Effect of prepreg format on defect control in out-of-autoclave processing

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Abstract: Prepreg format plays a key role in part quality for composites produced using vacuum bag only (VBO) techniques. To date, however, VBO prepregs have been produced by modifying existing autoclave formats. In this work, we introduce USCpreg, a prepreg format designed specifically for out-of-autoclave cure, featuring through-thickness permeability. We describe the fabrication and analysis of laminates processed with USCpreg, as well as laminates fabricated from traditional VBO prepreg formats. The through-thickness pathways for air transport in USCpreg result in near-zero internal porosity and defect-free surfaces in parts cured under VBO conditions, even under challenging processing conditions. Results highlight the fact that surface and internal porosity depend on prepreg format, and that through-thickness permeability is critical to achieving high quality parts in non-ideal manufacturing scenarios.

Key words: A. Prepreg; A. Polymer-matrix composites; B. Porosity; E. Out of autoclave processing.

1. INTRODUCTION

Out-of-autoclave processing of composites promises lower cost and increased throughput relative to autoclave manufacturing. The appeal of out-of-autoclave (OoA) processing over

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autoclave manufacturing stems from the reduced capital investment, elimination of the need for costly nitrogen gas, greater energy efficiency, and removal of size constraints. Vacuum bag only (VBO) processing of prepregs, in particular, is an appealing out-of-autoclave technique for high-performance applications. Advantages of prepreg over other OoA methods such as infusion or resin transfer molding include the capacity to use higher-performance and higher viscosity resins and incorporate modifiers, as well as the ability to precisely control fiber alignment and fiber volume fraction [1], [2] and [3]. Additionally, VBO processing of prepregs utilizes many of the same protocols and consumables as autoclave manufacturing, but enables the use of lower pressures and temperatures during cure, allowing for the use of inexpensive tooling and flexible curing environments [4] and [5].

VBO processing of prepregs has been driven by developments in prepreg technology that have evolved from autoclave prepreg formats. For autoclave processing, prepregs historically featured full resin impregnation of the fiber bed (or as much as possible) to enable near-net-shape lay-up of parts. Full impregnation was achieved by solution coating, a technique in which dry fabric is pulled through a bath of resin dissolved in a solvent. Following solution coating, solvent is removed from the prepreg in a sequence of drying ovens. Solvent dip prepregging, however, results in residual solvent in the resin, which can evolve gases during cure and lead to voids. For this reason, the bulk of commercial prepregging today has transitioned to hot-melt methods [2] and [5]. Hot-melt processing eliminates the need for solvent and solvent removal, as well as the environmental and health concerns associated with solvent dip methods. In this technique, resin is heated to reduce viscosity, after which thin films are transferred onto backing papers by passing through controlled thickness dies [1] and [2]. These films are then applied to both sides of dry fabric and compacted through rollers to impregnate the fiber, forming prepreg. Achieving full resin impregnation using hot-melt prepregging is difficult. However, reduced impregnation of the fiber bed in the initial condition of the prepreg actually leads to reduced void content in cured laminates. VBO prepregs with reduced impregnation can be used to produce parts with low defect levels. However, successful cure is contingent on careful material and process control, which are often difficult to achieve in practice. As a result, VBO prepreg processing remains insufficiently robust. Over the past decade, research has focused on developing effective defect control solutions for specific material and processing challenges. In this work, we demonstrate that the redesign of the VBO prepreg format itself can eliminate the key defect formation mechanisms that exist during OoA cure, and lead to higher material and process robustness.

1.1. Background

Material format is the key factor in the production of high-quality prepreg-processed parts both in and out of the autoclave [6] and [7]. The critical importance of prepreg format was demonstrated in the 1980s through the fabrication of a "half-and-half" panel in which plies of a fully impregnated prepreg and those of a partially impregnated prepreg were used to produce a laminate by autoclave cure [7]. To produce the panel, 100 plies of the fully impregnated material were laid up, followed by 100 plies of the partially impregnated material, resulting in a 200-ply laminate. Although the two prepregs were produced from the same resin batch with the same resin content and fiber lot, the upper half of the laminate, consisting of the partially impregnated plies, was nearly void-free, while the lower half exhibited extensive porosity [7].

The concept of partial impregnation, as demonstrated in the half-and-half panel, drove the recent transition from autoclave cure to vacuum-bag processing. In VBO materials, a permeable network of unimpregnated fibers, referred to as engineered vacuum channels (EVaCs) promote in-plane gas transport. Defects are controlled by the high in-plane permeability (on the order of

10−14–10−15 m2) afforded by partial impregnation [8]. Because air transport occurs in-plane, achieving void-free parts with VBO cure requires edge breathing of laminates to remove residual or evolved gas [8], [9] and [10]. In a VBO layup, edge-breathing "dams" are positioned next to a layer of breather cloth, to which vacuum is applied. In principle, air trapped within the laminate escapes to a breathing edge and subsequently, as the laminate is heated, the resin flows to fully impregnates the initially dry areas (EVaCs), yielding a void-free part.

Thick laminate prepreg (TLP) was the name given to the partially impregnated material used to fabricate the "half-and-half" panel [7]. TLP was initially developed for autoclave cure of thick parts, and remains the classic form of VBO prepreg, featuring two-sided partial impregnation with EVaCs at the center of each ply [6] and [7]. This design has been adapted to other commercial VBO prepregs. In some cases, two-side impregnated VBO prepregs have equal amounts of resin on each side, and in other cases the resin distribution is unbalanced [9]. In practice, the same amount of resin on both sides of the prepreg tends to be preferred by manufacturers for ease of layup.

In addition to TLP, various VBO prepreg formats have been introduced, each with different specific resin distributions, depending on the supplier and application [3] and [11]. All VBO prepregs, however, feature a common characteristic - the incorporation of dry fiber regions that provide pathways for air transport. One-side tacky prepregs represent an extreme of uneven resin distribution, and these variants have been produced since the advent of hot-melt processing [3], [11], [12] and [13]. In this prepreg format, resin is applied to only one side of the fabric, while the other side remains fully dry. One-sided prepregs, though not originally developed for VBO cure, result in low internal void contents when cured out of autoclave. They do not, however, consistently yield void contents of less than 1% [3] and [14]. One of the first commercial VBO prepregs (SPRINT) was designed in a manner inverse to that of TLP, with layers of dry fabric applied to either side of a central resin film [3]. Finally, there are formats of VBO prepreg that are variably impregnated (such as ZPREG), which consist of strips of resin applied to a non-crimp fabric, with gaps between strips. Various configurations of this prepreg are available, with different strip widths and spacing. Such variably impregnated prepreg formats exhibit superior drapability compared with one-side tacky materials and conventional prepregs [5], and feature high through-thickness permeability. However, depending on the relative proportion and size of dry and resin-rich regions, achieving full impregnation during cure can be challenging.

While multiple partially impregnated prepreg formats are available for OoA cure, not all VBO prepregs perform equally, and the simple presence of dry fiber regions does not ensure low void content and high part quality [3]. In addition to the specific method of resin application, resin properties and choice of fiber architecture are key aspects of VBO manufacturing. For a given fiber bed, a proper resin chemistry must be obtained such that EVaCs remain open sufficiently long to allow air to escape from every point within the laminate, yet full saturation can be achieved prior to completion of the cure cycle [10]. Current VBO prepregs, although produced in an automated process, exhibit large spatial variations in initial degree of impregnation, which can interfere with air removal and compromise final part quality [15]. Additionally, processing composites without autoclave pressure requires greater care in prepreg storage, handling, lay-up, bagging, and cure protocols [10]. While autoclave equivalent quality can be achieved under ideal VBO processing conditions [16], at present, the key factors preventing the widespread adoption of VBO cure for high-performance applications are lack of robustness and scalability to large parts.

