Compression molding of reused in-process waste – effects of material and process factors

M.-S. Wu a, T. Centea and S. R. Nutta

aDepartment of Chemical Engineering and Materials Science, University of Southern California, Los Angeles, CA, USA; bDepartment of Aerospace and Mechanical Engineering, University of Southern California, Los Angeles, CA, USA

ABSTRACT
Effective strategies for the reuse and recycling of in-process prepreg waste are needed to reduce economic and environmental costs. In this paper, we investigate the compression molding of prepreg waste converted into scrap “chips” (or strands). Material is randomly distributed within a lab-scale closed mold and cured with control of temperature and pressure. Material properties and process parameters such as chip geometry, fiber bed reinforcement, resin state, and cure cycle are varied and shown to influence porosity and thickness. These experiments clarify the phenomena governing microstructural quality and identify manufacturing pathways for high-quality parts. In addition, mechanical properties are measured for laminates with high and low defect levels. The study demonstrates the viability of prepreg reuse. Furthermore, the resulting insights provide a basis for practical science-based optimization of the reuse of production prepreg waste.

Introduction
Composites are increasingly used in multiple industrial sectors due to their high-specific mechanical properties (relative to traditional metals) and the ability to form lightweight, complex structures.1 Globally, the composites industry grew by 4.5% in 2015. Furthermore, the global composite materials market is projected to grow at a promising CAGR (Compound Annual Growth Rate) of 12.94% during the forecast period of 2015–2020.2,3 An estimated 1.8 million kilograms of carbon fiber-reinforced plastics is produced each year in the USA.4

Unfortunately, this market growth also increases the amount of material waste generated during production. For composites, in-process waste usually consists of scrap generated during cutting or time-expired materials and can comprise up to 30% of the initially purchased quantity. Traditionally, composite structures in aerospace applications begin as carbon fiber “prepreg” or carbon fiber beds “pre-impregnated” with an uncured polymer resin. During manufacturing, plies with the required shape, size and fiber orientation are cut from roll stock. During this process, some material is necessarily left over as waste. Furthermore, prepregs have a limited storage life because the uncured resin matrix is catalyzed and undergoes polymerization even at cold storage temperatures. Most applications require that prepreg not exceed a specified shelf life and out-life. Thus, when prepreg exceeds either of these limits, it is discarded. Altogether, a remarkable amount of processing waste is generated: according to literature,5 about 80% of carbon fiber is processed as prepreg and up to 40% of carbon fiber prepreg may be scrapped. The consumption rate of carbon fiber is growing at over 10% per year and is projected to reach a value of approximately $5 billion by 2020.6 Reducing up-front material costs can mitigate some of these economic losses. However, technologies and strategies that reduce the net amount of unused prepreg are essential to improve cost-effectiveness, reduce environmental impact and provide opportunities for downstream manufacturing.

Historically, processing waste has been incinerated or stored in landfills due to a lack of effective recycling or reuse technologies. Both disposal options have detrimental economic and environmental effects. Moreover, emerging environmental regulations are increasingly forcing composites users to adopt new reuse, recycling or disposal solutions.7 As a result, interest in recycling and reuse of composites has grown in recent years.

A major challenge in recycling composites stems from the presence of multiple constituent phases. For prepregs with a thermosetting polymer matrix, for which cure is irreversible, it has generally only been possible to reclaim the higher-value, inert fibers by eliminating the matrix through pyrolysis or chemical removal. Techniques for recycling of thermoset composites have been reviewed...
by Pimenta et al.\textsuperscript{7} and Oliveux et al.\textsuperscript{8} In addition, technologies for reclaiming recycled fibers have been commercialized (e.g. Materials Innovation Technologies and Adherent Technologies).\textsuperscript{9} However, in general, methods that discard the resin eliminate a highly optimized engineered material, as well as the economic and physical resources used to produce it. Thus, if and when possible, the direct reuse of prepreg waste is desirable, eliminating the need for chemical digestion or pyrolysis. Finally, while cured composites are recycled at the end of service life, in-process waste is generated during manufacturing, resulting in an immediate and growing problem.

Since prepreg offcuts commonly consist of random shapes, sizes and orientations, they must be converted into a more versatile product form. Randomly oriented strands (ROS) or prepreg "chips" have been proposed as an intermediate form that facilitates the fabrication of composites with complex geometries at the expense of some mechanical performance.\textsuperscript{10} Feraboli et al.\textsuperscript{11} characterized the elastic behavior and failure of compression-molded flat plates made from carbon fiber/epoxy prepreg chips, showing that moduli and strengths are influenced by the chip aspect ratio, and that failure is matrix-dominated. The authors also used digital image correlation (DIC), which offers both repeatable modulus measurements and critical insights about non-homogenous behavior.\textsuperscript{12} Further, Feraboli et al.\textsuperscript{13} investigated the notched behavior of chip-based parts, demonstrating that macroscopic response is largely notch-insensitive due to the complex stress state within the randomized meso-structure. Finally, Feraboli et al. analyzed the link between defect levels and performance. They identified defect classes, including porosity, sudden changes in fiber orientation, and resin-rich regions, but showed that defect locations did not consistently predict failure locations.

