In situ monitoring and analysis of void evolution in unidirectional prepreg

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Abstract: In the layup of prepreg laminates, air is inevitably entrapped between adjacent prepreg plies, yet the removal of this inter- ply air under vacuum bag cure conditions is not well understood. In this study, an in situ visualization technique was developed to dynamically observe inter-ply air removal during the cure of an out-of-autoclave prepreg. The technique was used to investigate mechanisms of air removal and void evolution in unidirectional prepreg. Prepreg impregnation was also tracked by inspection of laminate cross-sections prepared at different times during the cure cycle. From these data, a three-stage air removal mechanism was documented based on the relationships between void content, resin properties, and tow impregnation as functions of time. Furthermore, a positive correlation was observed between the rate of evacuation and bubble elongation. Though discussed here in the specific context of voids in unidirectional laminates, the in situ observation technique developed for this work has broad potential to enhance understanding of processing phenomena associated with out-of-autoclave prepregs.

Key words: Prepreg, porosity/voids, out-of-autoclave processing

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1. INTRODUCTION

Out-of-autoclave (OoA) processing of vacuum bag only (VBO) prepregs presents an appealing alternative to traditional autoclave manufacturing technology for high performance composites. Potential benefits include greater energy efficiency and lower capital and operating costs.[1] However, because the pressure supplied during cure is often at most 0.1 MPa (1 atm), composite parts produced by OoA prepreg processing are susceptible to voids, which adversely affect mechanical properties.[2–4]

VBO prepregs are partially impregnated by design, featuring dry fiber pathways at ply mid-planes. The dry fiber pathways, termed engineered vacuum channels (EVaCs), enhance air transport and removal during early stages of VBO cure (Fig. 1), before being saturated by surrounding resin at high temperature. Voids in composites are generally classified into two categories according to the source of the voids: flow-induced and gas-induced.[5] Flow-induced voids, also known as tow voids, are located within the dry fiber bundles.[6–8] EVaCs constitute an initial network of porosity in the prepreg. In the absence of ideal material and processing conditions, resin infiltration into the tows can cease before full saturation. Thus, the initial porosity can sometimes lead to residual flow-induced voids. Flow-induced voids have been observed in cured laminates if resin content or resin pressure was insufficient,[9] or if the resin viscosity profile did not allow sufficient flow prior to gelation.[10]

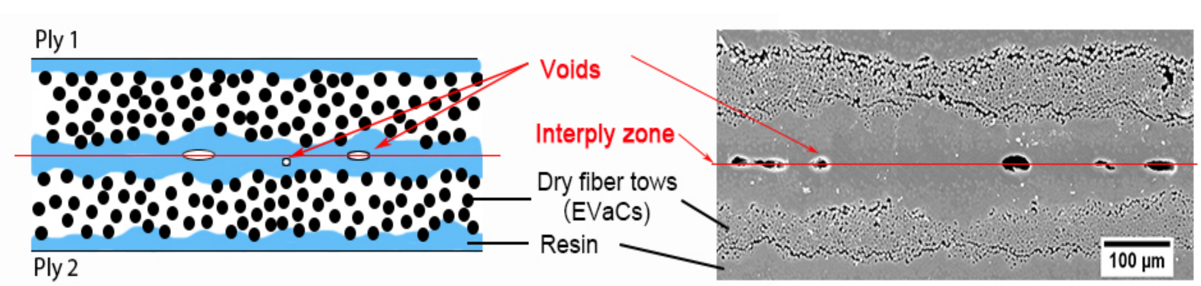

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Fig. 1. Micrograph and schematic of inter-ply voids.

The focus of the current study is gas-induced voids, which are formed primarily due to entrapped air during layup or volatiles evolving during cure. Gas-induced voids are morphologically different from flow-induced voids, being located, in cured parts, primarily within the resin-rich regions around tows and between prepreg plies.[5] Studies have indicated that initial inter-ply air entrapment correlates with the surface topology and tack of prepregs.[11] During cure, entrapped gas bubbles will expand or collapse if the gas pressure within the void is greater or less than the surrounding resin pressure, respectively. With access to an EVaC (or other flow channel), entrapped gasses can escape the laminate through edge breathing mechanisms. However, if access to an air removal pathway is occluded, or if the air is not fully removed prior to gelation of the resin, voids will remain as defects in the finished part. For example, gases located in inter-ply regions may not reach EVaCs, and must undergo a more complex removal process. For these reasons, producing void-free parts requires a thorough understanding of gas-induced void formation and evolution.

The scarcity of experimental techniques to effectively characterize inter-ply entrapped air has led to limited insight into the evolution and transport of gas-induced voids during processing. Ultrasound is commonly used in the composites industry to detect defects in laminates.[12] While ultrasound can be conveniently automated and is non-destructive, the resolution of ultrasound is relatively low (~hundreds of μm). An alternative NDI technique, X-ray computed micro-tomography (or micro-CT) generates three dimensional images at higher resolution, on the order of several microns.[13] However, the drawbacks of micro-CT include long scan and processing times, relatively small sample sizes, and high cost. Observation of laminate cross-sections at different time intervals during VBO processing with light microscopy has been used to examine void evolution.[14] While this approach provides valuable insights into the microstructure of the laminate at specific points within the processing cycle, the soft nature of the partially cured polymer matrix can introduce artifacts by alteration of void morphology during sample preparation or removal from curing conditions. Most importantly, none of the methods described above provide *in situ* monitoring of inter-ply void evolution during VBO cure.

Transparent glass tooling surfaces have been employed in previous studies to observe air entrapment, distribution, and surface porosity *in situ.*[15–18] For example, Cender et al.[19] used a transparent acrylic table and a CCD camera to monitor the dual scale flow of resin within fabric prepregs. Gangloff et al.[20] used the same method to investigate resin flow and bubble motion in fabric prepregs under constant pressure and temperature conditions. Finally, Hamill et al.[11] also investigated the formation and evacuation of surface porosity of prepregs using a glass plate and camera. However, most previous studies focused on void evolution and flow in woven fabric prepregs, and did not replicate the curing environment of VBO processing, i.e., a vacuum bagged laminate cured in an air circulating oven. In this study, laminates were laid up and bagged on the interior side of a transparent oven window, thereby re-creating a standard VBO cure environment while at the same time allowing direct observation *in situ*. Additionally, a perforated resin film with controlled pore size and distribution was placed between the glass window and the first prepreg ply, intentionally introducing entrapped air into the layup. With these modifications, air evacuation and entrapment between unidirectional (UD) prepreg plies was investigated throughout the VBO cure process.