VBO cure involves a maximum consolidation pressure of only 1 atm, and thus laminates are more prone to a range of production problems compared to autoclave processing, in which high external applied pressures are used to consolidate prepreg plies and prevent void formation. First, the bagging technique required for VBO cure is more complex, as the need for edge breathing requires that most ply edges be precisely aligned, and connected to breather material [10]. Edge-breathing is readily achieved for flat panels, but is often impossible with complex geometries [5]. In particular, problematic geometric constraints include common features of parts, such as embedded ply drop-offs, where the ends of select plies are truncated within the layup [17], as well as corners in which pressure differentials exist and EVaCs are easily pinched off [17] and [18]. Cuts and splices, which must often be introduced to drape complex parts, further interrupt in-plane air evacuation through EVaCs [18]. In addition to part complexity, part size is a key concern for VBO manufacturing. Void contents generally increase as a function of breathe-out distance, unless extensive room temperature vacuum holds are imposed. However, such holds increase processing time, and can result in the undesirable accrual of out-time [19]. Vacuum hold time is, in fact, the rate-limiting step for VBO production of large parts, increasing production time and decreasing production rates [20]. Finally, poor vacuum often results in voids with VBO processing [21], and void types that are generally suppressed by autoclave pressures, such as surface porosity [22] and [23] and moisture-induced voids [24], are common with low-pressure VBO processing.

To be used in high-performance applications, VBO prepreg and associated processes must consistently yield high-quality, void-free parts without difficult steps or protocols [2] and [22]. Additionally, prepregs may soon transition from high-cost high-performance sectors to commodity manufacturing for automotive, sporting goods, industrial applications, and infrastructure [2]. These high-volume market sectors require low-cost, robust processes, high surface quality, and a reduction in the number of steps and degree of difficulty required to manufacture parts [22]. The ability to achieve these requirements necessitates the concerted analysis and re-design of the prepreg format itself.

In this work, a new prepreg format is presented that features high through-thickness (z-direction) permeability. This experimental prepreg, designated "USCpreg," is produced using a simple one-step process designed specifically for VBO cure. The through-thickness pathways for air transport present in USCpreg result in near-zero internal porosity and flawless surface finish in parts cured out-of-autoclave under VBO conditions. The key advantage of USCpreg is that the material design renders air and volatile evacuation essentially size-independent, as gas transport occurs through the part thickness over distances of millimeters, as opposed to edge-breathing which is highly size dependent. This study describes the processing of lab-scale laminates with USCpreg and a quantitative comparison of part quality to that achieved with traditional VBO prepreg formats.

2. PREPREG FABRICATION AND CHARACTERIZATION

Three prepreg formats were selected for this study, all of which were fabricated using the same resin and carbon-fiber fabric. The only difference between the prepregs was the format (resin distribution). Images of each prepreg surface were acquired using a digital camera, and the surface topography of the prepreg and dry fabric was measured using an optical surface profilometer (NexView 8000, Zygo Corporation, USA).

2.1. Resin and fiber

The resin selected for this work was a standard urea-accelerated dicyandiamide epoxy, formulated and mixed using commercially available components. Specifically, the resin consisted of 60% solid epoxy derived from a liquid epoxy and bisphenol-A (EPON Resin 1001F, Momentive/Hexion) and 40% medium viscosity liquid epoxy produced from bisphenol-A and epichlorohydrin (Epikote Resin 828, Momentive). To this mixture, we added 6 phr of pulverized dicyandiamide (Technicure D-5, A&C Catalysts) and 3 phr of aromatic substituted urea (Technicure MDU-11, A&C Catalysts). This recipe yielded an epoxy resin with a room temperature viscosity sufficient to prevent cold flow, and low enough at high temperature (~1 Pa s minimum) to fully impregnate dry regions of the fiber bed during the cure cycle. The viscosity profile of the resin for the cure cycle used in this study is shown in Fig. 1. While a single epoxy formulation was used in this work, results are applicable to a variety of resin systems and have been reproduced using additional resin formulations

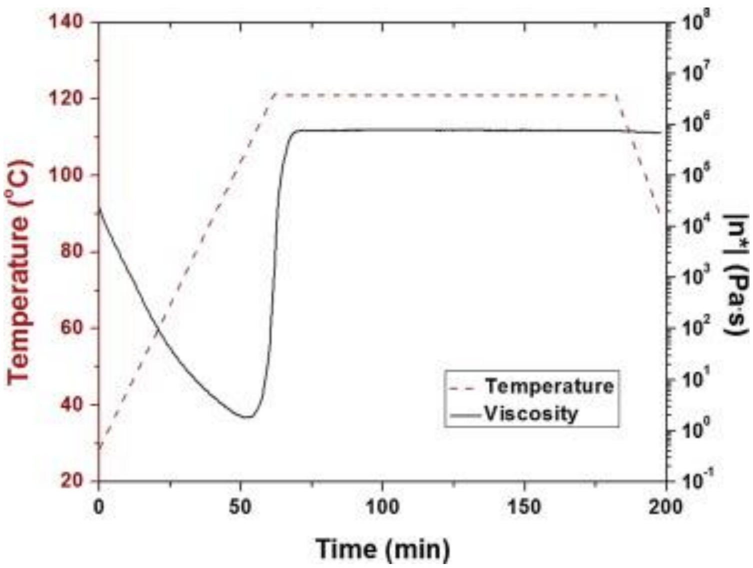

Figure 1. Viscosity profile of epoxy resin during cure cycle. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

To produce prepreg, the epoxy resin was partially impregnated into a 2 × 2 twill carbon fiber fabric with 6000 fibers per tow and an areal weight of 370 gsm. Resin content for all prepreg produced in this work was nominally 35% by weight. Resin impregnation was carried out with three distinct approaches, resulting in prepregs which will be referred to as: (1) control, (2) one-sided, and (3) USCpreg. The design of each prepreg type is depicted in Fig. 2, and described below.

2.2. Control

A control prepreg was fabricated to replicate the resin distribution in conventional VBO prepregs and serve as a benchmark (see Fig. 2a). The control material consisted of uniform resin films partially impregnated into the top and bottom surfaces of the carbon fiber fabric, leaving in-plane EVaC's. Resin content in USCpreg was 35% by weight, determined via weight measurements of stacks of prepreg plies. The measured resin content in the USCpreg was then used to determine required resin film thicknesses for the fabrication of control prepregs. The uniform resin films were produced on a conventional hot-melt line (Patz Materials & Technologies, Benicia, CA).

To achieve a resin content of 35% in a two-side impregnated prepreg, 100 gsm film was produced. Dry fabric, cut into pieces measuring 305 mm × 305 mm, was sandwiched between two resin films of the same size and partially impregnated at room temperature by applying 907 kg of force (1 ton-force) for 50 min in a hydraulic press (G30H-18-BCX, Wabash MPI). This force is equivalent to 8.9 kN over the sample area, or 95.7 kPa of pressure.