The production of composites from thermoplastic-matrix prepreg chips has been studied by Eguemann et al.\textsuperscript{14} who demonstrated that composite parts with complex geometries can be fabricated from reused carbon fiber/PEEK prepreg strands. Likewise, Selezneva et al.\textsuperscript{15,16} determined mechanical properties of such composites as a function of both material and test specimen characteristics, identifying trends similar to those observed for thermoset composites, and proposed a stochastic approach for modeling mechanical performance.\textsuperscript{17} Leblanc et al.\textsuperscript{18} studied the effect of processing conditions on the forming of ribbed features, showing that complex features can be produced if the molding pressure exceeds a sufficient filling pressure, and that initial strand placement can reduce mechanical performance if it creates merging flow fronts. Levy et al.\textsuperscript{19} proposed and demonstrated a model capable of predicting the inter-strand void content for discontinuous CF/PEEK parts, and produced design charts relating process parameters, chip properties, and final part characteristics. Finally, Landry and Hubert\textsuperscript{20} investigated defect formation mechanisms for such composites and identified non-uniform shrinkage due to thermal strains at the onset of crystallization as a critical process leading to voids and reduced mechanical performance.\textsuperscript{21,22}

The literature cited above has shown that prepreg chips can be used to produce composites by compression molding, but further research is needed. Unlike sheet molding compounds (SMCs), glass mat thermoplastics (GMTs), and long-fiber-reinforced thermoplastics (LFTs), prepregs are typically high fiber content materials optimized for automated or hand layup followed by autoclave or vacuum bag-only cure. Consequently, fiber bed, resin and prepreg properties are not designed for processing within a closed compression mold. Second, in the studies cited in the previous paragraph, prepreg chips have typically been sourced from pristine material and were likely unaffected by aging. Conversely, in-process waste is likely to accrue out-time (or exposure to ambient temperature) and exhibit sub-optimal, tack, viscosity and other process-relevant properties.

The reuse of in-process prepreg waste through conversion into randomly oriented chips and compression molding must also be assessed relative to compression molding of SMCs, GMTs, and LFTs. These mature manufacturing routes for low- to medium-performance structural parts consist of multiple steps, including compound/charge preparation, molding, secondary working, and subsequent finishing (e.g. bonding and painting).\textsuperscript{23,24} To achieve high production volumes and fast cycle times requires system-level optimization of materials, tooling, and processing, as well as practical manufacturing considerations. Such optimization requires a detailed understanding of material and process phenomena.

In this study, we investigate the viability of using scrap vacuum bag-only (VBO) thermoset prepreg from in-process waste to produce composite parts. We specifically consider differentiating characteristics between VBO prepreg and material specifically designed for compression molding, including initial prepreg microstructure, thermal cure cycle, and accrual of out-time. We manufacture lab-scale composites within a closed mold while varying material and process factors across manufacturing relevant ranges. Then, we characterize microstructural quality to determine the governing material – process – structure relationships. Finally, we assess mechanical performance for different processing scenarios. Overall, the results provide insights into the viability of compression molding in-process waste in the form of randomly oriented chips, highlight similarities and differences to traditional compression molding materials and processes, and identify opportunities for further optimization.
Methodology

Material

Two commercial thermosetting prepregs designed for out-of-autoclave (OoA) VBO cure and featuring a toughened epoxy matrix (CYCOM® 5320, Cytec Industries) were selected. The first prepreg featured a unidirectional (UD) carbon fiber bed (T40/800B) with a fabric areal weight of 145 g/m² and a total resin content of 33% by weight. The second prepreg consists of a five-harness satin (5HS) woven fabric (T650-35), with a fabric areal weight of 370 g/m² and a resin weight content of 37%. Upon receipt, both rolls were stored in freezers at −18 °C. The 5320 resin is designed for VBO processing in an oven at 93 and 121 °C. Vacuum bag pressures below 5 kPa are typically required to ensure air removal and compaction. The prepreg out-life of the 5320 resin is reportedly 21 days. VBO prepregs within the 5320 family exhibit a partially impregnated microstructure, with dry areas intended to allow extraction of entrapped air and volatiles under vacuum in the initial stages of processing.25 The dry areas are typically absent in conventional prepregs. The presence of dry areas increases the “bulk factor,” or ratio of initial volume to final volume, of the prepreg. The effect of these characteristics on chip-based manufacturing was not specifically addressed in this study, but should be noted.

Manufacturing

Chip production

Each prepreg roll was first removed from the freezer and allowed to thaw to room temperature for 8–12 h. Once cut, the chips were stored in sealed polyethylene bags in a freezer until future use, or first exposed to ambient temperature out-time for 14 or 28 days for conditioning. Note that while the unconditioned samples are designated “fresh” in later discussion, the prepreg rolls are believed to have a baseline out-time of 3–5 days. Chip lengths of 6.35, 12.7, and 25.4 mm were selected, while the chip width was maintained constant at 6.35 mm. Dimensions were controlled within a 5% tolerance.