In this work, we perform a case study to highlight the utility of the *in situ* method. Specifically, the study was undertaken to increase understanding of inter-ply void formation and removal mechanisms in unidirectional VBO prepregs. A thorough understanding of gas-induced void formation and evolution during VBO processing is needed to guide the design of more efficient and robust prepreg formats, and to optimize cure processes to minimize porosity in finished parts. The *in situ* observation technique developed for this work yielded new insights into bubble migration, expansion, and removal during VBO cure. Resin impregnation during cure was also tracked, and links between void evolution phenomena, resin properties, and tow impregnation were revealed. Finally, experimental observations were used to establish a three-stage void removal mechanism for unidirectional VBO prepregs.

2. EXPERIMENTAL DETAILS

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 manufacturer's recommended cure cycle for the material at the time of the research specified cure at 93 ºC for 12 hours or 121 ºC for 3 hours. The lower temperature cycle was used in this study, with a ramp rate of 2 °C/min. Micrographs of the initial condition of unconsolidated prepreg were obtained using an SEM (JEOL JSM-6610). Prepregs were laid up and accrued out-time at room temperature for six months, allowing the resin to vitrify prior to sectioning. Both cross-sectional views and surface images of samples without conductive coating were captured using a back-scatter electron detector at an accelerating voltage of 20 kV.

Neat resin films were also utilized in this work. The films (areal weight of 91.3±4.7 g/m^2, thickness of 49.5±6.5 µm) were composed of the same resin as the prepreg. To create perforated

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resin films, holes were punched manually into the film using a coring tool with a diameter of 0.25 mm.

2.2. In situ observation of VBO cure

The primary sources of gas-induced voids in composites are air entrapped during the layup and evolved gases (notably moisture). Entrapped air bubbles are typically located in the resin-rich regions between prepreg plies, as shown in Fig. 1. To accurately reproduce the conditions for air removal and gas entrapment described in the introduction, and to allow monitoring of multiple bubbles in the same field of view, an analogous configuration in which air bubbles are surrounded by resin must be created. This type of artificial inter-ply zone can be produced by incorporating a perforated neat resin film into the layup.

In this study, a perforated resin film containing holes of controlled size and distribution was laid up against a glass oven window, and prepreg plies were subsequently laid onto the film. In this way, air bubbles were intentionally introduced into the lay-up and surrounded by resin everywhere except at the bubble-glass interface. In general, the configuration used approximated the conditions surrounding an internal void located at the mid-plane of a stack of prepreg plies. Furthermore, incorporating voids of known diameter via a perforated film enabled quantitative measurements of porosity with controlled initial characteristics, as well as visualization of trapped air migration. In a parallel set of experiments, the potential effects of the bubble-glass interface on bubble behavior were investigtated by creating bubbles fully enclosed in resin. This condition was achieved by inserting an additional, non- perforated film between the glass tool plate and the perforated film.

Results from these experiments indicated bubble behavior was the same, regardless of which configuration was used.

To produce test panels, prepreg plies were cut to 127 mm × 127 mm. To visualize voids, air pockets were introduced via perforated resin films containing holes with a diameter of 0.25 mm and spacing between holes of 2 mm (Figs. 2(b) and (c)). These resin films were 38 ×38 mm. Each test was repeated 3 times. The moisture content of the as-received prepregs was 0.10 ± 0.01%.

Fig. 2(a) shows the experimental setup. Laminates were laid up and bagged vertically on the interior side of a transparent window in a programmable air-circulating oven (Thermal Products Solutions Blue M). The glass window was treated with liquid release agent (FreKote 770-NC) prior to layup. A layer of perforated resin film was positioned against the glass window. Four layers of prepreg were stacked on top of the perforated resin film, and the assembly was then vacuum bagged using standard consumables. The consumable arrangement consisted of a layer of non-perforated Teflon release film on the bag-side laminate surface, edge-breathing dams made of vacuum sealant tape wrapped with fiberglass boat cloth, and finally one layer of breather cloth followed by a vacuum bag. The placement of the resin film and prepregs was controlled to achieve similar initial void contents (12% to 15%) for each test. Six thermocouples were used to monitor temperature. Five thermocouples were placed against the Teflon release film, one at each of the four corners of the laminate and the last at the center, while the sixth thermocouple was located against the interior surface of the glass window. Time-lapse videos were recorded from outside the oven, using a portable microscope (Dino-Lite Premier2 Digital Microscope). Video was collected throughout the cure cycle at a magnification of 20 ×.

Gravity effects can be neglected in this study for the following reasons. First, the Bond number, *Bo*, was estimated to be 0.02. Bond number is a dimensionless number that characterizes the ratio of gravitational force to surface tension.

$$Bo = \frac{\Delta \rho g L^2}{\sigma} \quad (1)$$

where $\Delta\rho$ is density difference between epoxy resin (=1310 kg/m3)[21] and air bubbles (=1.2 kg/m3),[22] g is gravitational acceleration (=9.8m/s2), L is the characteristic length (the bubble diameter ~0.25mm was used), and σ is surface tension (= 0.035 N/m).[8] A value of Bo <<1 implies a weak dependence on gravitational force. Secondly, bubble movements during cure were observed to be horizontal (and along the fiber direction) in all tests. Accordingly, resin movement was also likely to occur in the horizontal direction. Furthermore, no laminate thickness gradients were observed in the vertical direction after cure, indicating that gravity-driven bulk resin flow did not occur.