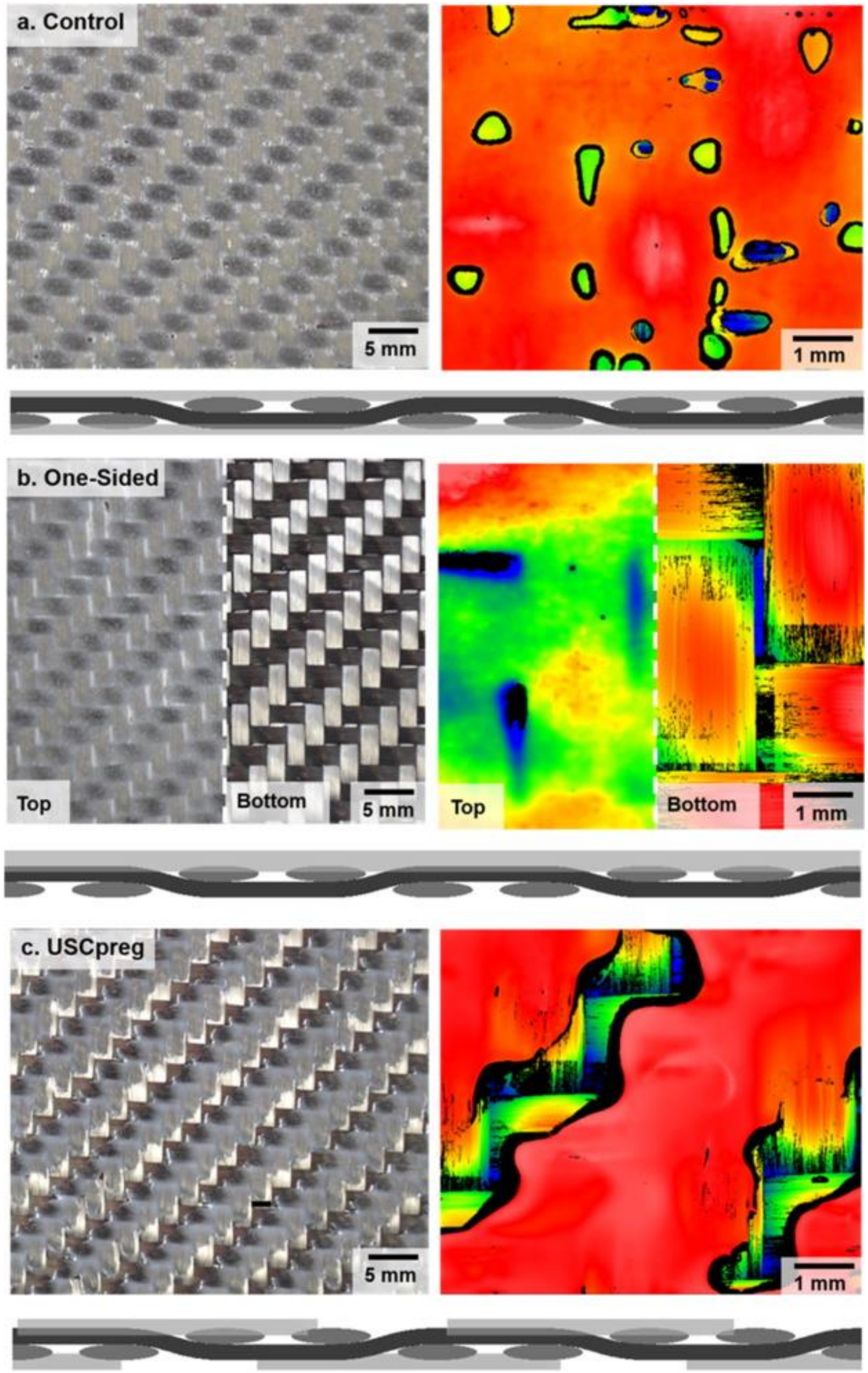

Figure 2. Details of prepreg formats - (a) Control, (b) one-sided, (c) USCpreg. For each prepreg type a photograph of the surface is presented on the left. On the right, a color map shows surface topography (highest regions in red, lowest regions in blue). Below, a schematic cross-section is presented, detailing resin distribution and partial impregnation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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2.3. One-sided

A second common commercial VBO prepreg format is one-side tacky, in which resin is applied to one side of the fabric and the other side remains dry. For completeness, one-sided prepregs were fabricated by partially impregnating 200 gsm uniform film (also processed by Patz Materials & Technologies) onto one side of dry carbon fiber fabric (see Fig. 2b). The impregnation process used to form one-sided prepreg was different, owing to the thicker resin film. For this prepreg, film was placed on top of the carbon fiber fabric and the assembly was vacuum bagged for 25 min at 50 °C to achieve partial impregnation of the fiber bed.

2.4. USCpreg

USCpreg was produced on a custom prepregging line designed, built, and operated by a USC collaborator (Tipton-Goss Advanced Materials Company, Corona,CA). The line differs from traditional hot-melt prepregging systems in that at no point in the prepregging process is a continuous resin film formed. Instead, woven fabric is directly coated with resin as it passes through a pair of heated rollers. Resin for each roller is supplied by heated troughs and metered by a gap set between the rollers and a pair of precision ground doctoring blades. Resin content is controlled by the gap, as well as by the roller temperature and rotational speed. Resin content is monitored on-line during prepregging using a nuclear density (or gamma) gauge. Additional details regarding the equipment and process used to fabricate USCpreg appear elsewhere [25].

Examination of the surface of USCpreg (Fig. 2c) reveals a discontinuous resin distribution characterized by diagonal strips of resin separated by dry fiber regions. This resin distribution is a direct consequence of the roll-coating technique and the woven fabric used to produce the prepreg. Woven fabrics are comprised of tows, or bundles of individual fibers. To create a fabric, tows in the weft direction are woven over and under tows in the warp direction. In a 2 × 2 twill fabric such as the one used in this work, each weft tow is woven over two warp tows and then under two. An offset between adjacent rows creates a diagonal pattern in the weave. By the nature of the weaving process, woven fabrics are neither smooth, nor flat [26] but rather contain periodic valleys and ridges, corresponding to tow underlaps and overlaps. The distribution of the hills and valleys in a fabric is unique to the weave type. The surface roughness inherent to the 2 × 2 twill fabric used in this work is shown in Fig. 3a, with a peak-to-valley amplitude of over 0.3 mm.

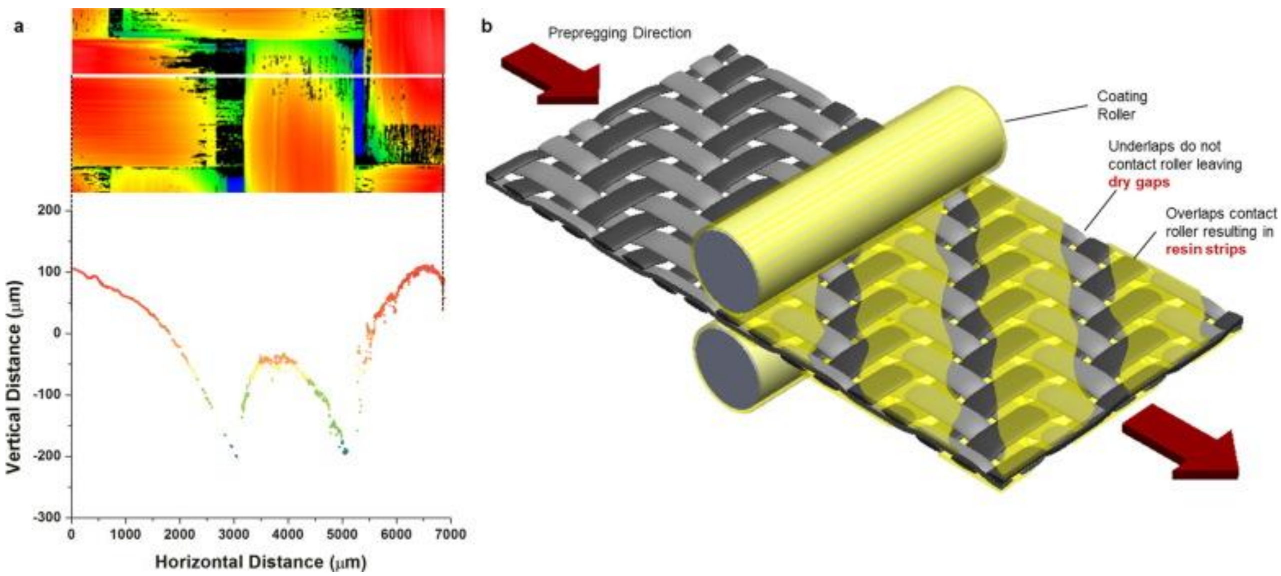

Fig. 3. (a) Topographical map of fabric surface roughness showing surface elevations in different regions of the woven architecture. (b) Schematic representation of prepregging process, in which dry fabric is passed through a pair of resin-coated rollers, producing a discontinuous resin distribution dependent on fabric architecture. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

When prepregged using a roll-coating technique, the raised fiber tow overlaps in the fabric contact the resin coated rollers, while the recessed underlaps do not. This leads to a distinctive "striped" resin distribution via a simple single-step production method. The roll coating technique is shown schematically in Fig. 3b. The technique is applied here on a 2 × 2 twill woven fabric, but the same principle applies to other woven fiber architectures (plain and satin weaves, etc.).