Closed cavity compression molding

A lab-scale closed cavity aluminum mold was used to investigate the effect of key material and process factors in a controlled and instrumented setting. The mold features a two-part “piston” with a circular cavity of 76.2 mm diameter and a matching cylindrical piston. The cavity-piston gap is ~0.25 mm and is sealed using an O-ring. The mold is designed to limit resin seepage from the sample. Figure 1(a) depicts the piston mold assembly, and Fig. 1(b) shows prepreg chip panels produced using closed molding.

To produce molded panels, the internal mold surfaces were first treated with a liquid release agent (Henkel Frekote 770-NC) to avoid bonding to the prepreg. The prepreg chips were distributed in the cavity in a 2-D random in-plane orientation (care was taken to avoid out-of-plane orientations), and the piston was inserted into the cavity. The assembly was then placed into the environmental chamber of a load frame (Instron 5567). The load frame was used to control the load applied by the piston while measuring the crosshead displacement. The mold was heated to the desired cure temperature using the environmental chamber. No vacuum was used during compression molding, and the convective heating and cooling rates were measured but not directly controlled.

Figure 1. (a) Perspective sketch of closed cavity mold, with base plate (1), sample cell (2), piston (3), and sealing O-ring (4). (b) Prepreg chips sample compressed by closed cavity molding method
Scale-up molding

Scale-up studies were conducted using a hydraulic hot press and a larger two-part flat square mold (203.2 mm × 203.2 mm). During these experiments, we compared the microstructure and mechanical properties of chip-based parts with the baseline of a continuous fiber composite laminate produced using the same materials. Based on the results of the initial investigation of processing parameters, we considered two extreme cases for comparison with reference continuous fiber eight-ply laminates with a quasi-isotropic layup [0/90/+45/−45/−45/+45/90/0]. The "high-porosity" sample was comprised of chips consolidated under low pressure in the hot press (0.93 MPa, 5HS, 25.4 mm long chips, fresh prepreg and 121 °C cure for 2 h), equivalent in mass to an 8-ply panel. At the other extreme, the "low-porosity" sample consisted of chips consolidated under high pressure in the hot press (5.60 MPa, 5HS, 6.35 mm (0.25")

Test matrix

Table 1 shows the controlled variables used within the closed cavity compression mold. Four load levels were used to determine the effect of applied pressure. Two material quantities revealed "mass scale" effects. Two dwell temperatures were used to study the influence of the thermally controlled cure kinetics. Three chip lengths and two fiber bed architectures were chosen to clarify the influence of product form on consolidation. Finally, three out-times were selected to determine if prepreg out-time affected consolidation quality, and the specific material and process parameters are listed in Table 2. A baseline condition consisting of intermediate factor levels (17 kN load, 10.8 g mass, 121 °C temperature, 12.7 mm chip length and "fresh" 5320/5HS prepreg) was chosen as a standard reference to make it more clear to readers.

| Sample # | Force applied (N) | Mass (g) | Dwell temp/time (°C/hours) | Chip length (mm) | Fabric type (Cycom 5320) | Out-time |
|----------|------------------|---------|-----------------------------|-----------------|--------------------------|---------|
| 1        | 17,000           | 10.8    | 121/2                       | 12.7            | SHS                      | Fresh   |
| 2        | 4250             | 10.8    | 121/2                       | 12.7            | SHS                      | Fresh   |
| 3        | 8500             | 10.8    | 121/2                       | 12.7            | SHS                      | Fresh   |
| 4        | 25,500           | 10.8    | 121/2                       | 6.35            | SHS                      | Fresh   |
| 5        | 17,000           | 10.8    | 121/2                       | 25.4 mm (1")    | Cycom 5320 SHS           | Fresh   |
| 6        | 4250             | 10.8    | 121/2                       | 25.4            | Cycom 5320 UD            | 14 days |
| 7        | 8500             | 10.8    | 121/2                       | 25.4            | Cycom 5320 SHS           | Fresh   |
| 8        | 25,500           | 10.8    | 121/2                       | 25.4            | Cycom 5320 UD            | 14 days |

Note: Bold fonts represent a baseline condition consisting of intermediate factor levels (17 kN load, 10.8 g mass, 121 °C temperature, 12.7 mm chip length and "fresh" 5320/5HS prepreg) was chosen as a standard reference to make it more clear to readers.

Table 1. Manufacturing parameters for panels from prepreg scrap.

| Force applied (process pressure) | Mass  | Dwell temp. | Chip length | Fabric type | Out-time |
|---------------------------------|-------|-------------|-------------|-------------|---------|
| 4250 N (0.93 MPa)               | 10.8 g (4-ply equivalent) | 121 °C (2 h) | 25.4 mm (1") | Cycom 5320 SHS | Fresh   |
| 8500 N (1.87 MPa)               | 21.6 g (8-ply equivalent) | 93 °C (8 h)  | 12.7 mm (0.5") | Cycom 5320 UD | 28 days |
| 17,000 N (3.73 MPa)             |       |             |             |             |         |
| 25,500 N (5.60 MPa)             |       |             |             |             |         |

Table 2. Test matrix for specific material and process parameters used for closed cavity compression molding test in this study. The bold variables represent the baseline condition as reference for the later mechanical results.
6.35 mm long chips, 14 days of out-time prepreg, and under 93 °C cure for 8 h), of similar mass.

