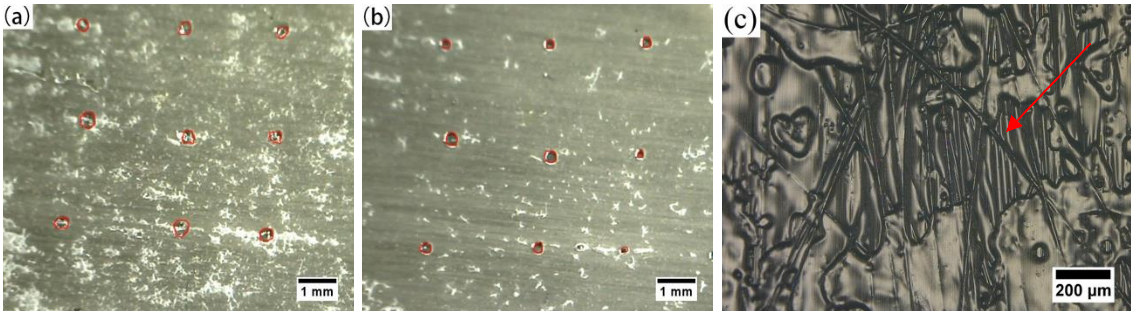

Figure 2 Time-lapse images: a) initial state; b) after 24-hour vacuum hold and c) micrograph of the surface of an uncured unidirectional prepreg.

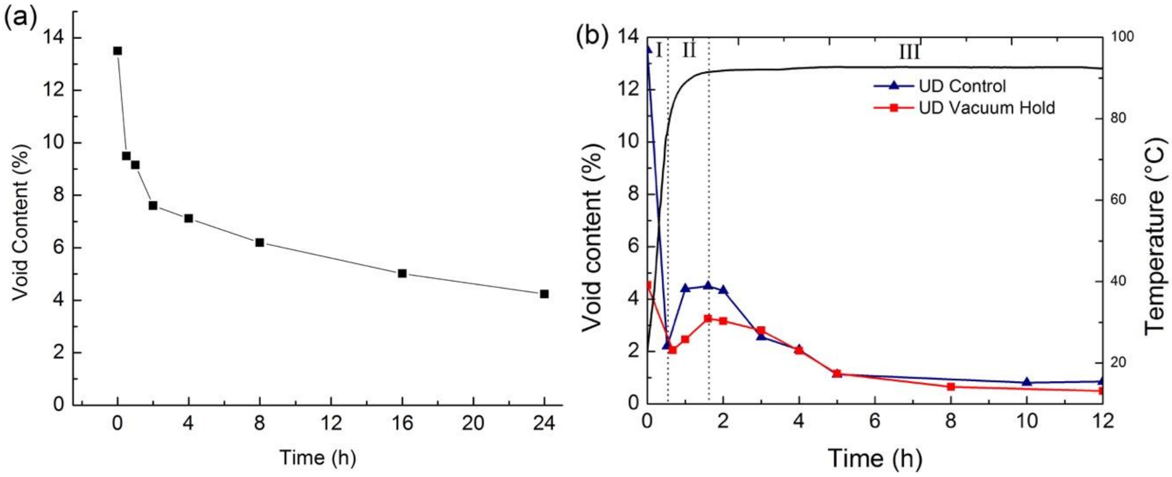

Figure 3 Void content of UD prepregs as a function of time (a) during 24-hours room temperature vacuum hold; (b) during the elevated cure cycle.