3. EXPERIMENTAL METHODS

3.1. Permeability

The intrinsic (slip corrected) transverse permeability of the prepregs examined in this work was determined using Darcy's law, which describes air flow through porous media. Specifically, a falling pressure method was implemented, in accordance with the experimental approach described by Sequeira-Tavares et al. [27] and [28]. A custom test fixture was utilized for permeability testing [29]. Prepreg was laid up over a cavity of known volume (supported by an open core), covered with a perforated release film and breather cloth, and vacuum bagged. Both the vacuum bag and the cavity were initially at atmospheric pressure. Vacuum was then applied to the assembly, thereby creating a pressure differential. A pressure transducer and data acquisition software (LABView, National Instruments) were used to monitor changes in the cavity pressure as a function of time. The edges of the prepreg were sealed with vacuum sealant tape, such that when vacuum was applied to the bag, air evacuation from the cavity could occur only through the thickness of the prepreg. Permeability was measured for single plies of each prepreg type, as well as for 2- and 4-ply stacks. All tests were conducted at room temperature. In addition to the prepregs produced for this work, the permeability of a commercial VBO prepreg with the same woven fiber architecture was measured for the purpose of comparison (TenCate 275-1 resin, HTS40 3 k fiber, 2 × 2 twill).

Two experiments were performed for each test configuration, and a minimum of three trials were carried out in each experiment. To obtain an average intrinsic permeability value, the pressure transducer was disconnected from the cavity in each trial at the point when the pressure either stabilized or neared zero (indicating air had been fully evacuated). The chamber was then repressurized (to 0.1 MPa) to begin the next trial. Data from the first vacuum cycle was omitted from the permeability test results, as the laminate and vacuum bagging consumables are not yet fully compressed prior to applying vacuum to the layup, allowing air to potentially evacuate more quickly through gaps between the tool, sample and breather or perforated release film. The test set-up and analysis technique are described elsewhere [29].

3.2. Laminate fabrication

A set of laminates were fabricated for this work with the aim of determining the effects of parameters known to lead to voids in parts manufactured with conventional VBO prepregs. To allow evaluation of surface porosity, panels were laid up on an aluminum tool plate using a liquid release agent (Frekote770-NC, Henkel). Release was applied to the tool plate with an areal density of 7.75E-2 kg/m^2. No intermittent debulking was performed during layup.

To replicate ideal processing conditions, baseline panels were fabricated with each of the three prepreg types (control, one-sided, and USCpreg). The panels were 8 plies thick, with a [0°/90°]4s stacking sequence and were 140 × 140 mm. The baseline panels were laid up using standard VBO consumables, including edge breathing dams consisting of vacuum sealant tape wrapped in fiberglass boat cloth. A perforated release film was placed on top of the prepreg stack, followed by a layer of breather cloth and finally a vacuum bag. Prior to the cure cycle, the laminate was held under vacuum at room temperature (~22 °C) for 4 h. Following the vacuum hold, each sample was cured according to the temperature profile shown in Fig. 1: a ramp of 1.5 °C/min to 121 °C, followed by a 2-h dwell at 121 °C, and finally a ramp down to room temperature at 1.5 °C/min.

An identical layup sequence, laminate size and cure cycle were utilized for the remainder of the test panels, with the following exceptions. For samples labeled “no RT vacuum hold” the 4 h room temperature vacuum hold prior to cure was eliminated. Secondly, for samples labeled “sealed edges," the breathable edge dams used in all other panels were removed, and the edges of the panels were instead sealed with vacuum sealant tape to eliminate edge-breathing. Third, for samples labeled "humidity conditioned," no changes were made to the layup or cure cycle when compared to the baseline case, but prior to layup and cure, the prepreg was conditioned for 24 h in a relative humidity of 90% at 35 °C. Finally, for samples labeled "embedded ply drop-offs," both the number of plies and the ply dimensions were altered. A [0°/90°] symmetric layup was used, but embedded ply drop panels were produced using 8 plies 229 × 229 mm, with 4 plies 76 × 76 mm centered at the mid-plane of the layup. All sample configurations and associated labels are shown in Fig. 4. For this proof of concept study, one panel was produced for each experimental test condition. Results have since been replicated in repeat experiments with additional batches of prepreg and various resin formulations, confirming the trends described.

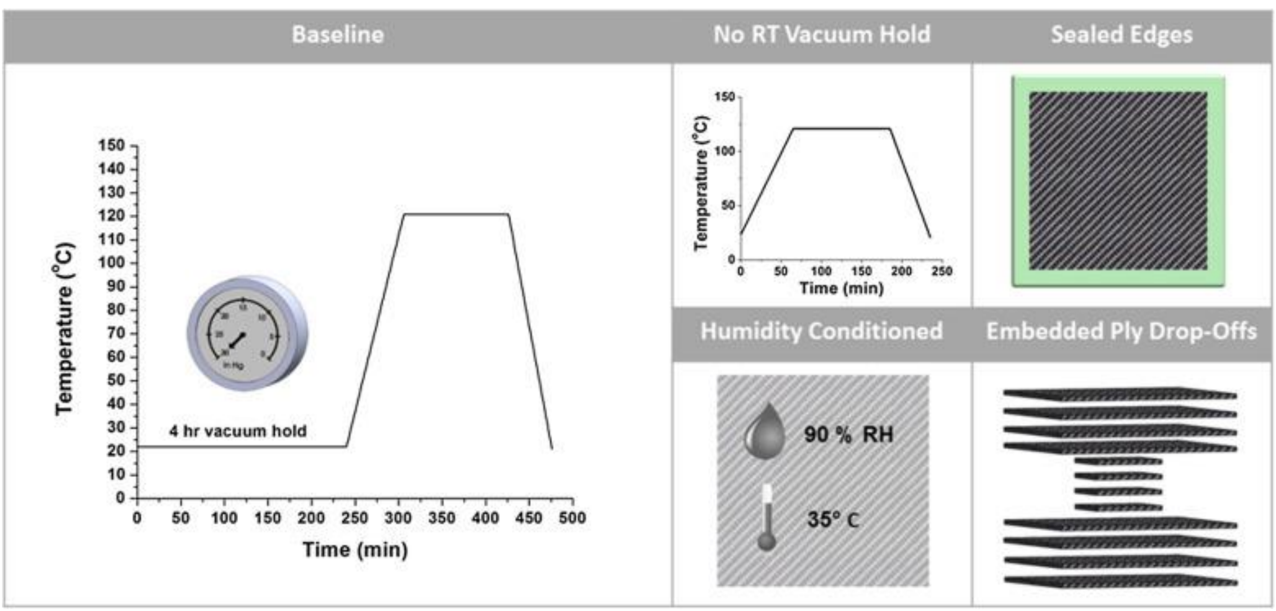

Fig. 4. Test matrix with name and processing details for each laminate type. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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3.3. Surface and bulk void contents

To measure surface porosity, the entire surface of each panel was divided into a grid of cells 12.7 × 19 mm. An image of each cell was recorded at 20× magnification using a handheld microscope (Premier2, Dino-Lite Digital Microscope, USA) and analyzed using image analysis software (ImageJ). In each image, voids were selected and converted to black pixels. All defect-free area was converted to white pixels. The binary image was then used to determine a percent area of voids by dividing the number of black pixels by the total number of pixels in the image, a technique which has been used elsewhere for surface voids [22]. Finally, an average was calculated over all images to provide a measure of surface porosity.

Internal void content was measured in a similar manner. Two samples sectioned from the center of each laminate were mounted, ground, and polished using graded abrasive papers. Images of polished sections were acquired using a stereo microscope (Keyence VHX-600). To image the entire length of each section, a series of approximately 20 micrographs was recorded. Micrographs were converted to binary images to calculate void contents. For internal void measurements, the average void area was taken as a measure of void volume fraction, following standard methods [30].