Three reference panels were fabricated for comparison. The first reference panel was an eight-ply laminate with no out-time manufactured by the VBO method and cured at 121 °C for two hours. The second consisted of the same laminate produced by hot press forming, under the same pressure and temperature as the “high-porosity case” chip panel. The third consisted of continuous plies exposed to 14 days of out-time, processed under the same pressure and temperature as the low-porosity case panel. As a result, the range of performance of the chip-based panels was compared to continuous fiber laminates manufactured under similar temperature and pressure conditions.

**Part quality and performance**

**Thickness**

After manufacturing, the thickness \( h \) of each sample was measured in five locations to assess dimensional uniformity and to estimate the fiber volume fraction \( (V_f) \).

**Resin bleed**

The mass of the samples cured in the piston mold \( (m_f) \) was compared to the initial mass of the prepreg \( (m_i) \). Assuming no fibers were removed during processing, the amount of resin bleed \( \Delta V_r \) was quantified per Equation (1), where \( W_r \) is the fractional resin weight content.

\[
\Delta V_r = \frac{m_i - m_f}{m_i} \cdot W_r
\]  

(1)

**Porosity**

Void contents were measured using microscopy of polished samples. Each sample was 50 mm long, cut from the center of the parts. Sample cross sections were polished by abrasives on a grinder-polisher (Struers® LaboPol-2) at grits of 150, 240, 400, 600, 1200, and 2400. Images of polished sections were acquired using a digital microscope (Keyence® VHX-600E) at 100×. A total of 40 images were necessary for a full picture of each cross section. Images were merged and processed using image processing software. Two cross sections were used to evaluate porosity for each sample.

For void content analysis, the images were converted to gray scale, and voids were manually selected and filled to distinguish from solid phases. Then, image analysis software (ImageJ®) was used to “threshold” each image into a binary map of void (black) and solid (white) pixels, and to analyze the resulting areal void ratio.

**Mechanical properties**

Mechanical test samples were prepared from both chip and continuous ply product forms using a hot press and a two-part flat square mold (215.9 mm × 215.9 mm). To assess panel quality, the microstructure and amount of resin bleed were analyzed before mechanical testing.

Specimens 203.2 mm long and 25.4 mm wide were cut and loaded to failure in tension using a load frame (Instron 5567) according to the ASTM D3039 standard. The loading rate was 2 mm/min to minimize strain rate effects. Eight coupons were prepared and tested per manufactured panel, and only valid gage section failure cases were chosen. The gauge length was set to 40 mm for every coupon.

Compression tests were carried out using a combined loading compression (CLC) fixture (Wyoming Test Fixtures, Salt Lake City, UT) according to ASTM D6641. Combined loading compressive strength is widely used for material specifications and design purposes. Eight test coupons (139.7 mm long by 12.7 mm wide) with a gauge length of 12.7 mm were tested from each panel. Coupons were aligned to the text fixture to prevent premature end crushing and limit bending or buckling during testing. The loading rate was 1.3 mm/min to failure.

**Results and discussion**

**Thickness**

From Equation (1), assuming no fibers lost during processing, the part thickness \( (h) \) will be inversely proportional to the fiber volume fraction \( (V_f) \). The \( V_f \) was estimated from the sample mass \( (m_f) \) and volume \( (obtained from the thickness h \times surface area A) \), and the carbon fiber density \( (\rho) \).

\[
V_f = \frac{m_f(1 - W_r)}{\rho_f A h}
\]  

(2)

Figure 2 shows the relationship between fiber volume fraction and thickness for various sample classes. For both materials, the fiber volume fraction decreased quasi-linearly with increasing thickness. Moreover, UD
prepreg samples exhibited fiber volume fractions 5–10% greater than 5HS samples. Finally, the $V_f$ increased by up to 17% in 5HS fabric and 22% in UD fabric as the compression force was increased from 4.25 to 25.5 kN, demonstrating that higher processing pressures can increase consolidation. Also, the figure shows that there is a reduction in $V_f$ from the fresh condition to the 14-day aged and to the 28-day aged materials, and a reduction in resin bleed. The figure also shows that for relatively short out-times, the matrix loses tack, which makes the chips easier to lay down flat and, consequently, improves manufacturability. However, exceeding out-times will cause partial curing of the matrix before manufacturing, which could lead to further reductions in resin bleed and poor adhesion between the chips.

**Microstructural quality**

Figure 3(a) depicts the common locations of voids formed during closed compression molding. Due to the morphology of the prepreg chips and the associated microstructure, most voids were located at chip edges and within the resin-rich areas formed by chip overlaps. The chip edges are likely locations for air bubbles, particularly if air is initially entrapped within the dry areas of the VBO prepreg chips. The resin-rich zones formed by chip overlaps often serve as air entrapment regions during layup (or chip placement). Figure 3(b) shows polished microstructural cross sections of the larger panels fabricated for mechanical testing, demonstrating that these parts exhibited the same defect types.

