REUSE AND UPCYCLING OF THERMOSET PREPREG SCRAP: CASE STUDY WITH OUT-OF-AUTOCLAVE CARBON FIBER/EPOXY PREPREG

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ABSTRACT: A large amount of uncured thermoset prepreg scrap is generated during manufacturing, including both ply cutter trim waste and out-of-spec material. While techniques to recycle end-of-life cured composite waste and reclaim carbon fiber are well established and commercialized, there is little effort made presently towards reusing uncured scrap prepreg. Here we present a viable and scalable technique to process scrap prepreg into intermediate forms that can be readily manufactured into commercial end-products. Using out-of-autoclave (OoA) carbon fiber / epoxy prepreg as an example, we report the mechanical properties, microstructure, and performance of composite laminates fabricated with scrap prepreg under various processing conditions. Demonstrator parts manufactured with scrap prepreg are also presented. Hybrid composite foams as a low-cost engineering material that is tough, strong, and fire retardant.

Key words: Prepreg, Reuse, Carbon Fiber, Thermoset, Recycling, Mechanical Properties, Compression Molding

1. INTRODUCTION

There has been a significant increase in the global demand for carbon fiber (CF) and carbon fiber composites from several industries, notably aerospace and automotive. The global prepreg industry
is expected to grow to an estimated USD 6.1 billion by 2019 [1], while the global demand for carbon fiber composites is projected to reach USD 20.1 billion by 2018 with an annual growth rate of 6.4% [2]. Newer aircraft from Boeing (e.g. 787, 777X) and Airbus (e.g. A380, A350) exemplify the increasing use of composites in aircraft structures. Boeing’s market outlook (2015-2035) projects a doubling of the world aircraft fleet size from approximately 23k in 2015 to 45k in 2035, valued around USD 5.9 trillion. Within the fleet, single aisle aircraft are projected to grow from 15k in 2015 to 32k in 2035. In the automotive industry, increased mileage requirements for US passenger cars of 23.1 km/l by 2025 has led automotive manufacturers to increasingly explore using carbon fiber to help reduce vehicular weight. Similarly, the sporting and recreation industry is also increasingly using carbon and other polymeric fibers (e.g. aramids) to produce lighter and stronger sports equipment.

Thermoset composite prepreg waste can be broadly classified into uncured waste generated during the manufacturing operation and cured waste from end-of-life parts. Techniques to handle these material streams can therefore be classified as “reuse” and “recycle,” respectively. Recycling efforts seek to reclaim carbon fibers from cured waste by removing the matrix through various techniques such as burn-off (pyrolysis), matrix digestion (acids and supercritical fluids), or depolymerization. Individual reclaimed carbon fibers demonstrate excellent moduli and strength retention properties compared to virgin fibers. However, they lose their continuous and aligned configuration and instead often appear as tangled arrays of fibers that must be reprocessed (realigned via carding, and often resized) before they can be processed into new CFRP parts. More information on composites recycling technologies, their impact, and market outlook can be obtained from Refs. [3-7]. There are only a handful of companies globally that have commercialized recycling processes, such as
Adherent Technologies and Carbon Conversions (formerly MIT-RCF) in the US, and ELG Carbon Fibre in the EU. Carbon fiber reclamation through pyrolysis and solvolysis continues to be an active research topic in academia as well as in organizations such as MAI Carbon Recycling (Germany), Siemens Corporate Technology (Germany), and Hitachi Chemical (Japan), to name a few.

Reuse efforts, on the other hand, seek to use both the fiber and the uncured matrix from scrap prepreg in their original configuration towards the manufacture of new CFRP parts. Sources of scrap prepreg include ply cutter waste, end-of-roll, and out-of-spec material such as those beyond the specified out-life or storage life. In the aircraft industry, for example, the task of recertifying out-of-spec prepreg rolls is cost prohibitive. The amount of scrap generated during ply cutting operations depends on the size and shape of the prepreg cut-outs as well as the efficiency of the nesting routines.