After the 24-hour vacuum hold, laminates were heated using the cure cycle shown in Fig. 3b, and bubble migration throughout cure was documented (Fig. 4). In previous work [9], we described a three-stage air removal mechanism based on bubble behavior, resin properties, and tow impregnation during cure of unidirectional prepregs. These three stages are (I) air evacuation, (II) bubble expansion, and (III) bubble shrinkage, denoted with Roman numerals and dashed lines in Fig. 3b. All stages were observed at roughly the same time during cure of prepregs having undergone a room-temperature (RT) vacuum hold. During Stage I, as the temperature increased, resin began to flow, and further air evacuation was achieved. At the end of the first stage, the void content at the visible surface was similar to that of the control panel. During Stage II, however, a smaller increase in the void content of room temperature-evacuated prepregs was observed compared to the control panel. Bubble expansion is attributed to moisture vaporization. One possible reason for the reduction in void content could be a reduction in moisture content. However, this is unlikely, as titration tests before and after 24 h RT vacuum hold showed no significant decrease in moisture content. Comparison of void distributions in both prepregs at the end of Stage II indicates that although the total void contents were similar, more small, discrete air pockets existed in the control panel. As moisture will tend to diffuse into pre-existing air pockets [10], more air pockets provide more hosts for moisture release in the control panel. The final void content of prepregs cured after a 24-hour hold was only slightly less than that of the control panel, indicating that a RT vacuum hold is not an efficient way for inter-ply air evacuation in UD prepregs. Super-ambient dwells are sometimes recommended by prepreg manufacturers to promote air evacuation. A relatively high temperature (usually 50°C-60°C) is used during super-ambient dwells to effectively remove air by reducing resin viscosity [12].

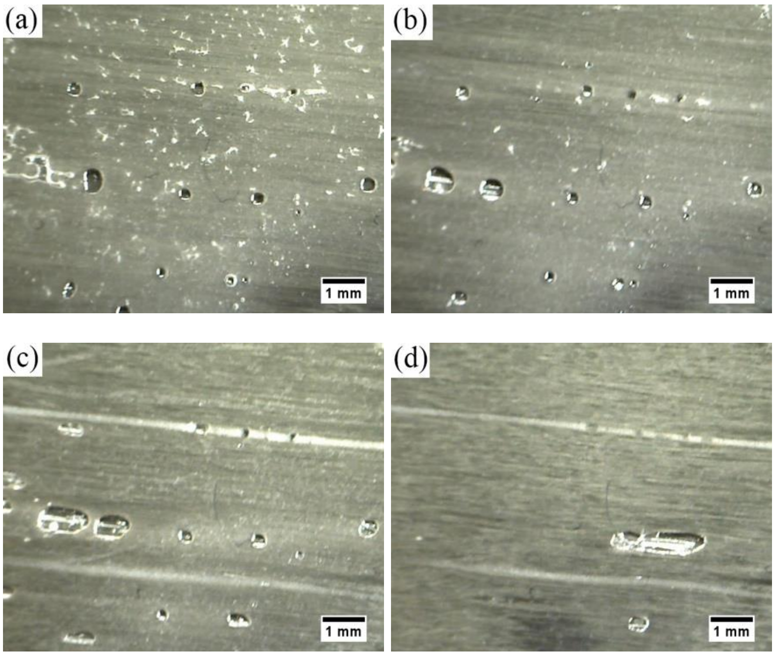

Figure 4 Time-lapse images of void evolution in unidirectional prepregs during heated cure. Images were taken at a) initial state, after 24h RT vacuum hold, b) 40min, c) 95min, d) 12h.

3.2 Bag pressure effects

The influence of vacuum quality on void evolution in UD prepregs is shown in Fig. 5. Two vacuum levels were used in this study – 80% (bag pressure of 21.3kPa) and 100% (~ 2kPa). At the end of the first stage, the void content in both prepregs decreased to a similar value, indicating that inter-ply air evacuation under 80% vacuum remained efficient. Bubble expansion also began at about 80°C under 80% vacuum. However, while the void content under full vacuum decreased after 1 hour of heated cure, the void content of prepreg cured under 80% vacuum kept increasing for another hour. The void content after expansion was 50% greater than that of the control panel. This difference indicates that during Stage II, prepregs are more sensitive to pressure deficiency compared to the Stage I. As discussed before, bubble expansion results from gas pressure in the voids exceeding the surrounding resin pressure. The explanation for the longer expansion time is that the decrease in consolidation pressure results in a drop in resin pressure. While the exact mechanism requires further investigation, observations indicate that strict control of vacuum quality is particularly important during the Stage II. Interestingly, the final void content under both conditions is almost identical.