4. CONCLUSIONS

4.1. Permeability

Results of through-thickness permeability measurements are summarized in Fig. 5, with error bars showing minimum and maximum values. Through-thickness permeability was measured for 1-, 2-, and 4-ply stacks of the three prepregs produced for this work. Additional measurements were performed on a commercial VBO prepreg with a 2 × 2 twill fiber architecture (labeled "commercial"). Regardless of number of plies, USCpreg displayed the highest through-thickness permeability, typically by orders of magnitude. In addition, while all other prepreg types displayed a drop in permeability with increasing ply count, the permeability of USCpreg was independent of number of plies.

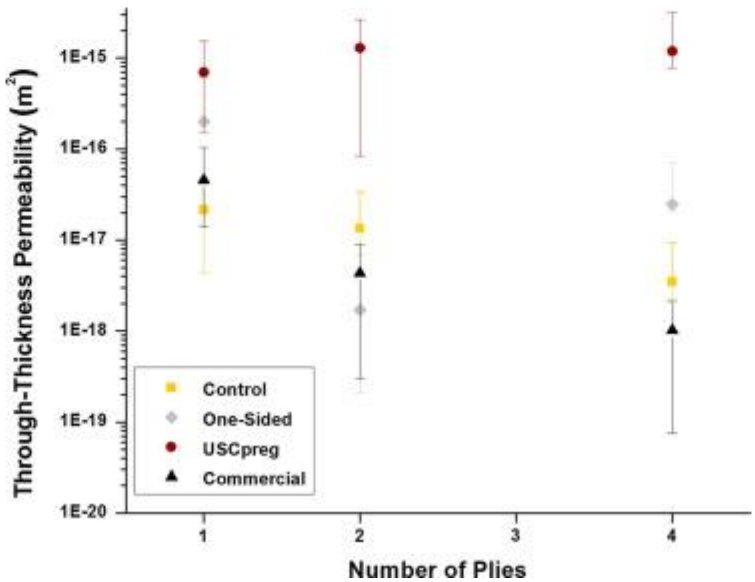

Fig. 5. Through-thickness permeability of the prepregs studied in this work, as a function of number of plies. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Permeability values for the commercial prepreg (10^{-17}–10^{-18}) are consistent with reported values for through-thickness permeability of various VBO prepregs [27], [29] and [31]. Values for the control prepreg fabricated here are greater than those of the commercial prepreg, a result of small perforations in the resin films used. In the coating of the thin film and partial impregnation of the fiber bed, pinhole openings in the resin film were created. These openings are evident in the image of the prepreg surface and in the topographical map in Fig. 2a. The presence of these perforations in the resin film was not intentional, and resulted in a control material with greater through-thickness permeability than typical commercial VBO prepregs.

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Pinhole openings were also present in the thicker film used to fabricate the one-sided prepreg (Fig. 2b). The openings in this case were less frequent, owing to the relative ease of uniformly applying a thicker film, but are larger than those in the control material. The larger holes in the one-sided prepreg are attributed to partial impregnation of the resin film at 50° C. Heating the resin to even a moderate temperature can cause resin to flow away from air pockets. This phenomenon is, in fact, a key attribute of a patent for treating prepregs post-production to enhance through-thickness permeability [26]. The larger size and more sporadic placement of surface openings in the one-sided prepreg is assumed to cause the variability and inconsistency of the measured permeability values. Note that while larger pinhole openings were observed in the resin after prepregging at 50 °C, no noticeable difference was observed in the amount of gas bubbles entrained in the resin prior to cure. Because of gaps in the uniform resin films, the control and one-sided prepregs fabricated here have greater through-thickness permeability than commercial VBO prepregs, but they are still more than an order of magnitude less permeable then USCpreg. This result is significant, as a strong correlation exists between prepreg permeability and the time and quality of vacuum required to achieve a high quality part [7].

The dry fiber channels produced by roll-coating of USCpreg lead to through-thickness permeability values of $\sim 10^{-15}$ m^2. These values are roughly equivalent to the in-plane permeability in conventional VBO prepregs with EVaCs [16]. Note that similar through-thickness values have been achieved in commercial VBO prepregs by using a spiked roller to perforate prepreg plies and increase through-thickness permeability[22] and [28]. Spiking, however, can damage fibers and reduce mechanical properties, and requires time and effort. Moreover, the perforations produced by spiking are localized, whereas the dry fiber regions within USCpreg are uniform at both global and tow scales.

The in-plane permeability of conventional VBO prepregs and one-side tacky prepregs is orders of magnitude greater than through-thickness permeability, which is negligible [16]. Because of this directional disparity, air removal relies almost exclusively on in-plane pathways of egress. When these pathways are occluded, which is often unavoidable as a result of corners, ply drops, or distance, voids will arise in the finished part. Prior studies on surface porosity have shown a strong negative correlation between room temperature vacuum hold time and porosity [22] and [23], sometimes requiring two days or more to fully eliminate porosity from a conventional VBO prepreg [23]. Similar trends have been reported with bulk porosity. Considering in-plane permeability alone, the time required to evacuate air from a layup scales with breathe-out distance as the length of the laminate squared [32], rendering fabrication of large parts with traditional VBO prepregs difficult because of the time and cost of part production. Altering the permeability of the prepreg, however, can reduce or altogether eliminate these size constraints. For example, for a 2.5 m^2 part, an order-of-magnitude increase in permeability (here considering still only in-plane air transport) has been estimated to reduce part cost by 20% by reducing the time required for air evacuation [20]. Incorporating through-thickness permeability and removing air and volatiles over short distances (millimeters) further reduces the time and cost associated with VBO processing.

USCpreg is not the first or only through-thickness permeable prepreg, although the format has advantages over other systems. One such product (ZPREG), is a through-thickness permeable prepreg made from wide strips of resin partially impregnated into one side of a non-crimp fiber bed. Various formats of this material are available. Two common varieties feature 50 mm resin strips with 10 mm gaps [3], and 13 mm strips with 7 mm gaps [35], [36] and [37]. The spacing between resin strips facilitates through-thickness air transport, but also results in long resin flow distances to fully wet-out the fiber bed, and requires the use of a low-viscosity resin. For comparison, we measured the width of the resin strips in USCpreg, as well as the spacing between strips. With the 2 × 2 twill fabric used in this work, the resin strips were 4.0 ± 0.8 mm wide with dry regions of 1.7 ± 0.6 mm between strips. The shorter distance between strips results in shorter resin flow distances in USCpreg than commercial variably impregnated materials, reducing the risk of flow-induced porosity and enabling the use of higher-viscosity resins. Closely spaced strips also ensure that any air entrapped within resin-rich regions is near a dry, permeable region.

The resin strips in commercial materials are independent of fiber architecture. USCpreg, in contrast, takes advantage of the fiber weave to obtain a patterned resin distribution. A different method to enable through-thickness air transport has been patented that, like USCpreg, takes advantage of specific woven fiber architectures [26]. In this technique, as discussed previously, surface openings in the resin are introduced via a thermal treatment performed after initial hot-melt prepregging [26]. The advantage of USCpreg is that the openings in the material are a natural result of fabrication by roll-coating, rendering production of the prepreg a one-step process with no additional treatments required. The greater through-thickness permeability in USCpreg that results from the roll coating method has significant benefits in terms of process robustness and reliability, as well as porosity content in finished parts.

4.2. Surface porosity

Surface porosity often arises in VBO prepreg-processed laminates. This porosity, referred to as "pitting," is a direct consequence of air trapped between the tool surface and the first ply [22]. If trapped air does not have access to an EVaC, it remains after cure as a surface defect. While surface pitting does not significantly alter mechanical performance, it is an aesthetic issue that must be addressed. Release films can be used to produce porosity free surfaces. However, films cannot be applied to molds with contours. Other methods to eliminate surface porosity, including resin-rich surfacing films, paints, and gelcoats, add extra time, weight, and/or cost [22]. Thus, practitioners seek methods to consistently produce composite parts free of surface defects without the need for additional materials or processing steps

Surface porosity was measured for each of the test panels produced, and the values are presented in Fig. 6, with standard deviation shown by error bars. For all three prepreg types, the baseline condition, which included a four-hour vacuum hold at 22 °C prior to cure, showed near-zero surface pitting. Air removal is a time-dependent process, so the lack of pitting in the baseline panels indicates that four hours provides sufficient time to evacuate air from all three of the prepregs for small laminates (140 × 140 mm).