According to varied sources [8-10], the buy-to-fly ratio can vary from 1.2:1 to as high as 3.5:1, with manufacturing waste between 6-19%, uncured prepreg forming 56-70% of the waste stream, ply cutter scrap between 25-50%, and post-processing (trimming, machining) waste between 2-40%. These reported ranges are merely exemplary. In reality, the precise estimates of waste vary greatly, depending on factors such as the type of industry (e.g. aerospace, automotive, recreation), type of product (e.g. high-volume production line or small-order customized parts), and the level of automation and technology utilized. Unfortunately there is little available published data that has consistently monitored thermoset prepreg waste classified by industry and tracked over time as manufacturing efficiency and technology improves. For example, Ref. [11] indicates 66% of the waste generated at the manufacturing source was in the form of unused prepreg material, although this figure was based on industry surveys [12,13] over a decade old. Prepreg scrap is generally sent to landfills, posing both an environmental challenge as well as a lost commercial opportunity to recover some of the original material cost.
The amount of energy required to reclaim carbon fiber is significantly less than that used to manufacture new carbon fiber [6], leading to a positive energy and carbon footprint. Some reclamation processes however can result in toxic by-products. Reuse processes directly utilize the original material form, resulting in a comparatively negligible carbon footprint and absence of toxic by-products. The issue of reusing scrap prepreg is time-sensitive and must be handled “here-and-now,” as it is continuously generated during the manufacturing process. In contrast, recycling efforts that target end-of-life parts could be implemented after a decade for an automobile and a service life of a few dozen years for an aircraft. There is an estimated 13M kg of uncured scrap carbon fiber prepreg landfilled annually in the US. This is clearly a growing concern that needs to be addressed. However to provide context to the reader, note that by comparison, the volumes of scrap thermoset prepreg are outweighed by the volumes of cured carbon fiber composite waste, in turn far outweighed by the volumes of cured glass fiber composite waste primarily from the boating industry. The issue of reusing scrap prepreg has gained traction in the European Union, where legislation limits the extent of landfilling and mandates minimum levels of reuse and recycling with strict penalties and fines being levied [6]. In contrast, less strict environmental regulations in the US have yielded little progress in composites recycling, and the reuse of prepreg scrap is largely ignored. While some companies may embrace green and sustainable practices and closely incorporate them into their corporate philosophy and strategy, others may consider reuse and recycling as a distraction from their core competency, especially if their bottom line is not affected. Some estimates of waste disposal fees in the US are as low as $0.09 per kg while other estimates specifically for prepreg waste disposal fees range between $1.7-6.0 per kg (obtained from two undisclosed aerospace and recycling companies). These costs can increase if the waste is considered hazardous and requires special handling. Note that these estimates are merely exemplary of a few cases and that variability
in waste disposal fees can be expected between various vendors. Depending on the type of industry and volumes of scrap prepreg generated, waste disposal fees may either have little effect on a company’s finances, or may prove to be a significant-enough driver of their cost. However, it is not yet cost prohibitive in the US to simply landfill all waste prepreg material, which is the de-facto practice at present. Nonetheless, there is increasing interest in the US in composite reuse and recycling efforts, as there is a general consensus in the industry that environmental regulations will emerge in the US along the lines of the European Union, and companies are slowly positioning themselves for such legislation. The potential savings from a carbon tax, avoidance of fines and penalties for non-compliance, and the possibility of additional revenue and job creation from reuse and recycling efforts make a compelling argument for re-use and recycling. For example, the Composites Recycling Technology Center (CRTC) was recently established in the state of Washington and backed with state and federal funding. The primary mission focuses on the reuse of scrap prepreg, and ultimately aims at spurring economic activity and jobs locally.