Internal void content was also examined, and the results showed that both laminates were free of inter-ply voids. During Stage III, air bubbles shrink as water solubility increases with increasing degree of cure [9]. As the cure process progresses, moisture in voids dissolves into the resin again. Initially, prepregs in both cases should have similar moisture content. Although void content after Stage II increased, resin was still able to absorb most of the moisture at the end of cure.

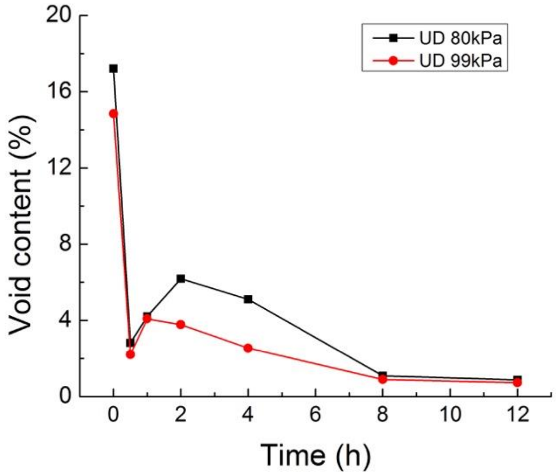

Figure 5 Void evolution during cure under reduced vacuum level.

3.3 Moisture effects

The effects of moisture on void removal were investigated. Images of the cured conditioned prepreg laminates are shown in Fig. 6. Both void size and density increase as the moisture content in the resin increases. Fig. 7 shows that prepreg conditioned at 99% RH (wet prepreg) exhibited the highest porosity, while prepreg dehydrated at 1% RH (dry prepreg) contained the lowest void content. At the end of Stage I, the observed void content in wet prepreg was about 3 times higher than that measured in dry and control prepregs. This increase is associated with the increased tack of wet prepreg, which promotes air entrapment and inhibits air evacuation. During Stage II, air bubbles in wet prepregs also underwent expansion for a longer period. Table 2 shows the moisture content in each prepreg after conditioning. Although the amount of moisture in the prepreg appears small when expressed as a weight percentage, it represents a significant potential volume fraction of water vapor. The moisture content in wet prepreg was measured to be 4 times greater than that of the control panel, and 6 times greater than that of dry prepreg. The void growth in wet prepreg occurred via diffusion of water from the surrounding resin [8]. Diffusion can favor either void growth or dissolution, depending on the solubility of moisture in the resin and the concentration gradient of water. As the moisture content in resin increases, the water concentration difference between the void and resin increases, resulting in more water vapor migrating into voids. The final void content of wet prepregs was ~ 4 times greater than that of the control panel.

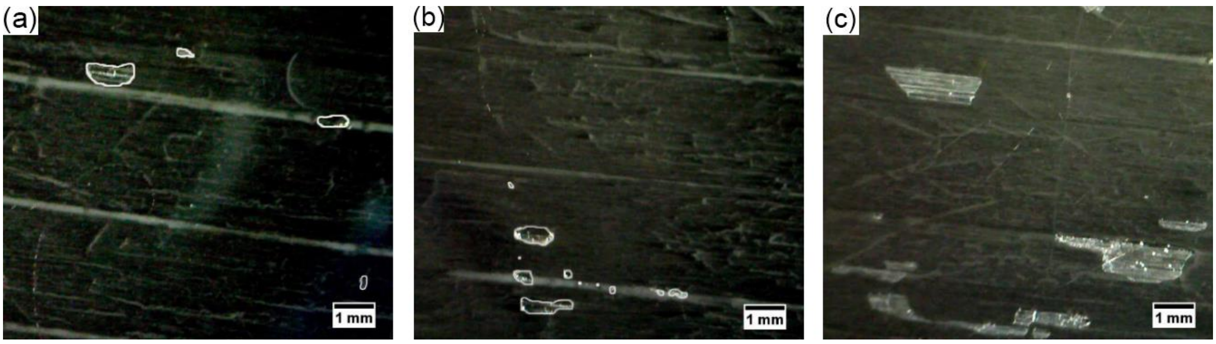

Figure 6 Effects of moisture on void removal during cure. Images were taken at the end of cure for prepregs humidity conditioned at a) 1%RH, b) 40%RH, c) 99%RH for UD prepregs, respectively.