In this study, the long axis of the chips was oriented along the fiber direction of the original prepreg. The longer chip dimension was therefore used to maximize the effective length of the fibers and potentially improve mechanical properties. In practice, chips cut from in-process waste in the form of prepreg “skeletons” are likely to have different lengths, depending on the skeleton geometry. Understanding the effects of chip length is both important and useful.

Figures 4(a) and 5(a) show the effects of consolidation load and chip length on void content and resin bleed. For the 25.4 and 12.7 mm chips, increasing the consolidation force from 4.25 to 17 kN reduced the void content from about 3.5% to approximately 2%. However, an increase

![Figure 3](image-url)

**Figure 3.** (a) Common void locations in fabricated parts: edges of the chips (red rectangle) or resin-rich areas (blue rectangle). (b) Polished sections showing void contents under (i) the high-porosity case, (ii) baseline case, and (iii) the low-porosity case.
Furthermore, for a given load, bleed percentages increased with chip size, indicating that smaller chips formed a more permeable porous medium, and provided less resistance to resin flow. Note that the nonzero bleed percentages show that the lab-scale fixture used to compact the samples limited, but did not fully eliminate, resin loss, and that during processing, the resin did not carry the entirety of the applied load.

Figures 4(b) and 5(b) illustrate the influence of dwell temperature on part quality. The lower dwell temperature (93 °C) reduced the void content in all cases and from 17 to 25.5 kN did not have a noticeable effect in void content but multiple smaller voids discovered in these higher force applied cases. Conversely, for 6.35 mm chips, the void content decreased to approximately 0.9% at 17 kN and to 0.4% at 25.5 kN. These observations indicate that shorter chips facilitated consolidation, due to a more uniform initial distribution and more effective re-arrangement and nesting once pressure was applied. The resin bleeding (or mass loss) percentages increased from 10–20% at low loads to 25–35% at high loads, indicating that higher resin pressures induced more bleed. Furthermore, for a given load, bleed percentages increased with chip size, indicating that smaller chips formed a more permeable porous medium, and provided less resistance to resin flow. Note that the nonzero bleed percentages show that the lab-scale fixture used to compact the samples limited, but did not fully eliminate, resin loss, and that during processing, the resin did not carry the entirety of the applied load.

Figures 4(b) and 5(b) illustrate the influence of dwell temperature on part quality. The lower dwell temperature (93 °C) reduced the void content in all cases and from 17 to 25.5 kN did not have a noticeable effect in void content but multiple smaller voids discovered in these higher force applied cases. Conversely, for 6.35 mm chips, the void content decreased to approximately 0.9% at 17 kN and to 0.4% at 25.5 kN. These observations indicate that shorter chips facilitated consolidation, due to a more uniform initial distribution and more effective re-arrangement and nesting once pressure was applied. The resin bleeding (or mass loss) percentages increased from 10–20% at low loads to 25–35% at high loads, indicating that higher resin pressures induced more bleed. Furthermore, for a given load, bleed percentages increased with chip size, indicating that smaller chips formed a more permeable porous medium, and provided less resistance to resin flow. Note that the nonzero bleed percentages show that the lab-scale fixture used to compact the samples limited, but did not fully eliminate, resin loss, and that during processing, the resin did not carry the entirety of the applied load.

Figures 4(b) and 5(b) illustrate the influence of dwell temperature on part quality. The lower dwell temperature (93 °C) reduced the void content in all cases and
had a more significant effect at higher consolidation pressures. These relationships indicate that in prepreg chip parts, voids are primarily gas-induced rather than flow-induced. Flow-induced voids are generally mitigated by higher temperatures, which reduce the resin viscosity and increase flow rates. Conversely, gas-induced voids are generally suppressed by lower temperatures and higher pressures, as demonstrated in Figs. 4(b) and 5(b). The resin bleed increased with temperature (121 °C), but stabilized at 25.5 kN, indicating that there is a limit to the total amount of resin outflow, which is likely to be dictated by the nonlinear influence of increasing fiber volume fraction and decreasing permeability.

The influence of fiber bed architecture is shown in Figs. 4(c) and 5(c). The 5320/UD panels exhibited markedly lower void contents than their 5320/5HS counterparts for all consolidation loads and near-zero porosity.
at 25.5 kN. Two factors may explain this finding. First, the unidirectional chips are relatively flat due to fiber alignment and the absence of tow overlaps, and are likely to re-arrange and nest during compaction. Second, previous work has shown that the UD panels are less likely to entrap air than textiles.\textsuperscript{28} Since air is more likely to be entrapped between chips rather than at surface undulations of each chip, this effect may reduce the likelihood of entrapment. The resin bleed is less in the 5320/UD parts than in the 5320/5HS due to both the lower initial resin content and the comparably low fiber bed permeability of unidirectional reinforcements.