The literature dealing with basic research into and commercialization of prepreg scrap reuse and upcycling is sparse. Several of these works, perhaps ahead of their time, are over a decade old [12-15]. Unser [9] explored the direct reuse of prepreg scrap, where scrap pieces of varying sizes and shapes were either butt spliced or overlap spliced to form continuous panels. Also, a patent by Imparato et al. [10] involved randomly placing predetermined sized fragments of prepreg scrap between strips of backing and protective material and then compacting the assembly to form a strip of constant areal weight and thickness. A patent application by Evans [16] involved milling thermoset scrap prepreg at sub-zero temperatures into a dough molding compound (DMC) comprised of filament lengths between 1-2 mm. Ward and Potter recommended various techniques
to reduce the volume of prepreg scrap generated [8,17,18]. Additional information and references on composite reuse and recycling can be obtained from composite handbooks [11,19].

An unusual prepreg material form, although comprised of virgin (fresh) prepreg and not scrap, is created by cutting plies of unidirectional prepreg into rectangular chips that can be compression molded into composite parts. This chopped prepreg molding compound (CPMC) was studied by Fudge [20]. For example, TenCate Advanced Composites (USA) sells a bulk molding compound (BMC) comprised of chopped unidirectional carbon fiber/epoxy prepreg. Hexcel (USA) also sells a molding compound comprised of unidirectional carbon fiber and a quick curing epoxy prepreg. The compound is provided in a roll form (HexMC®) comprised of randomly oriented chips of 50 mm x 8 mm in size. A similar prepreg product form with BMI resin (HexTool®) is used for tooling.

Here we describe a technique to reuse and upcycle scrap prepreg, starting with the raw material and culminating in a multiple demonstrator parts, including a hat stiffened panel and a carbon fiber prosthetic foot. Broadly, the process involves reducing scrap pieces into a chip-like form that is compression molded into the final part. In addition, the effects of material, form, and processing such as chip size, fiber orientation, cure cycle, and consolidation pressure on the mechanical properties, performance, and microstructure of scrap-based laminates is studied in detail using scrap derived from carbon fiber/epoxy prepreg (Cytec CYCOM® 5320 5HS). Additional information on our prepreg scrap reuse efforts appear elsewhere [21-23].

2. AN OVERVIEW OF THE REUSE AND UPCYCLING PROCESS

2.1. SOURCES OF SCRAP PREPREG
CF / epoxy prepregs contain partially cured (B-staged) epoxy resin and are stored in freezers to retard further curing. Commercial prepreg materials have a specified tack life, out-life, and freezer storage life which respectively defines the recommended maximum time frames within which a prepreg ply should be laid down, can be left out in the shop floor at room temperature (RT), and can be stored in a freezer, beyond which the material is no longer considered within specification. Scrap prepreg can consist of unused rolls that have simply exceeded these limits of storage life or out time. A second common form of prepreg scrap consists of irregular cutouts or “skeletons” left over from kitting or other ply cutter operations. Scrap prepreg is often mixed with other composite manufacturing waste such as the consumables (backing paper, breather, sealant tape, vacuum bag, etc.) in waste streams, and therefore must be separated prior to reuse. In some cases, prepreg scraps also must be separated and classified based on the underlying fiber and resin system, especially if they are incompatible to co-process, or doing so could lead to reduction in resulting part performance. Once separated, suppliers recommend that the scrap prepreg be re-frozen as quickly as possible to minimize further curing of the resin. The time frame for this re-freezing depends on several factors, such as the existing tack and out-time of the scrap prepreg, how often and how quickly the scrap is collected from the shop floor, whether the scrap processing facility is co-located with the source of scrap, or whether the scrap must be transported to it, and whether the volumes of generated scrap are too large to be processed in a timely manner, thereby necessitating freezer storage until it is ready to be processed. Scrap prepreg is typically in the form of various geometric shapes and sizes with the backing paper still intact [22]. Because of the high variability in scrap streams, this form of scrap prepreg cannot be readily or efficiently upcycled and must first be converted into an intermediate and more consistent form.
2.2. INTERMEDIATE SCRAP PREPREG PRODUCT FORMS