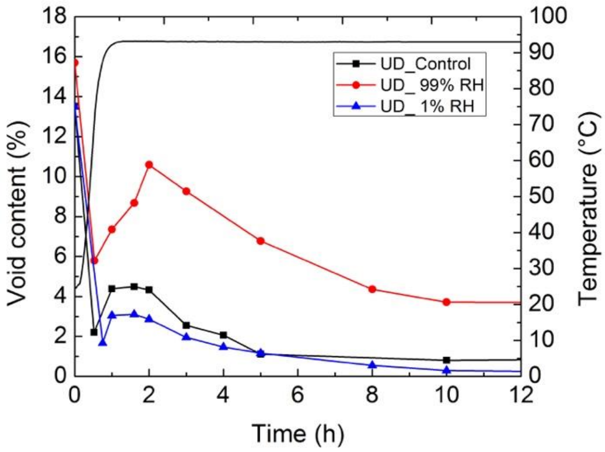

Figure 7 Void content as a function of time during OoA cure.

Table 2 Moisture content of UD prepreg determined by Fischer titration

Samples	UD
Dry (1%RH)	0.063±0.016
Control (40%RH)	0.103±0.013
Wet (99% RH)	0.394±0.020

3.4 Bubble expansion

The effects of process parameters on the expansion of individual bubbles were also investigated. While overall porosity data provides important information about the final part quality (which is related to mechanical performance), data on individual bubble expansion rate provides insight into the mechanisms through which processing parameters influence defect formation. Fig. 8 shows the average areal expansion rate under each condition. At least 20 individual bubbles of each test were tracked, and the area of each bubble before and after expansion was measured. The large variability in the dataset indicates that bubble expansion depends on the specific characteristics and environment surrounding each bubble. The average bubble expansion rate of prepregs after vacuum hold was similar to that measured in the control panel, further indicating that RT vacuum holds will not reduce the moisture content in the resin. (Though air in the voids will also expand according to the ideal gas law, its effects in both cases should be similar, as the temperature profile was similar.) Increased moisture contents and reduced vacuum levels increased expansion rates by increasing gas pressure in voids or decreasing the resin pressure, respectively. Although comparable expansion rates were measured, prepregs cured under 80kPa exhibited much less porosity than humidity-conditioned prepregs, indicating that moisture acts as the dominant parameter affecting final porosity.

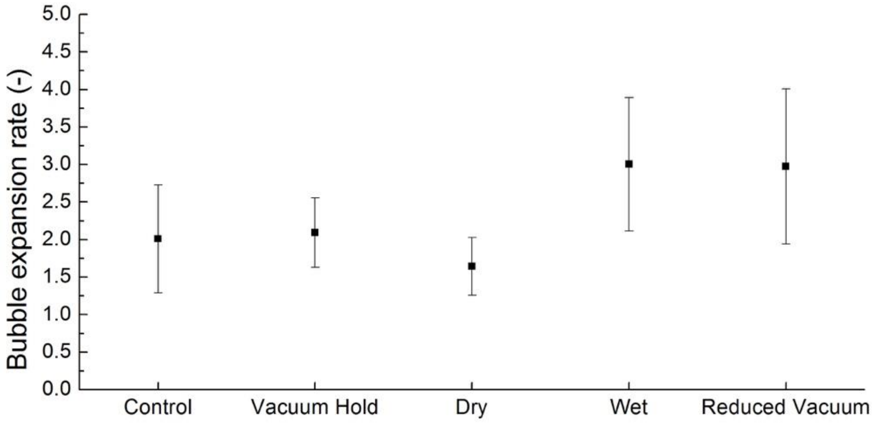

Figure 8 The effect of process parameters on bubble expansion rate.