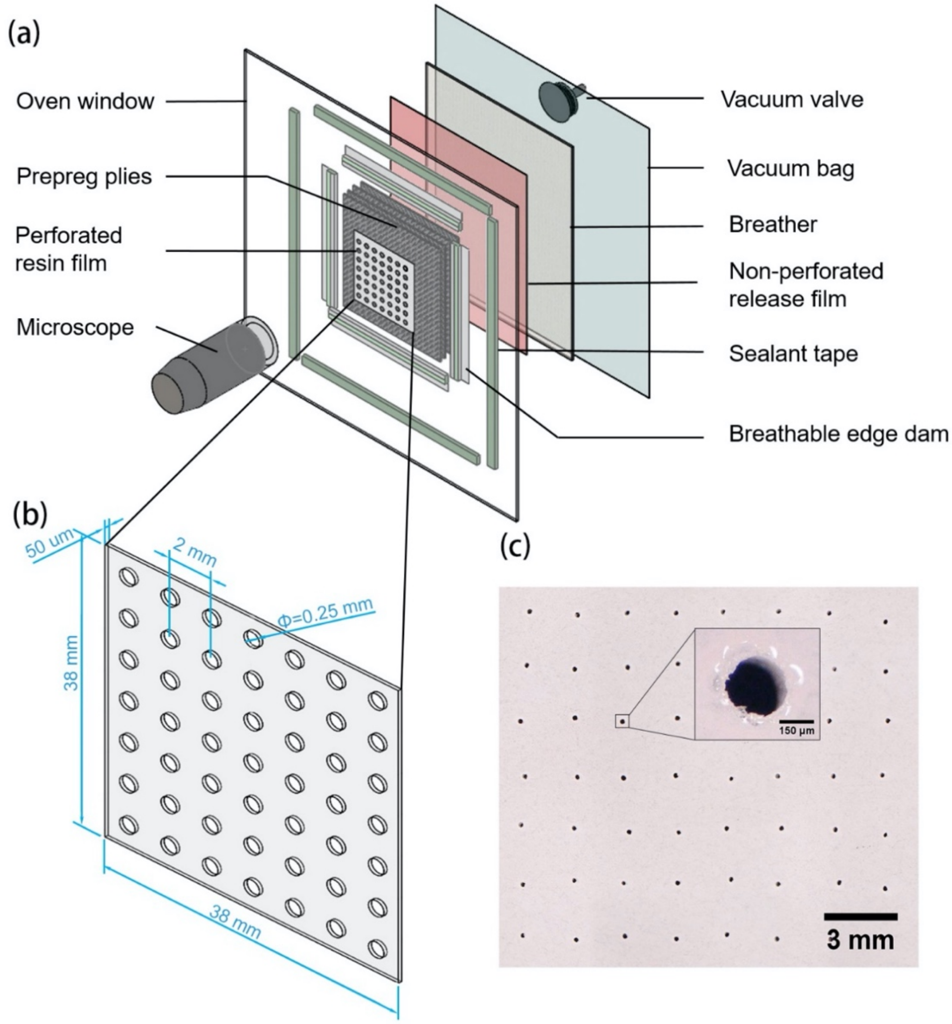

Fig. 2. (a) Schematic of *in situ* observation set-up, (b) Dimensions of perforated resin film, (c) Micrograph of perforated resin film.

2.3. Void content

Void content as a function of time was calculated for each test panel using image analysis software (ImageJ). Ten frames, recorded at different points throughout the cure cycle, were chosen from the time-lapse videos, and the voids present in the image were manually selected. The images were then converted to binary, with voids represented in black, and the remaining area in white. A percent value for porosity was determined by dividing the number of black pixels by the total number of pixels in the image.

2.4. Partial cure sample preparation

To better understand fiber bed compaction, resin impregnation, and the relationship of both factors to void evolution, laminates were prepared by interrupting the prepreg cure cycle at different points, as shown in Fig. 3. Each laminate prepared by partial cure processing consisted of 8 plies of prepreg 127 mm × 127mm with a unidirectional $[0]_8$ layup. To preserve the morphology of the laminates at each point of interest, panels were removed from the oven at the desired point in the cure cycle while maintaining vacuum and rapidly quenched with liquid nitrogen to prevent further resin flow. The skin temperature of each laminate was tracked using thermocouples.

Partially cured laminates were then placed in an ammonia environment at room temperature for 10 days. Ammonia vapor reacts with epoxy at a low temperature, acting as a curing agent to achieve a hard and stiff structure [23]. This technique enables the microstructure of samples to be assessed at various points in the cure cycle without altering the internal structure. Following ammonia cure, samples were sectioned at the center of each panel, mounted and polished using a series of graded abrasive papers. Polished sections were imaged using a stereo microscope (Keyence VH-Z100R).

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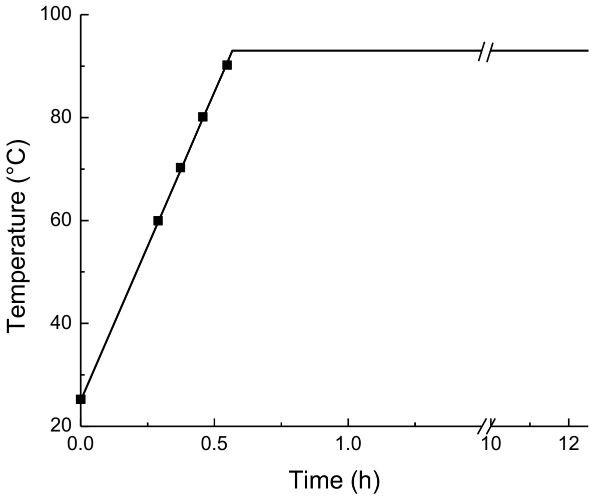

Fig. 3. Cure cycle with partial cure sampling points marked with black squares.

2.5. Water solubility in epoxy resin

To support hypothesized explanations of void growth and collapse during the cure process, the effect of degree of cure on the water solubility of epoxy resin was investigated. Neat resin samples were prepared to different degree of cure values following a cure kinetics model previously developed for the same resin system.[24] Next, samples were humidity-conditioned under 99% (± 1%) relative humidity for 24 hours at room temperature (21 ± 2°C). 99% relative humidity was achieved in a sealed container with saturated K_2SO_4 solution. Modulated-DSC measurements (TA Instruments Q2000) were performed to verify the degree of cure of each sample after humidity conditioning. Moisture content of each sample was measured by Fischer titration using a coulometric titrator (Mettler Toledo C20 with D0308 drying oven).