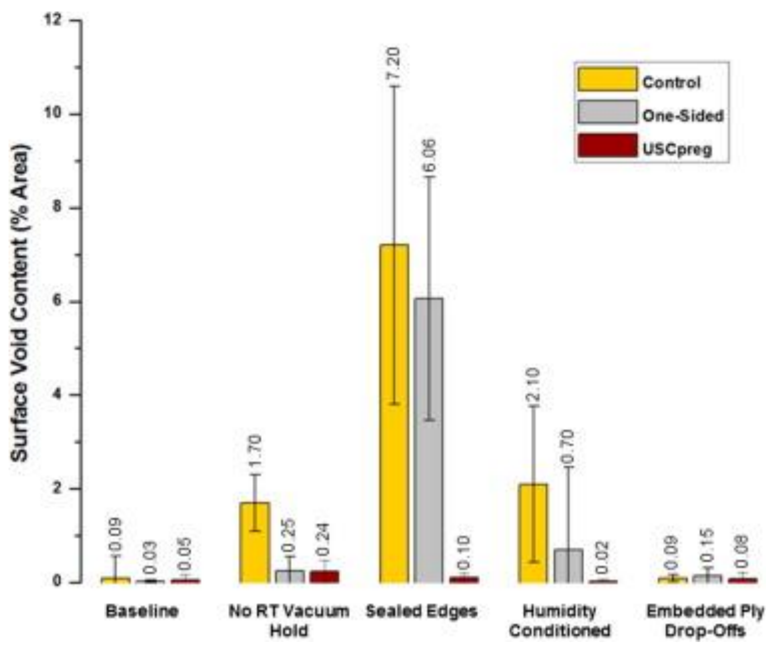

Fig. 6. Average surface porosity for each panel. (Details of panel layup and cure appear in Fig. 4.) (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Eliminating a room temperature vacuum hold caused surface porosity levels to increase for all prepreg types. The greatest increase in surface porosity was in the control material. In the case of the control, air bubbles trapped between the tool and the first ply of prepreg are fully surrounded

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by resin. Trapped air must, therefore, travel long distances in-plane to reach either a breathing edge or a pinhole opening in the resin film that will provide access to the dry fiber EVaC at the ply mid-plane. One-sided prepreg, in contrast, is laid up with the dry side against the tool plate, resulting in an efficient in-plane air evacuation pathway. USCpreg features both in-plane and through-thickness pathways that are readily accessible to air trapped at the tool surface.

The presence of minor surface pitting in all three no-vacuum-hold panels indicates that for the resin system used here, viscosity decreases rapidly enough during cure to occlude evacuation pathways before all air is completely removed (see Fig. 1). Comparison of surface void content in the panel made with control prepreg and the more permeable one-sided and USCpreg materials, however, reveals that while a room temperature vacuum hold is required prior to cure, the duration of the hold can be greatly reduced with the more permeable prepregs.

With a baseline cure and omitting the room temperature vacuum hold, the one-sided prepreg and USCpreg exhibit no significant differences in surface porosity. This finding is consistent with past studies which have shown that one-side tacky prepregs yield high-quality surfaces when edge breathing is feasible [5]. While the potential for low void content is an attractive feature of one-sided prepregs, there are non-trivial drawbacks to the product form. For hand layup, resin on both sides of a prepreg is desired for tack and ease of layup [4] and [9]. Additionally, using prepregs with consistent resin content on both sides reduces the likelihood of technician error in the event that a ply is placed “upside down.” One-side tacky prepreg is used to make laminates through what is essentially a resin-film infusion process. The fiber is minimally impregnated with resin in the initial condition [12] and [13], raising concerns over bulk factor [10] and [14]. Finally, one-sided prepregs have limited through-thickness permeability (see Fig. 5). The benefits of through-thickness permeability for VBO cure are apparent when considering void contents observed in the absence of edge breathing.

A critical requirement for the production of high quality parts with current commercial VBO prepregs is adequate edge breathing [16] and [33]. This requirement is often impossible to achieve, because of part complexity and/or size. Because of these issues, part quality for each of the prepreg types utilized here was evaluated when the edges of each laminate were sealed with vacuum sealant tape, eliminating edge breathing altogether. Elimination of edge breathing also simulates conditions that arise with large parts and prohibitively long breathe-out distances. In this extreme condition, panels produced with USCpreg were nearly free of surface pitting. In contrast, laminates fabricated with the control material and the one-sided prepreg yielded panels with >6% surface porosity (6.06 and 7.20% respectively).

The micrographs in Fig. 7 show the typical morphology of surface porosity for the three prepreg types. The panels pictured were cured with sealed edges (no edge breathing), the condition which resulted in the highest levels of surface porosity of all the laminates produced. The panel produced with the control prepreg (Fig. 7a) shows surface porosity typical of commercial VBO prepregs, consisting predominantly of round pits located near the underlaps of fiber tows [22] and [23]. For panels produced with one-sided prepreg, surface pits are more elongated and larger because the tool-side surface of the ply is initially unimpregnated (Fig. 7b). With one-side tacky prepreg, surface voids are located along the fiber tows and tow underlaps. Finally, in panels produced with USCpreg, the surface finish is nearly perfect (Fig. 7c). Any surface pits that exist are small and circular, occurring near the tow underlaps. Surface voids tend to appear near tow underlaps as a result of air entrapment during layup and/or interrupted bubble migration during evacuation and cure.