4 CONCLUSIONS

In this study, *in-situ* visualization was used to monitor air removal during cure of VBO prepregs under different processing conditions. This technique provided insights into air removal mechanisms. A three-stage void evolution process, previously described, was observed in all tests, demonstrating that this technique can reliably capture key phenomena associated with inter-ply void evolution.

The results increase the understanding of fundamental mechanisms of air removal, and can be used to optimize cure and process robustness. Data shows that once large air pockets are trapped between plies of UD prepregs, they are unlikely to be removed, even by extended RT vacuum holds, due to high resin viscosity and lack of access to dry fiber tows. Poor vacuum quality increased bubble expansion rates and times due to the decrease in resin pressure. However, the final porosity in finished parts was similar for the 80% and 100% cases, indicating that the final porosity largely depends on the amount of water that can dissolve back into the matrix during Stage III. Examining the void content-time curve of each experiment, we found that the final porosity was always less than the corresponding porosity at the end of Stage I, which indicates that water that diffuses into voids during bubble expansion (Stage II) can always be absorbed by the matrix during Stage III due to the increased solubility. It seems that the inter-ply porosity is determined by the amount of entrapped air which is not removed before full impregnation. This assumption is supported by the results of humidity-conditioned tests. Increases in moisture content of prepregs result in severe porosity, as moisture not only introduces more initial trapped air during lay-up, but also inhibits air evacuation due to increased tack. The results reported provide insights into air removal mechanisms during prepreg consolidation.

5 ACKNOWLEDGEMENT

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REFRENCES

[1] Helmus R, Kratz J, Potter K, Hubert P, Hinterho lzl R. An experimental technique to characterize interply void formation in unidirectional prepregs. J Compos Mater

2016:21998316650273. doi:10.1177/0021998316650273.

[2] Roychowdhury S, Gillespie JW, Advani SG, Gillespie JW, Advani SG. Volatile-Induced Void Formation in Amorphous Thermoplastic Polymeric Materials: I. Modeling and Parametric Studies. J Compos Mater 2001;35:340–66. doi:10.1177/002199801772662208.

[3] Hamill L, Centea T, Nutt S. Surface porosity during vacuum bag-only prepreg processing: Causes and mitigation strategies. Compos Part A Appl Sci Manuf 2015;75:1–10. doi:10.1016/j.compositesa.2015.04.009.

[4] Yamaleev N, Mohan R. Effect of the phase transition on intra-tow flow behavior and void formation in liquid composite molding. Int J Multiph Flow 2006;32:1219–33. doi:10.1016/j.ijmultiphaseflow.2006.05.011.

[5] Farhang L, Fernlund G. Void and porosity characterization of uncured and partially cured prepregs. J Compos Mater 2015:0021998315583924-. doi:10.1177/0021998315583924.

[6] Centea T, Hubert P. Out-of-autoclave prepreg consolidation under deficient pressure conditions. J Compos Mater 2013:0021998313494101-. doi:10.1177/0021998313494101.

[7] Kay J, Fernlund G. Process conditions and voids in out of autoclave prepregs. Proc SAMPE Tech Conf Balt Maryl May 21-24 2012:12.

[8] Grunenfelder LK, Nutt SR. Void formation in composite prepregs - Effect of dissolved moisture. Compos Sci Technol 2010;70:2304–9. doi:10.1016/j.compscitech.2010.09.009.

[9] Hu W, Grunenfelder LK, Centea T, Nutt S. In-situ monitoring of void evolution in unidirectional prepreg. J Compos Mater 2017.(Under Review)

[10] Wells J, Kay J, Poursartip A, Lane M, Fernlund G. Surface and Bulk Porosity in Out-of-Autoclave Prepregs. Proc Int Conf Compos Mater (ICCM-20) Copenhagen, Denmark July 19-24 2015:12.

[11] Cender TA, Gangloff Jr JJ, Simacek P, Advani SG. Void reduction during out-of-autoclave thermoset prepreg composite processing. Soc Adv Mater Process Eng Tech Conf 2014.

[12] Rigard C. Process Selection and Optimization for Out of Autoclave Prepreg Structures. Proc CAMX 2015 2015:12.