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3. RESULTS AND DISCUSSION

3.1. Air removal in unidirectional prepregs during VBO cure

The actual means of inter-ply void suppression are not well understood, although three possible removal mechanisms are generally considered possible. The first is gas transport through the inter-ply zone (Fig. 1). Because prepreg is not flat, gaps are created between adjacent plies during the layup process. These gaps, a consequence of the inherent surface roughness of the prepreg, provide a potential pathway for in-plane air transport, which can occur as continuum gas flow (if the gap is empty) or as bubble migration through viscous resin (if the gap is resin-filled). The second potential evacuation route is along the engineered vacuum channels at the center of each prepreg ply. Gas initially entrapped within the pores that form the EVaCs can easily flow to the laminate boundaries. Conversely, gas entrapped between plies must migrate short distances in the through-thickness direction to reach the dry fiber tows. Once the gas reaches an EVaC, it can be rapidly removed from the laminate, in-plane, via edge breathing. The third evacuation route is out-of-plane transport through the thickness of the laminate. However, for unidirectional prepregs, literature has shown that the through-thickness permeability is up to three orders of magnitude less than the in-plane permeability, and can essentially preclude through-thickness air transport.[25,26] In this study, most bubble transport occurred in the in-plane direction, in agreement with prior observations, although concurrent through-thickness transport is possible as well, as discussed later.

To observe inter-ply air evacuation and entrapment during the VBO cure process, a custom-built experimental setup was used, incorporating *in situ* visual observation. Bubble transport and the progression of gas evacuation during out-of-autoclave cure of unidirectional prepregs were monitored using the *in situ* observation technique. Select still images from the recorded video of a representative experiment are presented in Fig. 4. During layup, air is initially trapped both in the intentionally created (artificial) pores (Fig. 4(a) circled in red) as well as between the perforated resin film and the first ply of prepreg (white regions in Fig. 4(a)). The naturally formed air pockets are randomly located around the artificial pores. When vacuum is applied at the beginning of the cure cycle, the visible area of natural pores (and therefore the area of the white regions) gradually shrink. The artificial pores also slightly decrease in size, but the shape and position of the pores remain stable.

At low temperatures, inter-ply air evacuation in unidirectional prepregs is a slow process, because the resin viscosity is high and there is no direct route for air flow to a breathing edge. Close inspection of natural pores formed between the prepreg and the perforated resin film indicates that small regions of trapped air are more likely to be evacuated as the cure cycle progresses (Fig. 4(a-c)). The gas transport process, not surprisingly, occurs more easily at elevated temperature as a result of reduced resin viscosity. The images in Figs. 4(b-d) show that as the temperature increases, bubble size continues to decrease and even artificial air bubbles migrate at ~ 70°C (observed from the video). All movement of artificial air bubbles studied here occurred in-plane, along the fiber direction, and toward the laminate edges. The driving force for bubble movement is assumed to be the pressure differential supplied by the vacuum source. Throughout the cure cycle, some air bubbles were observed to coalesce, forming one larger bubble. At 80°C, a point roughly 30 minutes into the cure cycle, most of the naturally formed air pockets were no longer visible, and the overall void content reached a minimum value (Fig. 4(c)).

Following this point in the cure cycle, an unanticipated phenomenon was observed. Beyond 80°C, as the temperature ramp continued and the dwell at 93°C began, existing air bubbles increased in

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size. Additionally, some small bubbles grew from the residual naturally entrapped air. This bubble expansion continued for approximately one hour. After the expansion, the morphology of the bubbles also changed, with initially round bubbles elongating to different extents. Bubble expansion, in general, indicates that the internal gas pressure within the bubble exceeds the surrounding resin pressure and the resistance to growth created by surface tension. Following the growth phase, bubbles gradually shrank until resin gelation occurred. Prior to completion of the cure cycle, most bubbles were removed, resulting in a nearly void-free laminate.