The influence of out-time is shown in Figs. 4(d) and 5(d). The results indicate that both 14-day and 28-day conditioning decreased the void content. The out-time effect on degree of cure and tack of the 5HS material has been investigated previously.\textsuperscript{29} The reduction in prepreg tack and increase in resin viscosity induced by out-time facilitated a more uniform initial chip distribution and enabled chip re-arrangement and nesting during consolidation. This explanation is supported by the resin bleed data, which shows that for the 14- and 28-day samples, resin loss did not increase with increased compaction loading, and that the 28-day parts lost less than 5% resin overall. Moreover, the higher resin viscosity and limited resin outflow in aged samples may have increased the resin pressure during processing, relative to the fresh case, for a given applied compaction pressure, since pressure relaxation due to bleeding was reduced. Note that the 28-day samples exhibited higher void contents than the 14-day samples for all loadings, although this increase was within the measurement variability. This trend, if valid, indicates that at 28 days, out-time has a more important detrimental effect, with the viscous resin now unable to fully fill the dry areas within the partially impregnated microstructure of each prepreg chip.

Figures 4(e) and 5(e) illustrate the influence of sample mass on void content and resin bleed for panels produced from 5320/5HS chips. Eight-ply equivalent samples exhibited lower void content levels than four-ply equivalents, indicating that a minimum quantity of chips is required to eliminate gaps within the part and potential void formation sites. Furthermore, the results show that this phenomenon may interact with the consolidation force, since the reduction in void content between four-ply and eight-ply equivalents is more pronounced at higher pressing loads. The resin bleed was slightly

![Figure 6. (a) Tensile and (b) compressive properties of three reference panels and two select cases. Error bars refer to the standard deviation](image-url)
greater for the heavier parts, suggesting that the lighter panels may also suffer from resin starvation.

These results demonstrate that the microstructural quality of compression-molded panels made from randomly oriented chips can be affected by multiple material and process factors. Higher applied compaction pressures typically result in reduced porosity. However, this improvement often is limited or compounded by other factors. The capacity of individual chips to distribute and nest under applied pressure, enabled by shorter chip lengths and/or reduced tack, is critical and can yield marked reductions in void content, particularly at high pressures. Lower cure temperatures limit the driving forces for gas-induced void growth, at the cost of longer cure times and higher resin viscosity. Conversely, insufficient material can lead to non-uniform chip distribution, leaving gaps that entrap air and result in voids. Finally, relatively flat, uniform prepreg chips facilitate chip reorganization and consolidation, reducing air entrapment and resulting in low porosity.

These insights can be compared and contrasted to the knowledge base on compression molding. The ability to improve consolidation quality by increasing the applied pressure or ensuring full coverage of the mold by material is aligned with common practices for SMCs, GMTs, and LFTs. Conversely, the effects of chip size, tack/out-time, and cure temperature emphasize that prepreg chips sourced from in-process waste can exhibit properties distinct from optimized SMC, GMT, and LFT feedstock. As a result, compression molding of chips requires a detailed understanding of the fiber bed characteristics and resin properties, and the development and pressure/temperature cycles that optimize the in-process behavior for closed molding.

**Mechanical properties**

In Fig. 3(b), cross sections for low-porosity and high-porosity panels were compared with the baseline condition. Voids affect the mechanical properties and durability of composites. The difference in void contents for the low-porosity case (0.29 ± 0.03%) and the high-porosity case (4.21 ± 0.59%) is nearly a factor of 15×.

Figure 6 shows (a) tensile and (b) compressive properties for the high-porosity case and the low-porosity case, the two compression molding scenarios identified in this study, and reference panels of continuous fiber laminates fabricated by VBO cure, and by compression molding (from fresh and out-timed material). The results for chip-based parts showed tensile strength retention of 44 and 25% in the low-porosity and high-porosity cases, respectively, relative to the continuous fiber references. In contrast, the tensile modulus retention was ~70% in both cases. Compressive strength/modulus measurements exhibited similar trends – strength retention of 50–60% and modulus retention of 70–80%.

The specific properties of the chip panels compare favorably with those of typical aluminum alloys. For example, approximate calculations yield specific strength values of 125 MPa for chip panels vs. 110 MPa for Al alloys (based on strength/specific gravity for composites and Al, or 200 MPa/1.6 and 300 MPa/2.7, respectively). The specific modulus values were 28 for chip panels and 26 for Al alloys (based on 45 GPa/1.6 and 70 GPa/2.7). Aluminum data corresponds to 6061 Al-T6, a general-purpose Al alloy.

For the continuous fiber reference panels, processing in a heated press after 14 days out-time marginally increased tensile strength and maintained compressive strength and moduli at the same levels as the VBO reference. Thus, we assert that the manufacturing process of the reference panels has a negligible effect on mechanical properties. In contrast, the high-porosity case chip-based scenario exhibits a large decrease in strength in comparison with the low-porosity case, but a relatively modest decrease in stiffness. The significant strength decrease derives from the fact that the discontinuous reinforcement and short fiber lengths require the resin to carry load between chips – thus, matrix strength is limiting for such materials. Note that the difference is within statistical variability for the tensile data. However, in both cases, the tensile and compressive modulus values are maintained at levels similar to panels made from virgin prepreg by VBO processing. Modulus values were less affected by higher defect levels because, at low strains, the stiffness of composite parts is largely governed by fiber loading, orientation, and length, and by the fiber–matrix interface, rather than by the presence of defects within the matrix.

