THROUGH-THICKNESS PERMEABLE PREPREGS FOR ROBUST MANUFACTURING

William T. Edwards¹, Patricio Martinez², Timotei Centea³, and Steven R. Nutt⁴

M.C. Gill Composites Center, Department of Chemical Engineering and Materials Science, University of Southern California, 3651 Watt Way, VHE-406, Los Angeles, CA 90089, USA
¹Email: wtedward@usc.edu, ²Email: mart136@usc.edu,
³Email: centea@usc.edu, ⁴Email: nutt@usc.edu

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Abstract
To improve the economics, flexibility, and speed of composite manufacturing, out-of-autoclave/vacuum bag-only (OoA/VBO) processing emerged as a low-cost alternative to traditional autoclave cure. While such techniques have been demonstrated to produce defect-free laminates under ideal conditions, OoA prepregs and VBO processing not sufficiently robust for widespread commercial adoption, particularly in the aerospace industry. This paper explores the relationship between prepreg format, through-thickness permeability, and robust OoA manufacturing of prepreg laminates. A method for producing USCprepreg using a mask-and-press technique is described for the creation of prepregs with customized resin distribution. The format of uncured prepreg is characterized using light microscopy, and a custom-built fixture is employed to confirm that USCpreg exhibits exceptional through-thickness permeability. Samples are cured from USCpreg and a conventional prepreg format under ideal and sub-optimal VBO conditions, and surface and bulk defects in the resulting laminate are characterized to determine the relationship between format and the sensitivity of part quality to non-ideal processing. These results are compared to conventional OoA prepregs, and results confirm a clear relationship between through-thickness permeability and reliable part quality. Defect formation mechanisms are identified and evidence is presented that shows that even minor differences in resin topography affect part quality.

1. Introduction

Autoclave processing of composites can reliably produce components with defect levels inferior to standard part rejection thresholds in risk-averse industries, including aerospace. The super-ambient compaction pressure applied during autoclave cure ensures that laminates are fully consolidated and void-free, but acquisition, operation, and maintenance costs for autoclaves can be cost-prohibitive. Additionally, autoclaves limit production flexibility and part size. As such, composites manufacturers have long sought alternatives to autoclave processing, developing out-of-autoclave (OoA) techniques such as vacuum infusion and compression molding. Of these techniques, vacuum-bag-only (VBO) prepreg processing is uniquely suited for precise control of laminate fiber volume fraction, resin content uniformity, and fiber orientation [1].

The distribution of resin in commercial VBO prepregs has evolved from prepreg formats initially used in autoclave processing. A solution-dipping method was developed in which dry carbon fabric and unidirectional (UD) tape was immersed in a bath of solvated resin, fully saturating the fiber bed. Complete removal of solvent after fiber bed impregnation, however, was not viable, and residual volatile species manifested as porosity in cured parts. Hot-melt prepping avoids solvents altogether and has largely replaced solution-dipped prepreg in commercial use [2]. Hot-melt prepreg fabrication consists of applying resin films onto one or both sides of dry fabrics or tape. Fully or partially-saturated prepreg
formats can be produced using hot-melt processes by controlling lamination speed and levels of applied heat and pressure, yielding symmetric or asymmetric resin distribution relative to the ply midplane [3].

In 1987, Thorfinnson and Biermann [4] compared the quality of laminates produced from fully and partially-saturated prepregs. Data indicated that laminates manufactured from fully-saturated prepreg exhibited obvious porosity, while those produced from prepregs with incomplete saturation were nearly defect-free. This study provided the first published evidence that resin distribution within the ply (prepreg format) is critical to part quality. Repecka and Boyd [5] identified that unimpregnated regions of fiber tows created a network of pathways through which entrapped and evolved gases could be evacuated during cure. Because these channels were oriented along the fiber direction, air transport occurred primarily in-plane. For such evacuation paths to be effective, laminate edges must remain permeable to gas transport during debulking and the initial stages of cure, requiring edge breathing dams.