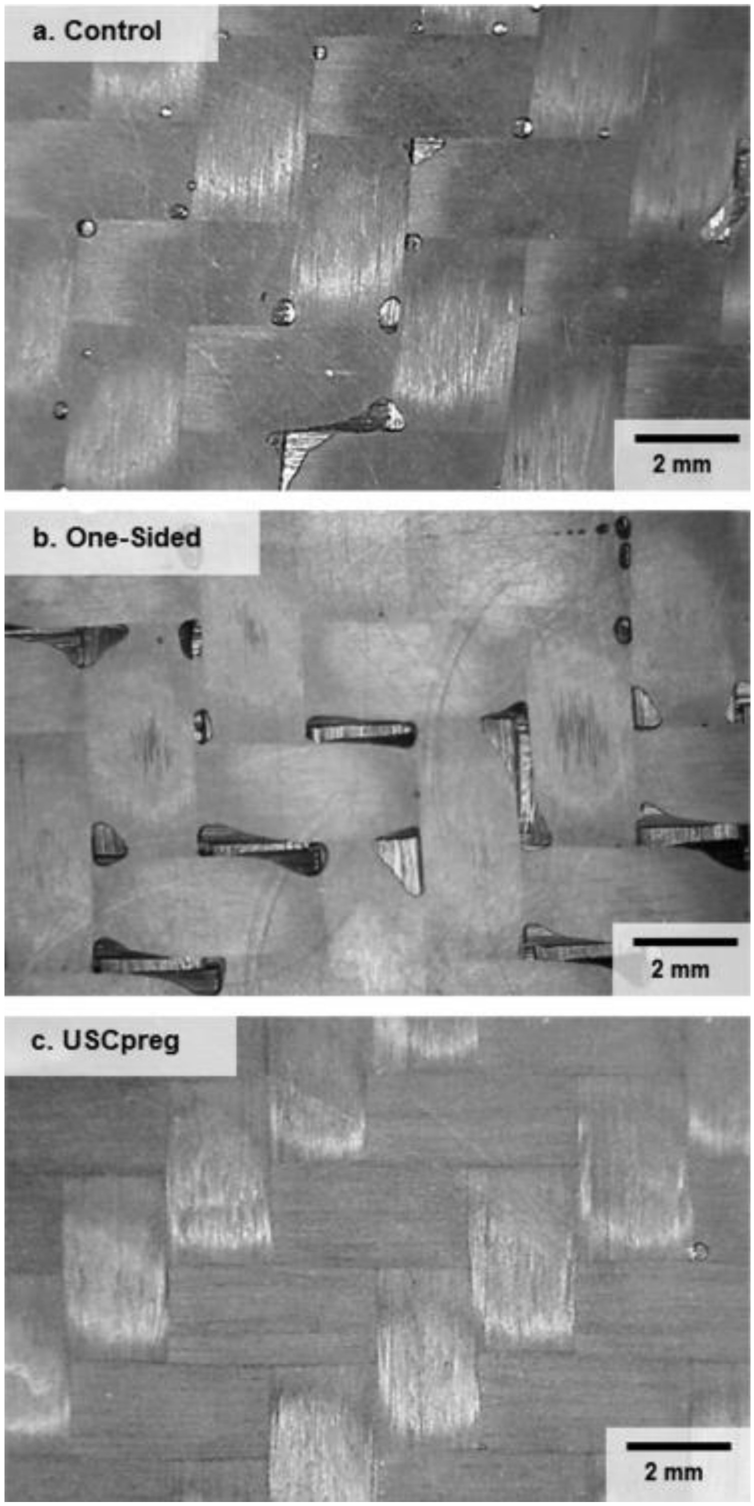

Fig. 7. Micrographs of representative surface porosity for parts cured with sealed edges using (a) control prepreg, (b) one-sided prepreg, and (c) USCpreg.

Embedded ply drops are a common feature of composite parts, and are problematic for VBO prepregs, in which edge breathing is the predominant gas evacuation method. Embedded doublers, for example, are a common element used to increase local thickness where holes will be introduced for fasteners. Because the embedded layers do not have in-plane connections to a breathing edge, porosity commonly arises in and around doublers when traditional VBO prepregs are used [17]. To accommodate embedded layers and to avoid edge effects, the panels fabricated with embedded ply drop-offs were larger than the panels produced to examine other processing conditions. Low surface porosity was observed in panels produced with embedded ply drop-offs, with pitting more pronounced in the region immediately below the embedded layers.

Finally, the role of moisture content was investigated using prepreg plies conditioned in elevated humidity environments. Surface porosity in these panels was closest to values measured for the baseline condition. Past studies have shown that, much like entrapped air, the influence of moisture is less apparent in small laminates, as a room temperature vacuum hold effectively extracts trapped air and water vapor over short distances [19] and [21]. In the absence of through-thickness permeability, however, void contents generally increase with increasing part size and/or reduced vacuum hold time prior to cure [19]. The pathways for air removal designed into USCpreg are equally effective for removal of other volatiles and gases trapped in the layup or generated during cure (such as water vapor) [6]. Enhancing prepreg permeability through engineered and optimized formats represents an effective strategy against gas-induced porosity in prepreg-processed parts, rendering part quality independent of size or moisture content.

4.3. Bulk porosity

Internal porosity values strongly influence mechanical properties and generally dictate part acceptance. Bulk void volume fractions were determined for each of the panels produced in this work, as shown in Fig. 8. Again, error bars show standard deviation.

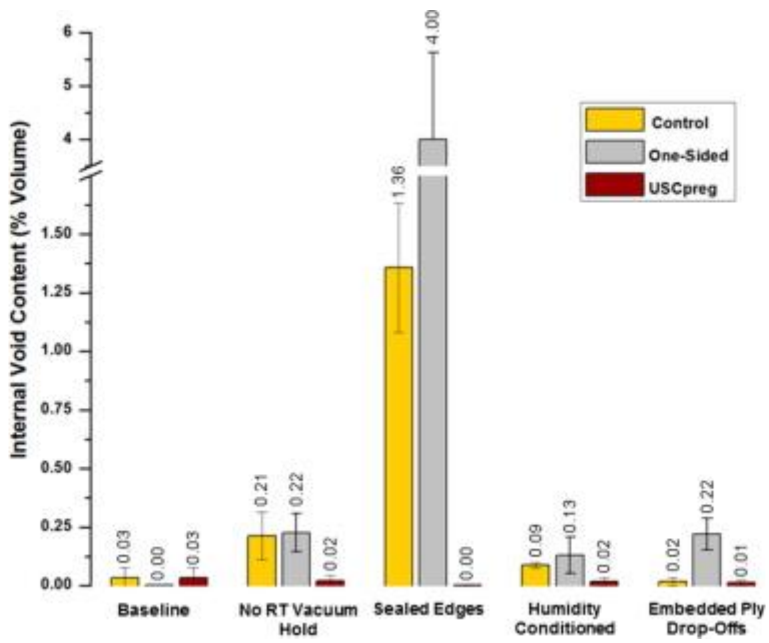

Fig. 8. Average internal void volume fraction for each panel. For details of panel layup and cure, refer to Fig. 4. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Values for internal porosity follow trends similar to those observed in surface porosity results (Fig. 6). In the case of surface porosity, values for the control material tended to be highest, as the tack of the resin film trapped more air at the tool surface than was trapped by the dry side of the one-sided prepreg. For internal porosity, the trend was reversed, and the one-sided prepreg produced the highest void contents. This result is a consequence of the large amount of air initially present in the layup because of the minimal level of initial impregnation of the resin film.

Panels produced with USCpreg showed negligible internal porosity in all conditions studied (Fig. 8). The layup and processing configurations chosen for this study were specifically selected because they represent well-known challenges for VBO manufacturing. Not surprisingly, for the baseline condition, all three prepreg types again produced panels with near-zero void content. The cured panel thickness of each baseline laminate was measured, with all three prepregs producing laminates with an equivalent thickness (3.0 ± 0.1 mm). In void-free laminates, therefore, fiber volume fraction of cured parts is consistent across the three prepreg types. For the other test laminates, however, the control material and one-sided prepreg produced higher void contents and therefore generally thicker laminates compared to USCpreg.

The primary causes of both surface and bulk void formation stem from air entrapment. However, unlike surface porosity, which arises from air trapped between the first ply and the tool surface, internal porosity is caused by air trapped between adjacent plies. In traditional VBO prepregs, air is trapped in resin-rich regions. Without sufficient through-thickness permeability, this trapped air often cannot reach an evacuation channel and subsequently a breathing edge. The high through-thickness permeability of USCpreg eliminates this issue and allows for rapid removal of air from between prepreg plies.

Fig. 9 shows the morphology of internal voids in the three prepreg types. The micrographs, like those presented for surface porosity in Fig. 7, were acquired from panels cured with sealed edges. These samples were chosen because they had the largest void contents of all test configurations studied. In the control material, both macro-porosity and micro-porosity are evident. Macro-porosity consists of large voids between plies, typically near the ends of fiber tows (one macro-void is circled with solid line in Fig. 9a). In addition to these macro-voids that arise from trapped air [34], micro-voids are also visible, located near the center of fiber tows (see dashed circle in Fig.

9a). Micro-voids result from incomplete wet-out of the dry fiber EVaCs with resin, and occur for one of two reasons. The first reason is an improper cure cycle or non-ideal resin properties, both of which can result in a resin viscosity profile that does not allow sufficient time at low viscosity for full impregnation to occur. The other possibility is incomplete removal of air from the initially dry regions at the center of the fiber tows, which prevents complete saturation by the resin [13].