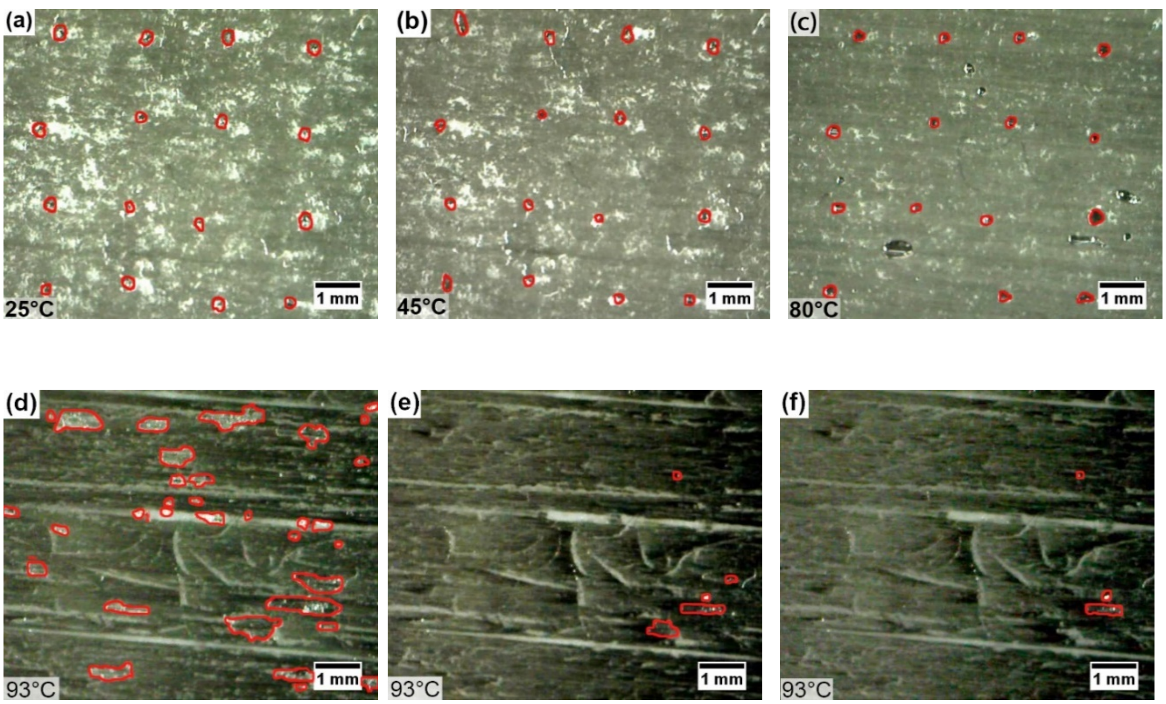

Fig. 4. Time-lapse images of void evolution in unidirectional prepregs during cure. Images were taken at a) initial state, before vacuum applied, b) 20min, c) 36min, d) 86min, e) 8h and f) 12h. The video file can be viewed on-line (URL: http://composites.usc.edu/projects/insitu.htm)

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3.2. Tow impregnation during cure

While *in situ* monitoring provides insights into inter-ply air removal, tow impregnation is more readily apparent in cross sections of partially cured prepregs. For this reason, the morphology and impregnation of EVaCs were investigated via partial processing of a sequence of laminates. Representative micrographs from different stages of the cure cycle are presented in Fig. 5. The as-laid-up state of a stack of unidirectional prepreg plies, prior to compaction and cure, is shown in Fig. 5(a). Within each ply, dry fiber tow areas are surrounded by resin-rich regions on both sides. Large, elongated gaps are present between plies as a result of air entrapped during laminate preparation. When heat and pressure are applied, the progression of fiber bed compaction and resin infiltration begins. At 60°C (Fig. 5(b)), the fiber tows remain partially dry, while the fiber bed is compacted and the inter-ply voids have reduced in size and become round. At 80°C when bubble expansion is observed, dry fiber tows are almost fully impregnated. For this material system, full impregnation is complete between 80°C (Fig. 5(c)) and 90°C (Fig. 5(d)).

Resin infiltration (tow impregnation) has a complex effect on entrapped air removal. Resin flow can enhance air removal, as air bubbles trapped in resin-rich regions are able to migrate with the resin flow front to dry fiber tows, from which in-plane evacuation can occur. However, resin infiltrating the dry fiber tows can also impede air removal by occluding the air evacuation pathways. Prepreg impregnation also affects local pressure and moisture concentration, both of which evolve with time, and can affect void nucleation and/or growth in a complex manner.

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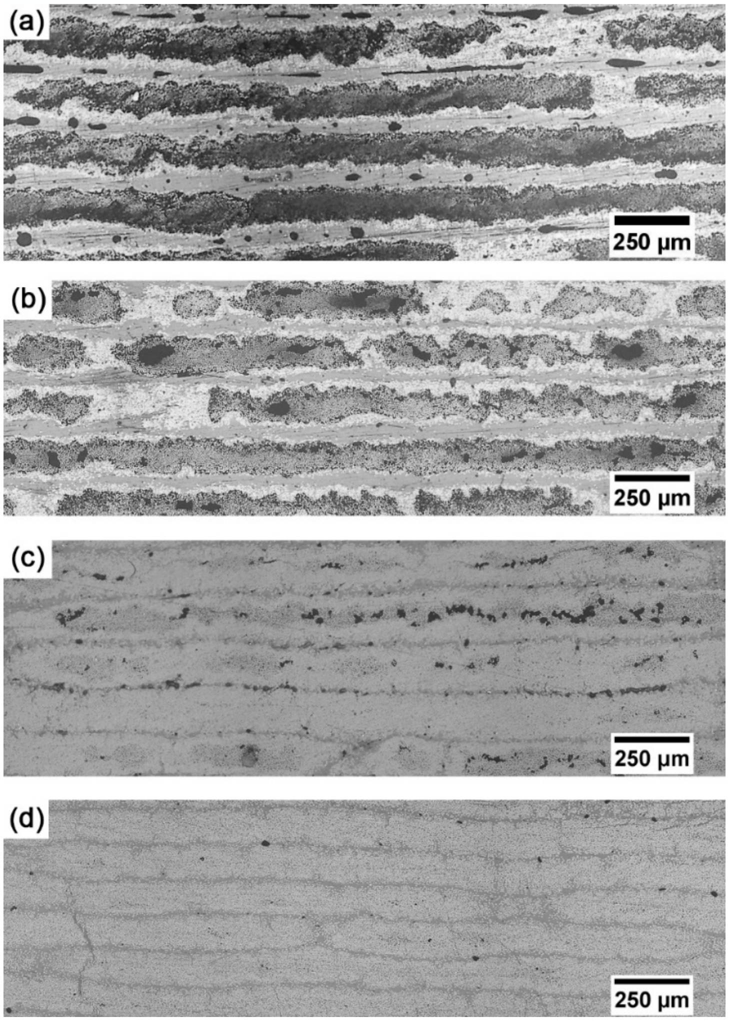

Fig. 5. Micrographs of tow impregnation in UD prepreg laminates during cure process. Laminates were (a) at initial state after lay-up, cured to (b) 60°C, (c) 80°C and (d) 90°C.

3.3. Void removal mechanisms

The relationship between resin properties, tow impregnation and bubble behavior was investigated to gain an improved understanding of air removal mechanisms in UD prepregs. Resin viscosity and degree of cure were calculated using published viscosity and cure kinetics models[24], while degree of impregnation was quantified by measuring the ratio of visible dry fiber tow area to total ply area in polished cross-sections of partially processed samples. Void content was determined by analyzing time-lapse videos from *in situ* cure monitoring experiments (with three replicates).