Most commercially-available OoA/VBO prepregs rely on in-plane evacuation of gasses to limit void content. Under ideal circumstances, VBO prepregs can yield nearly defect-free parts, rendering oven cure competitive with the autoclave [6]. Frequently, however, variations in storage, layup, bagging, vacuum level, and other processing conditions can compromise part quality [6–9]. Further, large parts, complex geometries, and internal ply drops can present additional challenges for OoA processing, leading to defect-filled parts even under ideal processing conditions [10–12]. Because part quality in VBO processed prepregs is sensitive to these issues, VBO processing has not yet established the necessary track record of reliability necessary for widespread adoption, and risk-averse industries continue to manufacture laminates in the autoclave.

New developments have indicated that the limited reliability of OoA/VBO processing can be overcome through innovation in prepreg format design. A novel prepreg format that allows through-thickness gas transport has been used to produce near-zero internal porosity and defect-free surfaces even in challenging VBO conditions [13]. This format, dubbed “USCpreg”, is characterized by a discontinuous resin distribution on each side of a ply. However, the process used to produce the initial embodiment cannot tailor resin distribution for a given fabric geometry, and cannot be used with unidirectional (UD) fiber beds. The development and validation of UD prepregs with high transverse permeability is motivated by the large fraction of the overall market held by UD mats and tapes.

This paper describes a lab-scale technique for producing prepregs with customizable resin distribution is described. The technique can be used to produce both woven and UD prepregs, although here, efforts focus on the unidirectional case. A UD prepreg with discontinuous resin distribution (U0SCpreg) was fabricated and characterized, with results confirming that the material exhibited a through-thickness permeability orders of magnitude greater than that of conventional UD prepregs formats. Parts were fabricated from U0SCpreg and conventional OoA prepreg under baseline and a variety of sub-optimal processing conditions, and the resulting bulk and surface porosities were measured using light microscopy. Insight into fundamental void formation mechanisms was gleaned from this data and used to inform an investigation into the influence of subtle differences in resin topography. Resin topography modifications were made to U0SCpreg plies, plies were cured, and surface defect content was compared. Overall, the results confirm that discontinuous resin distributions provide significant advantages during OoA/VBO cure, and impart markedly higher robustness to sub-optimal process conditions.

2. Materials, Prepreg Fabrication, and Characterization

2.1. Materials and Prepreg Fabrication

Prepreg (U0SCpreg) was fabricated in-house from 305-315 g/m² unidirectional non-crimp carbon tape (#2583, FibreGlast) consisting of 12K tows held together with minimal binder powder (0.7 wt. %) and supported by polyester threads. Polyester support material ran orthogonal to the fiber direction and was hot-bonded to the surface of the fiber bed every 8.5 mm. An epoxy resin film (PMT-F4, Patz Materials...
& Technologies) was used for all U\textsubscript{UD}SCpreg. Resin film was produced and delivered on a backing paper and was filmed to achieve an average areal weight of 152.5 g/m\textsuperscript{2}. A commercially-available unidirectional tape prepreg (Cycom 5320-1 IM7 12K 145gsm 33\% RW UD, Cytec Solvay) representative of state-of-the-art OoA materials was selected as a control for benchmarking the formats described below.

Prepreg was fabricated using a mask-and-press technique by selectively depositing resin film on the fiber bed. Resin film was laminated with a release paper, and passed through an automated cutting tool (R19 Desktop Vinyl Cutter, Vinyl Express) in which the release paper was cut in a customizable pattern. Portions of the release paper that had been cut were then removed to create a mask (Figure 1A). Masks were designed such that 5.0 mm wide strips of resin were exposed, between which there was a 5.0 mm wide strip of mask. Masked resin film was aligned on each side of a carbon fiber ply (Figure 1B) and pressed for 50 minutes at 400 kPa to transfer exposed resin film onto the fiber bed. Masks and the covered resin were peeled away, revealing a prepreg ply ready for storage or layup (Figure 1C). During this process 50\% of the resin film was transferred to each side of the dry fibers, creating plies with 33 wt. \% resin content.