**Conclusions**

We have investigated a strategy for direct reuse of prepreg scrap generated during production, using compression molding of randomly oriented prepreg chips. A parametric study was performed using small samples to determine the effect of key material and processing factors on part thickness and microstructural characteristics. Then, mechanical testing was performed for the ideal and non-ideal cases.

Several key manufacturing considerations emerge from the results presented here. First, parts with high microstructural quality (low-porosity, high fiber volume fractions) can be manufactured from prepreg chips if the materials and molding conditions are judiciously selected. Second, material and process parameter selection is critical, with void contents for parts made from the same material (5320/5HS) varying between 0.4 and 3.50%. High-quality parts resulted from high consolidation pressures, low molding temperatures, and prepreg properties favoring easy chip distribution, re-arrangement, and nesting (i.e. short, unidirectional, tack-free
chips). Furthermore, the low-porosity and high-porosity case scenarios identified during lab-scale testing were shown to influence the mechanical performance of prepreg chip panels. The highest specific mechanical properties obtained were comparable to those of conventional age-hardened aluminum alloys, a necessary requirement for future substitution. These guidelines form an initial basis for further optimization of the manufacturing process.

Further research and development steps are required before the viability of reusing prepreg waste using this method is confirmed. The manufacture of larger and more complex structures must be investigated to identify applications best suited to the materials and process, and to further optimize processes. Furthermore, the economic and environmental life cycle characteristics of this material reuse approach must be assessed to clarify the benefits relative to competing reuse and recycling methods, such as fiber reclamation. Note that while mechanical properties of chip panels were compared to a continuous prepreg baseline, it may be more appropriate to compare the specific properties of chip panels with those of monolithic aluminum alloys. Indeed, it is more realistic to speculate that chip panels may one day be considered as viable substitutions for such alloys. Finally, the logistical challenges associated with collecting, transporting, and storing prepreg waste, as well as with developing market supply and demand, must be addressed.

This initial study demonstrates that parts with high microstructural quality can be produced from prepreg waste without matrix removal, and the practice of prepreg scrap reuse offers additional avenues for improving materials use and manufacturing efficiency. The reuse approach returns both fibers and matrix to service. In relative terms, fiber reclamation processes claim only the fibers and do not recycle or recover the matrix. This issue becomes larger in second generation recycling processes which begin with composites that typically have much higher matrix contents – 60–75% – in which the polymer matrix is the primary component. At this point, commercial applications for prepreg scrap must be developed to further refine and scale up the processes of collection, cutting, sorting, distributing, and consolidation.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by the National Science Foundation (NSF) G8 Initiative “Material Efficiency: A First Step towards Sustainable Manufacturing of Composite Materials” [grant number CMMI-1229011], Cytec Industries and Northrop Grumman donated the prepreg materials used in this study.

ORCID

M.-S. Wu http://orcid.org/0000-0001-8820-4734

References

1. W. G. Roeseler, B. Sarh and M. U. Kismarton: ‘Composite structures: the first 100 years’, Presented at the 16th International Conference on Composite Materials, July 8–13, 2007.
2. K. A. Iyer, J. Lechanski and J. M. Torkelson: ‘Green polypropylene/waste paper composites with superior modulus and crystallization behavior: optimizing specific energy in solid-state shear pulverization for filler size reduction and dispersion’, Compos. Part A Appl. Sci. Manuf., 2016, 83, 47–55.
3. C. Kazmierski: ‘Growth opportunities in global composites industry 2015–2020’, Tech. Rep., Lucintel, Irving, TX, 2015.
4. T. Roberts: ‘Rapid growth forecast for carbon fibre market’, Reinf. Plast., 2007, 51, 10–13.
5. J. F. Unser and T. Staley: ‘Advanced composites recycling’, 41st International SAMPE Symposium, March 1996.
6. Composite Industry Survey: ‘Report prepared by McDonnell Douglas aerospace’, 27 November 1991.
7. S. Pimenta and S. T. Pinho: ‘Recycling carbon fibres reinforced polymers for structural applications: Technology review and market outlook’, Waste Manag., 2011, 31, 378–392.
8. G. Oliveux, L. O. Dandy and G. A. Leeke: ‘Degradation of a model epoxy resin by solvolysis routes’, Prog. Mater. Sci., 2015, 72, 96–103.
9. K. Wood: ‘Carbon fiber reclamation: going commercial’, High perform. Compos., 2010, 3, 1–2.
10. P. Feraboli, T. Cleveland, M. Ciccu, P. Stickler and L. DeOto: ‘Defect and damage analysis of advanced discontinuous carbon/epoxy composite materials’, Compos. Part A Appl. Sci. Manuf., 2010, 41, 888–901.
11. P. Feraboli, E. Petiso, F. Deleo, T. Cleveland and P. B. Stickler: ‘Characterization of prepreg-based discontinuous carbon fiber/epoxy systems’, J. Reinf. Plast. Compos., 2009, 28, 1191–1214.
12. P. Feraboli, E. Petiso, T. Cleveland and P. B. Stickler: ‘Modulus measurement for prepreg-based discontinuous carbon fiber/epoxy systems’, J. Compos. Mater., 2009, 43, 1947–1965.
13. P. Feraboli, E. Petiso, T. Cleveland, P. B. Stickler and J. C. Halpin: ‘Notched behavior of prepreg-based discontinuous carbon fiber/epoxy systems’, Compos. Part A Appl. Sci. Manuf., 2009, 40, 289–299.
14. N. Egueumann, L. Giger, M. Roux, C. Dransfeld, F. Thiebaud and D. Perreux: ‘Compression moulding of complex parts for the aerospace with discontinuous novel and recycled thermoplastic composite materials’, Proc. 19th Int. Conf. Compos. Mater., 2013.
15. M. Seleznева, G.-P. Pichcr-Martel, B. Landry, B. Trudel-Boucher, S. Roy L. Khoun, M. Hovjati, L. Lessard, P. Hubert: ‘Compression moulding of discontinuous-fibre carbon/PEEK composites: Study of mechanical properties’, Proc. SAMPE 2012 Conf., Baltimore, MD, Society for the Advancement of Material and Process Engineering, 2012.
16. M. Seleznева and L. Lessard: ‘Characterization of mechanical properties of randomly oriented strand thermoplastic composites’, J. Compos. Mater., 2015, 50, 2833–2851.
17. M. Selezneva, S. Meldrum, S. Roy, L. Lessard and A. Yousefpour: ‘Modelling of mechanical properties of randomly oriented strands thermoplastic composites’, J. Compos. Mater., 2016, 51, 831–845.