The results of these experiments are summarized in Fig. 6, with all variables tracked as a function of time, and correlated to temperature (Fig. 6(a)). Fig. 6(b) shows the evolution of void content during the cure cycle. Initial void content, representing both naturally trapped air and artificially introduced air bubbles, was approximately 14%. In the first 30 minutes of the cure cycle, during the temperature ramp, void content decreased from 14% to ~2.5%. At this point, near the end of the temperature ramp, void growth was repeatedly and consistently observed (without nucleation of new voids), producing a ~2% increase in porosity (Fig. 6(b)). This void growth was followed by a period of gradual decrease. The void content reached an equilibrium state after approximately 7 hours at the cure temperature of 93° C. The final void content in all test panels was less than 1%.

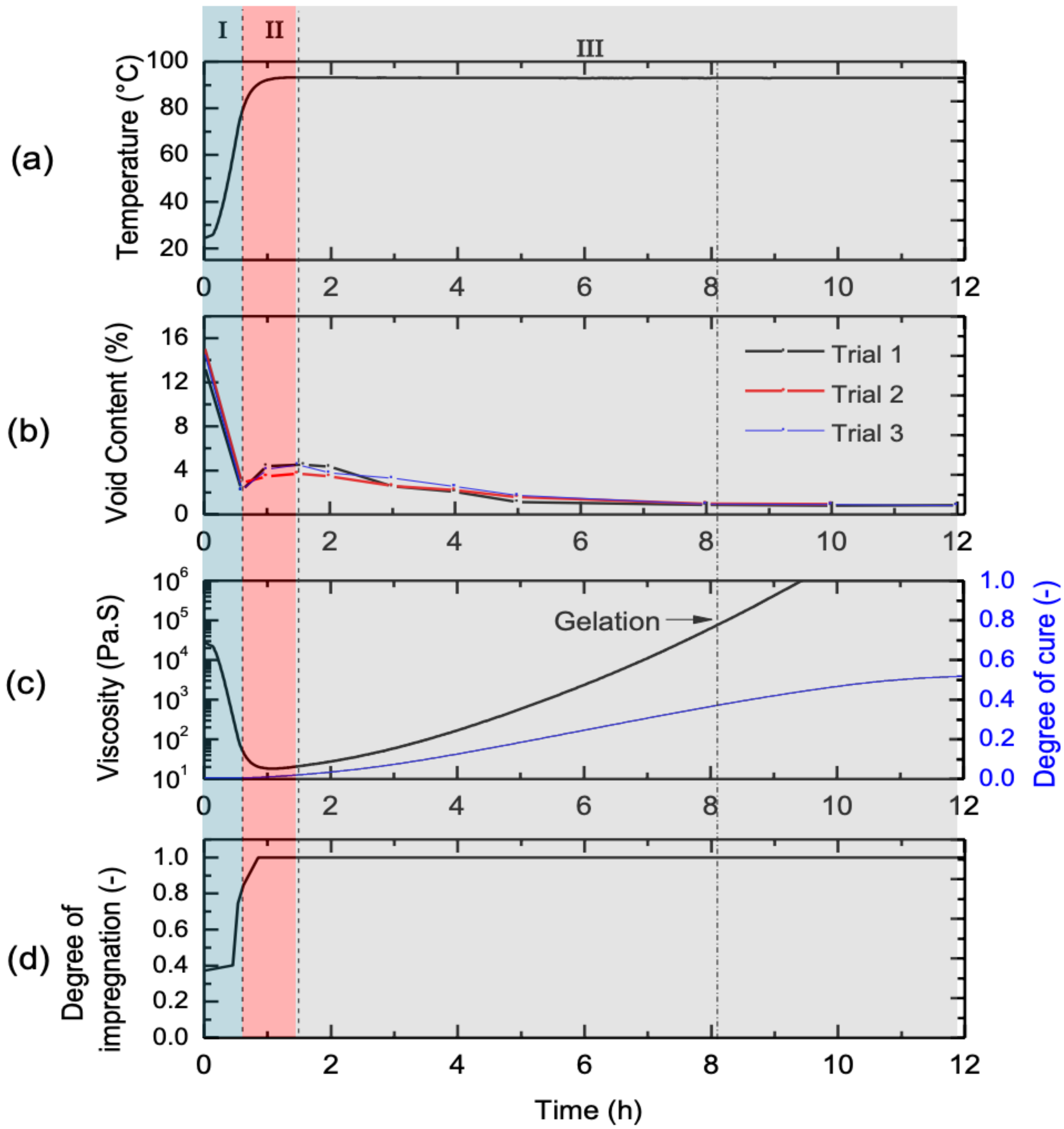

Fig. 6. The relationship between void content, resin properties, and tow impregnation. The three stages of void evolution are denoted with roman numerals.

Examining void content as a function of time, the evolution of inter-ply air bubbles in UD prepregs during cure can be divided into three stages: (I) air evacuation, (II) bubble expansion, and (III) bubble shrinkage. These stages are denoted with Roman numerals and dashed lines in Fig 6.

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During air evacuation, resin viscosity remains relatively high (Fig. 6(c)), and fiber tows are partially dry (Fig. 6(d)), facilitating air flow through the prepreg to breathing edges. Air naturally entrapped between the first ply of prepreg and the perforated resin film is gradually evacuated via either resin flow or resin-starved regions on the surface of the prepreg (see Fig. 7). The artificially induced air bubbles, in contrast, remain stationary because they are separated from the air evacuation channels by the resin-rich surface of the prepreg. As temperature increases, resin viscosity decreases, and large air bubbles begin to migrate in-plane, along the fiber direction.

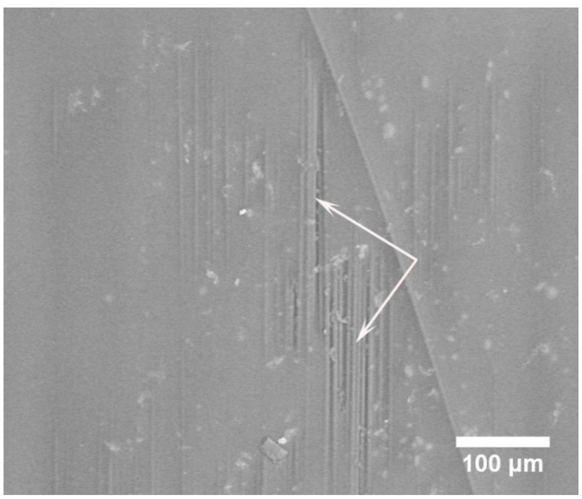

Fig. 7. Surface of unidirectional prepreg, showing resin rich and resin starved regions (arrows). Dry regions on the surface of the prepreg enable through-thickness gas transport to an air evacuation pathway.