![Figure 1](image)

**Figure 1.** Key steps in manufacturing customized prepregs using mask-and-press technique and a photograph of the resulting prepreg: (A) laminate resin film with release paper, cut and remove strips; (B) sandwich fibers between opposing masked resin films and press; (C) remove masks and excess film revealing completed prepreg.

To investigate the influence of subtle differences in prepreg resin topography, several plies of U\textsubscript{UD}SCpreg were modified by attaching as-manufactured UDSCpreg plies to patterned and un-patterned release paper and pressing the resulting assembly for a second time at 400 kPa for 50 minutes, embossing release film patterns into the resin strips. Three modifications to the resin strips were explored: an embossed crosshatch pattern, an embossed diamond pattern, and a smoother-than-manufactured surface (created using a flat release film).

### 2.2. Characterization

A custom fixture was used to measure through-thickness permeability of the as-manufactured USCpreg, the control prepreg, and dry UD carbon tape [14]. A single ply of each material was laid up over a cavity of known volume and supported by a honeycomb insert. Vacuum sealant tape was used to fix each ply in place during testing and seal ply edges, minimizing in-plane gas transport. Standard consumables were placed sequentially over each ply: a perforated release film and breather cloth were placed atop the prepreg, and sealed under a vacuum bag.

Falling pressure tests and Darcy’s Law were used to calculate a permeability coefficient for all
materials [15,16]. A vacuum pump and pressure sensor (PX32B1, Omega) were connected to the bag side of the custom fixture and a second identical pressure sensor was connected to the cavity to allow monitoring of the pressure differential across each prepreg or dry fiber sample. Data acquisition was initiated and the vacuum pump was activated. Tests were terminated once the pressure in the cavity was within 1% of the pressure in the vacuum bag or after several hours. At least three falling pressure measurements were conducted on each prepreg format.

The viscosity profile of the epoxy resin used for in-house prepregs was determined for a standard ramp-and-hold cure cycle using a parallel plate rheometer (TA Instruments AR 2000ex). The rheological properties of the resin in the control prepreg have been thoroughly studied, and a model was used to predict viscosity during cure [17]. Figure 2 shows viscosity and temperature vs. time for each resin. This figure highlights that the viscosity profile of the epoxy resin comprising UdSCpreg could likely be improved. Specifically, the resin system of the commercially available prepreg achieves a lower minimum viscosity and remains at reduced viscosity for longer than the UdSCpreg resin formulation. These characteristics both facilitate fiber bed impregnation and gas evacuation and allow more time for such processes to occur.

![Viscosity vs. Time](image)

**Figure 2.** Viscosity profile of resins studied during a standard cure cycle.

3. Laminate Fabrication and Analysis

3.1. Laminate Fabrication

The laminates fabricated can be divided into two classes: those produced to demonstrate the efficacy of introducing through-thickness gas transport pathways, and those created to explore the effect of subtle differences in resin topography on part quality. These groups of laminates will be denoted “as-manufactured” and “embossed”.

Laminates made of as-manufactured UdSCpreg and control prepregs were fabricated on a release agent-treated (Frekote770-NC, Henkel) polished aluminum plate. UdSCpreg laminates followed a [0°, 90°]_{2S} stacking sequence, and a [0°, 90°]_{2S} sequence was used for the control prepreg laminates to achieve equivalent part thickness. Unless otherwise noted, all as-manufactured UdSCpreg and control laminates measured 140 mm by 140 mm by 3.2 mm, were laid up using the same process, and cured using an identical cure cycle. Layup consisted of edge breathing dams, a perforated release film, breather cloth, and a vacuum bag. The cure cycle consisted of a four-hour room temperature vacuum hold, a 1.5 °C/min ramp to 121 °C, a two-hour dwell, and a -1.5 °C/min ramp back down to room temperature.

Exceptions to these baseline conditions were imposed to simulate processing conditions commonly encountered in industrial practice. A “no room temperature (RT) vacuum hold” sample was not held under vacuum for any extended period of time prior to beginning the temperature ramp. A “no edge breathing” sample was laid up with vacuum tape sealing all edges instead of edge breathing dams. A
“humidity conditioned” sample was exposed to 90 % relative humidity at 35 °C for 24 hours prior to cure. Finally, the layup was modified for an “embedded ply drop” sample, in which ply dimensions and stacking order were altered. Specifically, for the embedded ply drop sample, ply dimensions were changed modified and two smaller plies were added at the midplane (for the control prepreg embedded ply drop sample, four additional plies were added at the midplane).