18. D. LeBlanc, B. Landry, A. Levy, P. Hubert S. Roy, A. Yousefpour, et al: ‘Study of processing conditions on the forming of ribbed features using randomly oriented strands thermoplastic composites’, Proc. 70th Annu. Forum Am. Helicopter Soc., Montreal, 2014.

19. A. Levy and P. Hubert: ‘Interstrand void content evolution in compression moulding of randomly oriented strands (ROS) of thermoplastic composites’, Compos. Part A Appl. Sci. Manuf., 2015, 70, 121–131.

20. B. Landry and P. Hubert: ‘Experimental study of defect formation during processing of randomly-oriented strand carbon/PEEK composites’, Compos. Part A Appl. Sci. Manuf., 2015, 77, 301–309.

21. M. Selezneva, S. Roy, L. Lessard and A. Yousefpour: ‘Analytical model for prediction of strength and fracture paths characteristic to randomly oriented strand (ROS) composites’, Compos. Part A Appl. Sci. Manuf., 2016, 96, 103–111.

22. Y. Wan and J. Takahashi: ‘Tensile and compressive properties of chopped carbon fiber tapes reinforced thermoplastics with different fiber lengths and molding pressures’, Compos. Part A Appl. Sci. Manuf., 2016, 87, 271–281.

23. M. Revellino, L. Saggese, E. Gaiero, K. Anthony and Z. Carl: ‘Compression molding of SMCs’, in ‘Comprehensive composite materials’, (ed. A. Kelly and C. Zweben), 763–805; 2000, Amsterdam, Elsevier.

24. M. D. Wakeman and C. D. Rudd: ‘Compression molding of thermoplastic composites’, in ‘Comprehensive composite materials’ (eds A. Kelly and C. Zweben), 915–963; 2000, Amsterdam, Elsevier.

25. C.-H. Park and W. I. Lee: ‘Compression molding in polymer matrix composites’, in ‘Manufacturing techniques for polymer matrix composites (PMCs)’, (eds S. G. Advani and K.-T. Hsiao), 512; 2012, Amsterdam, Elsevier Science.

26. T. Centea, J. Kratz, and P. Hubert: ‘The effect of out-time and cure cycle on the consolidation of out-of-autoclave prepregs’, Proc. 11th Int. Conf. Flow Processes Compos. Mater, Auckland, New Zealand, 2012.

27. L. K. Grunenfelder and S. R. Nutt: ‘Void formation in composite prepregs – Effect of dissolved moisture’, Compos. Sci. Technol., 2010, 70, 2304–2309.

28. T. Centea and P. Hubert: ‘Out-of-autoclave prepreg consolidation under deficient pressure conditions’, J. Compos. Mater., 2013, 48, 2033–2045.

29. L. K. Grunenfelder and S. R. Nutt: ‘Out time effects on VBO prepreg and laminate properties’, SAMPE J., 2011, 47, 6–12.

30. Metals handbook: ‘Vol.2 – properties and selection: Nonferrous alloys and special-purpose materials’, 10th edn, 1990, Geauga, OH, ASM International.

31. J. M. Holt, C. Gibson, and C. Y. Ho: ‘Structural alloys handbook’, 1996 edn, 1996, West Lafayette, IN, CINDAS/ Purdue University, ©1997.

32. H. E. Boyer and T. L. Gall: ‘Metals handbook’, 1985, Materials Park, OH, American Society for Metals.