Bubble expansion occurs during Stage II, which begins at approximately 80°C, slightly after bubble movement initiates. At this point in the cure cycle, resin viscosity is near minimum, and dry-fiber gas evacuation pathways are almost fully sealed off, as shown in Figs. 5(d) and 6(d). Full tow impregnation represents a significant change in the microstructure of the laminates, potentially affecting gradients in pressure and moisture concentration. Data in Fig. 6 show that these changes coincide with bubble expansion.

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Bubble expansion/shrinkage depends on the equilibrium between the gas pressure within the bubble P_{void}, the surrounding hydrostatic resin pressure P_{resin}, and surface tension effects γ_{LV}. If $P_{void} > P_{resin} + 2\,\gamma_{LV}/R_{void}$, a bubble will grow. During Stage II, both P_{resin} and P_{void} are likely to change due to full tow impregnation. For example, P_{void} is driven by moisture diffusion from the resin, which now occurs solely towards bubbles (versus towards the bubbles and the flow front). Moreover, diffusion is temperature-accelerated and water vapor pressure in the void increases exponentially with temperature.[27] Full tow impregnation also represents a change in pressure boundary conditions within the resin. Resin pressure is difficult to measure during prepreg cure, because the resin is viscous, and because any sensor must form intimate contact with the resin without interacting with the fibers. Preliminary efforts using a melt pressure sensor embedded within a metallic tool plate and connected to the resin using a transfer fluid indicate that the resin pressure remains between 50 – 100 kPa during processing. Studies have shown that during prepreg processing, the internal vapor pressure in a void can exceed this value.[18,27,28] The interplay between these factors is complex, but the data clearly show that bubble expansion in Stage II coincides with full impregnation (Figs. 6(b) and (d)).

During the third and final stage of void evolution, bubbles shrink, and the dry fiber tows are fully impregnated with resin. Meanwhile, the resin viscosity is increasing to a final plateau as cross-linking completes. Because the tows are fully impregnated at this point in the cure cycle, gas evacuation via EVaCs to a breathing edge is no longer possible. The decrease in void size, therefore, is attributed to changes in moisture absorption in the resin as a function of degree of cure. Fig. 8 shows moisture content as a function of degree of cure for the resin system studied here. The data indicate that water solubility increases with degree of cure, a finding consistent with past experimental studies in which the phenomenon is explained as a result of attractive forces

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between water molecules and polar groups formed during epoxy cross-linking.[29] As the degree of cure increases in the final stage of processing, water previously released into growing voids once again dissolves into the resin, resulting in the final stage of bubble shrinkage. During this stage, bubbles continued to migrate in-plane towards the edges. While this void migration could lead to a decrease in void content, bubble movement did not affect void size measurements.

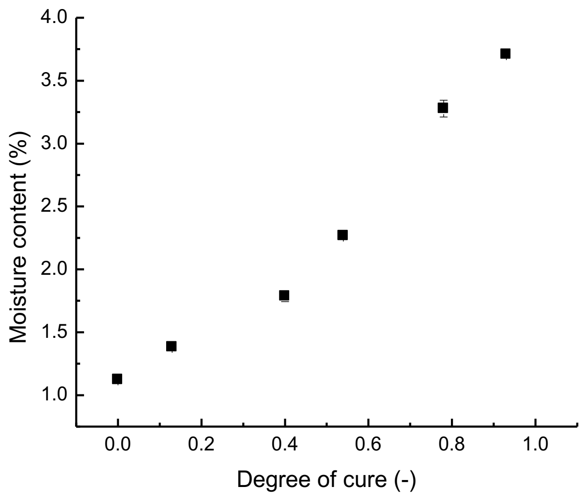

Fig. 8. Water solubility in epoxy (Cytec 5320-1) resin as a function of degree of cure.

3.4. Bubble mobility

During Stage II, air bubbles in the resin were observed migrating in-plane, along the fiber direction, towards the laminate edge. To clarify the physics of bubble migration, ten bubbles were selected and tracked for quantitative analysis. These ten bubbles were independent but in close proximity, ensuring that the local environments (i.e., local pressure/pressure gradient, resin flow, etc.) were comparable. The morphology of each bubble was measured using a best-fit ellipse (ImageJ), providing the bubble area, length (l, where $l=2a$ and a is the half-major axis length) and width (w, where $w=2b$ and b is the half-minor axis). The average bubble velocity was determined by the displacement of the center of each bubble over time. Bubble velocity was calculated over three separate five-minute time periods during Stage II, labeled A, B and C, as shown in Fig. 9(a).

The relationship between bubble velocity and resin viscosity, bubble size, and bubble aspect ratio was analyzed. No obvious correlations between bubble velocity, resin viscosity, and bubble size were observed. However, the bubble velocity showed a strong correlation to the aspect ratio of the best-fit ellipse. Indeed, Fig. 9(b) shows that bubbles with larger aspect ratios exhibited higher migration rates, as well as that the effects of bubble aspect ratio became less pronounced as cure progressed. During bubble migration, the bubble tended to grow larger, and became more elongated. Elongation can increase the pressure gradient across the bubble via buoyancy, driving faster bubble migration. Conversely, surface tension also increases as the bubble surface area increases, impeding bubble migration.[20] Because of these competing factors, bubble velocity increases only when the increasing buoyancy force overcomes the increasing surface tension.

Previously, authors have used the Hele-Shaw model to estimate bubble motion in a viscous fluid.[16,20,30] In a Hele-Shaw cell, the moving bubble is assumed to travel in a Newtonian fluid between two parallel plates with an infinitely small gap. For small bubbles in a Hele-Shaw cell, the ratio of average bubble velocity to average resin velocity (*U/V*) depends on the aspect ratio (*a/b*) of the bubble.[20]

$$\frac{U}{V} = 1 + \frac{a}{b} \qquad (2)$$

The average fluid velocity V is given by[20]

$$V = \frac{-h^2}{12\mu}\frac{dP}{dx} \qquad (3)$$

where μ is resin viscosity, h is the channel height and dP/dx is the pressure gradient in the resin.