Embossed U₀SCpreg samples followed a [0°, 90°] stacking sequence, cured on a glass tool plate (coated with release agent), and measured approximately 100 mm by 100 mm, but otherwise standard layup and processing conditions were used during cure.

3.2. Bulk and Surface Porosity Measurement

Bulk porosity was measured in as-manufactured U₀SCpreg and control laminates, surface porosity was measured for all laminates. To measure bulk porosity, sections were cut from the center of each sample, potted, polished, and imaged at 150 times magnification (VHX-5000, Keyence). For surface quality quantification, the tool-side of each laminate was imaged in sections at twenty times magnification (Edge AM7815MZTL, Dino-Lite Digital Microscope, USA). Images were manually binarized with defects indicated in black and passed to a script for calculation of the fractional defect content.

4. Results and Discussion

4.1. Through-Thickness Permeability

Gas transport through the dry fiber bed was most efficient. The pressure difference across dry fibers decreased to less than 1 kPa in approximately 30 s and the associated through-thickness permeability was calculated to range from approximately 4E-13 m² to 5E-12 m². When U₀SCpreg was tested, the pressure difference across the ply decayed to less than 1 kPa in approximately 200 s. The associated permeability of the in-house manufactured prepreg was calculated to range from approximately 4E-14 m² to 2E-13 m². The control prepreg was tested for ten hours, but the pressure difference across the ply did not decay below 25 kPa prior to test termination. The permeability values calculated from the control prepreg ranged from 3E-17 m² to 3E-16 m².

These data indicate that the discontinuities in resin distribution lead to formation of through-thickness gas transport paths. The permeability of U₀SCpreg was measured to be about three orders of magnitude higher than the permeability of the conventional prepreg. Additionally, while the presence of resin strips on the surface of U₀SCpreg does reduce the permeability of the dry fiber ply by one order of magnitude (for a single ply), efficient gas transport through the thickness of the ply remains possible. Indeed, the permeability values extracted from trials on dry fibers and U₀SCpreg are likely skewed by the speed with which the pressure across each ply equilibrated.

4.2. Bulk Porosity

Figure 3A shows the average bulk porosity for as-manufactured U₀SCpreg and the control prepreg. Bulk porosity below 2.0% was observed for all samples fabricated from as-manufactured U₀SCpreg. Under baseline conditions, the as-manufactured U₀SCpreg laminate exhibited 0.46% void content. When the part size was increased and an internal ply drop was added, bulk porosity increased slightly to 0.51%. Laminates with no RT hold and no edge breathing exhibited the highest porosity (1.26% and 1.33%, respectively). The humidity-conditioned sample showed the lowest measured bulk porosity (0.30%).

The reduced bulk porosity after humidity exposure was likely caused by changes in prepreg format that occurred during conditioning. Optical profilometry conducted on humidity-conditioned U₀SCpreg
showed that resin flow occurred during conditioning, modifying the resin strip cross-section from rectangular to bell-shaped. These changes outweighed the detrimental effect of additional volatiles (moisture) introduced during humidity exposure, which would typically manifest as a greater void content compared to the baseline.

Bulk porosity in control laminates was more sensitive to sub-optimal processing. At 0.05%, the control prepreg cured under baseline conditions exhibited the lowest bulk void content of all samples produced. The laminate with no RT hold showed increased void content at 0.81%, and laminates produced with other sub-optimal conditions showed a minimum of 2.60% porosity. UDSCpreg contained more defects than the control material under baseline conditions, but nevertheless produced an acceptable part. Conversely, control material was remarkably vulnerable to sub-optimal cure, whereas UDSCpreg was robust. Figure 3B compares micrographs of UDSCpreg and control prepreg laminates cured after humidity conditioning. The control material exhibits considerable interplay porosity, while UDSCpreg is nearly void free. This difference in performance occurred despite UDSCpreg format and the constituent materials having not been optimized.