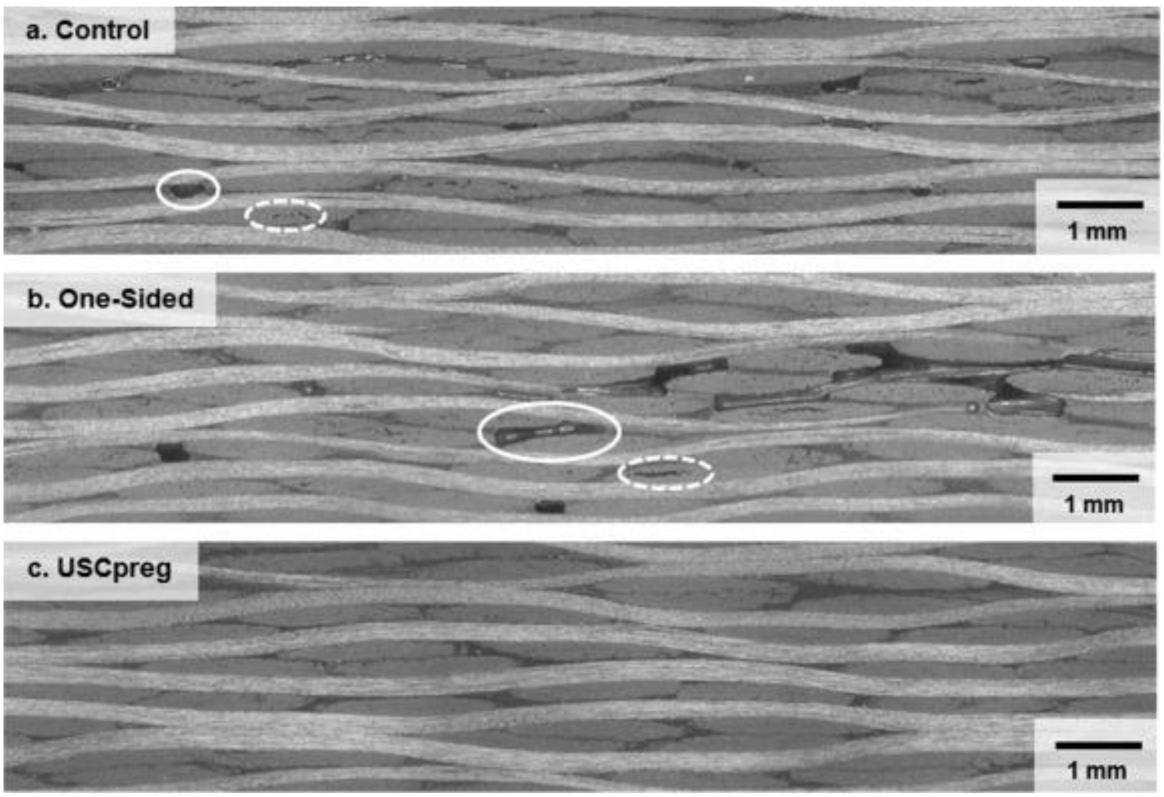

Fig. 9. Micrographs of polished cross-sections for parts cured with sealed edges using (a) control prepreg, (b) one-sided prepreg, and (c) USCpreg. Solid circles highlight macro-voids and dashed circles highlight micro-voids. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In the panel produced with one-sided prepreg (Fig. 9b), both macro and micro-voids are visible (circled with solid and dashed lines, respectively). In this case, however, macro-voids are much larger and more elongated, similar to the surface porosity observed with the one-sided prepreg.

Panels produced with USCpreg showed negligible internal porosity (Fig. 9c). In the instances where small amounts of internal porosity were observed in panels from USCpreg, the voids were small, generally spherical macro-voids. No micro-voids were observed in any USCpreg samples, indicating that the micro-voids observed in panels made with control and one-sided prepreg resulted from incomplete air removal from the dry fiber tows, and not resin flow issues. Incomplete air removal is undoubtedly responsible for voids in samples cured with no edge breathing, because direct pathways for intra-tow air removal do not exist in control and one-sided prepregs, and the only available breathing direction is through-thickness.

The results presented show that prepreg format is critical to final part quality when VBO processing is used. The prepregs produced for this study contained the same resin and fiber bed, and the same resin content. Additionally, all three prepregs were partially impregnated. The method of impregnation, however, and the prepreg format (i.e., the distribution of resin and dry fiber regions) greatly influenced the robustness of part production under a range of processing conditions. The resin distribution in USCpreg is a consequence of the woven architecture of the carbon fiber fabric, and has not yet been modified or optimized.

5. CONCLUSIONS

We have described basic material and process characteristics for USCpreg, a robust prepreg format for processing composite parts out-of-autoclave. We produced USCpreg using a direct roll-coating technique, which results in dry regions on the surface of the prepreg, promoting through-thickness air transport. Using prepreg with this novel format, parts cured even under challenging conditions, such as absence of edge breathing, were void free and contained negligible surface pitting, even in the absence of a release film. Comparisons between USCpreg, one-side impregnated prepreg, and control material made to replicate the uniform resin film structure of commercial VBO prepreg systems, revealed that defects can be virtually eliminated in VBO laminates by simply altering the prepreg format appropriately.

Both surface and internal porosity levels (Figs. 6 and 8) were near-zero in all laminates produced with USCpreg. Laminates produced with control prepreg or one-side impregnated material, however, in some cases showed levels of porosity that normally would be considered unacceptable. While the specific void fractions in some of the test cases examined were minimal (specifically, humidity conditioned samples and laminates containing internal ply drop-offs), void sources generally increase with increasing part size and complexity, and the samples produced in this work were small and flat. The low porosity levels observed, therefore, are expected to translate to higher porosity levels in larger parts. Additionally, for the purposes of this study, each manufacturing issue of interest was isolated, and addressed individually. Real parts often contain multiple complexities, and may present a combination of embedded ply drops, corners, and long in-plane breathe-out distances. In such cases, void formation issues can be expected to compound. For traditional prepregs, this compounding will have an additive effect, with void content rising steeply with part size and complexity. However, void-free parts are expected from USCpreg, regardless of size and complexity, because void contents were essentially zero in all test samples. Finally, note that the control and one-sided prepregs produced for this work had greater through-thickness permeability than commercial VBO prepregs, and thus void contents from commercial VBO prepregs can be expected to be higher than those reported here.

The scope of this work was restricted to gas-induced void formation mechanisms. Void formation in general, however, can be separated into gas-induced and flow-induced mechanisms [16]. While the required flow distances in USCpreg (on the order of 2 mm) are shorter than those in comparable products (ZPREG), they are longer than flow distances required to achieve full fiber wet-out in conventional VBO prepregs. While the longer flow distances are not problematic with fresh prepreg, flow-induced porosity may be more pronounced with increasing out-time and increased resin viscosity [38]. Out-time effects are a subject of current investigation, along with the relative bulk factor of USCpreg in comparison to traditional VBO materials. Bulk factor emerges as an issue because lower initial degrees of impregnation increase layup thickness and thus require greater reductions in thickness during cure. This thickness change can lead to problems of bridging and wrinkling in corners [10]. While these potential drawbacks to the product form are being addressed, so too are applications that would benefit from the USCpreg design. Specifically, USCpreg is expected to be useful for composite tooling, where near-perfect surface finish is required, as well as for co-cure over honeycomb core and composite scarf repair, both of which require through-thickness air removal.

The unconventional format of USCpreg consistently yields high-quality parts without an autoclave, and without the need for complex lay-up arrangements, or specialized procedures. While the benefits of processing composites out-of-autoclave are apparent, the principal shortcoming of vacuum-bag prepreg has been, to this point, a lack of robustness and part consistency. USCpreg represents a potential solution to these issues, and a format suitable for both high-performance applications and consumer products.

The current paradigm shift in the commercial airplane industry involves a transition from metallic structures to composite designs. Autoclave processed parts are currently in service on the Boeing 787, a wide-body commercial aircraft. For single-aisle aircraft (737 or equivalent), however, production demands are considerably higher than for wide-body models. In this context, autoclaves constitute a bottleneck that limits throughput. The forecast production rates for single-aisle aircraft (~50/month) cannot be met with autoclave processing, and will require a manufacturing solution that can bypass the autoclave while at the same time producing large, contoured parts with high quality. USCpreg could offer a potential solution pathway to enable production of aerospace-grade structures.

In addition to high-performance applications, there is strong interest in translating composite materials and processing to commodity manufacturing. The reduction in cost and cycle time, and the increase in part quality achievable with USCpreg may lead to adoption of composite manufacturing in automotive, sporting goods, and infrastructure applications. Promoting through-thickness permeability, and thereby eliminating the need for edge breathing, increases the robustness of materials and processes, which may ultimately enable expansion of VBO manufacturing to new parts and markets.

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