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For the case studies in this article, the physics of bubble transport are more complex than those captured by this model, although the equations above provide useful insights into potential factors affecting bubble migration. In Hele-Shaw flow, the channel height is assumed to be constant. For our case, this channel height is related to the distance between the impermeable tool plate and the fiber bed, or the instantaneous thickness of the perforated resin film. During experiments, this channel height decreased as resin infiltrated the dry fiber tows, thereby reducing bubble velocity. Moreover, as the degree of impregnation increased, the pressure gradient in the resin also decreased, as the resin pressure equilibrated within the fiber bed. Thus, the effect of bubble aspect ratio on bubble velocity diminished as time progressed. After reaching minimum viscosity, the cure reaction led to increasing non-Newtonian behavior within the resin. These deviations from the expected flow regime are expected to be minor during Stage II, since changes in measured viscosity were small.

The observations described above emphasize that inter-ply bubble migration during prepreg cure can be affected by multiple factors, including bubble morphology, resin viscosity, and pressure gradient. These factors are inter-related, and exert mutual influence in a complex manner. Furthermore, these factors are generally difficult to determine due to stochastic variability in materials and processes (for example, this study included observations that the migration of some bubbles can be significantly impeded by a single off-axis fiber lying on the tow perimeter). Overall, the results demonstrate that bubble flow models previously used by others provide a useful baseline from which to explain experimental observations, but may not fully capture the complexity of the physics involved and the mechanisms that govern defect formation during cure.

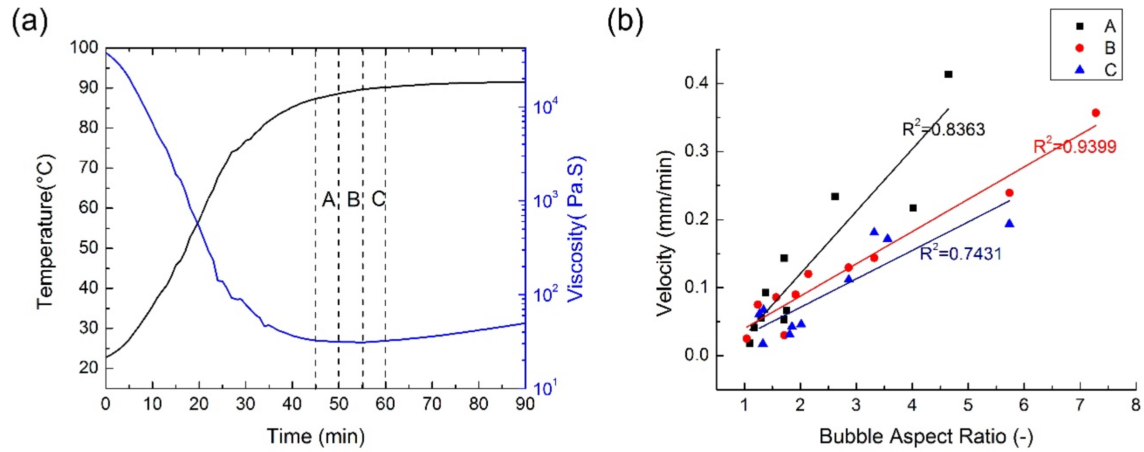

Fig. 9. (a) Temperature and resin viscosity profile versus time, indicating three time intervals used to analyze bubble mobility, (b) correlation of bubble velocity with aspect ratio.

4. CONCLUSIONS

In this work, an *in situ* observation technique is described and used to monitor air removal during the cure of out-of-autoclave carbon fiber/epoxy prepregs. The technique accurately captures the phenomena that occur between the plies within a laminate during VBO cure. Simulating the conditions of air removal during VBO cure is accomplished by incorporating a perforated resin film into the lay-up to enable not only visualization of void evolution throughout cure, but also quantitative measurements. Additionally, cure is carried out in an air circulating oven with a standard vacuum bag configuration, reproducing a realistic cure environment. The method can be utilized to ascertain mechanisms of inter-ply air removal (as in this work), as well as in parametric studies to address key aspects of void formation and evolution in out-of-autoclave prepregs.

The method demonstrated here was applied to the analysis of inter-ply void evolution during cure in unidirectional prepregs. Data obtained from *in situ* measurements were combined with tow impregnation data obtained through cross-sectional analysis of partially processed laminates. A comprehensive explanation of inter-ply air removal was established by inspecting void evolution and prepreg morphology in both in-plane and cross-sectional views. The explanation included a three-stage air removal mechanism based on void behavior, resin properties, and tow impregnation which, to our knowledge, has not been previously observed or reported in the literature. The resulting insights support and are consistent with our current understanding of prepreg consolidation processes, and thus confirm that the method proposed here captures the key phenomena governing inter-ply void evolution. The observations exhibit some of the trends predicted by simple models, but highlight the complexity of the physics governing bubble mobility and migration. In principle, this understanding can be used to optimize the cure of prepregs under VBO conditions and modify cure cycles to facilitate void reduction. Porosity is the major defect type arising during VBO processing, in the absence of elevated consolidation pressure. Consequently, the development of science-based void reduction strategies can support the ongoing shift from autoclave processing towards lower-cost, higher-efficiency methods.

Insights gained through *in situ* observation of process phenomena contribute to an improved understanding of the mechanisms at play in composite manufacturing. For decades, composite processing has been improved incrementally through a predominantly trial-and-error approach. The development of *in situ* process diagnostics and the ability to monitor phenomena in real time can provide valuable insights and understanding that will lead to improvements in both process efficiency and part quality. The technique presented here was applied to void evolution in UD prepreg to demonstrate proof of concept. The method, however, is widely applicable, and is presently being used to assess void formation mechanisms with woven fiber architectures, novel prepreg formats, and non-traditional cure cycles.

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