**Figure 3.** (A) Bulk porosity data for samples fabricated from UDSCpreg and control prepreg under baseline and a variety of sub-optimal conditions and (B) micrographs showing bulk defects in UDSCpreg and control laminates comprised of humidity conditioned prepreg.

### 4.3. Surface Porosity

Figure 4A shows the average surface porosity measured for each of the as-manufactured UDSCpreg and control laminates. Defect levels in the control prepreg depend strongly on processing conditions. While a nearly flawless surface (0.02% defect coverage) resulted from baseline conditions, when edges were sealed, control prepreg was humidity conditioned, or an internal ply drop was added, surface porosity increased to at least 9.9%. Control prepreg surface quality was less sensitive to the removal of the RT vacuum hold as to other sub-optimal conditions, likely because UD tape surfaces entrap little air.

Defect levels in UDSCpreg laminates ranged from 0.20% to 0.93%. Large error bars indicate that the surface quality varied by location for all samples fabricated. The as-manufactured UDSCpreg laminate cured with no edge breathing exhibited the lowest surface quality, while the humidity-conditioned laminate exhibited the fewest defects. The lower pore content in humidity-conditioned UDSCpreg was attributed to the changes in resin strip topography induced by conditioning.

Further evidence that subtle differences in resin surface topography influenced laminate quality is presented in Figure 4B. UDSCpreg embossed with a diamond pattern was characterized by the highest surface defect levels, measuring 1.54%. The diamond embossing pattern entrapped more air than the as-manufactured surface between resin strips and the tool plate during layup, leading to greater surface defects. The smooth-pressed and hatch-embossed UDSCpreg laminates displayed the lowest surface porosity (0.08% and 0.12%, respectively). After smooth pressing, the convex resin strip cross-section reduced the volume of gas entrapped between resin and tool plate during layup. After hatch embossing,
the pattern transferred to the resin strip created evacuation pathways for gases that would have otherwise been entrapped.

![Figure 4](image)

Figure 4. Surface porosity data for (A) samples fabricated from U₀Scpreg and control prepreg under baseline and a variety of sub-optimal conditions and (B) samples of embossed U₀Scpreg cured under baseline conditions.

5. Conclusions

This work introduces a means for customizing prepreg formats and leverages this method to identify fundamental consolidation mechanisms. Better understanding of such mechanisms can inform the design of next-generation OoA prepreg formats. The results highlight the importance of through-thickness gas transport pathways for resistance to commonly encountered sub-optimal processing conditions. Additionally, evidence indicates that benefits are not unique to woven prepregs, and can meaningfully improve process robustness in UD tapes. The results comparing as-manufactured U₀Scpreg to the conventional hot-melt format are particularly significant considering the rheological differences between the two resin systems compared. Specifically, the resin used in U₀Scpreg formats does not reach the same minimum viscosity as the control prepreg and remains at its minimum viscosity for a considerably shorter period, limiting the time during which entrapped or evolved gasses can be removed from the laminate.

The U₀Scpreg materials provide convincing indications of the efficacy of introducing through-thickness evacuation pathways, despite being unoptimized and likely sub-optimal. Indeed, the surface porosity data collected on embossed U₀Scpreg samples clearly demonstrates that subtle differences in resin topography can meaningfully influence laminate quality. Similar mechanisms operating to create surface porosity are likely responsible for a significant fraction of bulk defects, and future work will aim to investigate the effects of resin distribution, degree of penetration, and surface topography on defect control phenomena and robustness.

Broadly, this work represents a step toward addressing a growing need for rapid, flexible, robust, and cost-effective high-performance composite manufacturing techniques. Growing demand for composite structures, limited availability of autoclaves, and the inherently unsuitable nature of autoclave processing in some scenarios (e.g. in-field repair, large structures) will continue to drive the need for such techniques. The approach presented here effectively transfers the robustness of autoclave cure into the material itself, and further development will accelerate the adoption of VBO prepregs as enabling technologies for numerous applications of composites while reducing barriers to entry.

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References