Figure (1) shows five possible intermediate forms that we have identified for potential use in upcycling operations. The loose chip form (LCF) consists of chopped scrap prepreg, where the scrap pieces have been cut into uniform rectangular chips. Contrary to virgin prepreg, it is actually preferable for the scrap prepreg and resulting LCF to have as little tack as possible to facilitate distribution and dispersal of chips. LCF is similar to bulk molding compound (BMC) consisting of chopped virgin prepreg (typically unidirectional), except that it is comprised of scrap prepreg of various fiber bed architectures (unidirectional, fabrics, etc. and combinations thereof). The cutting process can be fully automated, wherein a conveyor belt continually feeds incoming scrap pieces into a set of linear and rotary cutters that initially cut the pieces into linear strips and subsequently into chips of desired dimensions. The process can also be semi-automated or batch-processed, wherein a clicker die cutting press is used to cut pieces of scrap one at a time into chips. The size of the resulting chips is determined by the shape and pattern of the rule die used for the cutting. In both operations, the incoming scrap pieces may require orientation in some manner with respect to the orientation of the blades to maximize the fiber length in the resulting chips (this may be the case with unidirectional scrap prepreg). Such orientation may be done manually or using machine vision. The protective backing paper must be removed prior to cutting the prepreg into chips, and the chips must be stored frozen to prevent binding.
A second intermediate product form is the reused scrap roll form (RSRF), a continuous flat and smooth sheet comprised of LCF. RSRF is customizable in terms of composition, thickness, areal weight, and flexibility, and can be laid up and processed in the same manner as virgin prepreg, although it is most suitable to compression molding. The process to convert LCF into RSRF involves evenly distributing the chips between two plies of backing paper or release film, then pressing between heated platens under low temperature and high pressure. The low temperature is just sufficient to bond the chips together, which is critical when the LCF has little or no tack. In an automated process, the distributed chips sandwiched between backing paper can be carried on a conveyor belt through a heated oven and then through cylindrical pinch rollers of progressively decreasing gaps. The resulting RSRF is similar to sheet molding compound (SMC), and can be wound onto a take-up roll. In a semi-automated or batch process, RSRF can also be produced in the
form of large sheets by hot pressing vertically stacked layers of backing paper and LCF together, similar to the production of oriented strand board (OSB). The sheets can either be cut to the requisite size or wound onto continuous rolls. Pressing between heated platens allows high compaction pressures, creating a smooth, flat, and consistent sheet. If needed, additional resin can be added to the LCF before it is pressed into RSRF.

Regular cutout form (RCF) refers to smaller plies of regular shapes that are cut from scraps of larger pieces. This is shown schematically in Figure (1), based on some of the large scrap pieces shown in Ref [22]. The remaining scrap can then be converted to LCF. The backing paper remains intact in RCF and thus does not require further processing. RCF can be vacuum bagged or compression molded into small parts, or butt- and overlap- spliced to fabricate larger parts. Resold prepreg roll form (RPRF) consists of out-of-spec prepreg rolls that may be unopened or only partially consumed, e.g., when the rolls have exceeded their freezer life. It is cost-prohibitive to recertify these materials for manufacturing in the aircraft industry, and they are typically sent to landfills, donated to research universities, or used for in-house R&D work. However, RPRF can be resold to commercial entities downstream from the major manufacturers for use in non-primary and non-critical structures. RPRF can also be converted into LCF or used as the surfacing plies in RSRF sheets, thereby achieving high-quality surface and visual appearance of prepreg composites. The final intermediate product form, composite construction material (CCMat), resembles sheet lumber and intended to be used as construction material. CCMat can also be sold as pre-machined and pre-fabricated boards for assembly. CCMat can be produced by hot pressing stacks of RSRF.
The collection and sorting of scrap prepreg, manufacturing of these intermediate product forms, sales, marketing, and distribution could potentially form a new niche industry where one currently does not exist. Such an industry could help large businesses dispose of scrap prepreg in a sustainable way and in compliance with future legislation without distracting them from their core competency. At the same time, the new industry would provide a low-cost, processable, and high-performance raw material to small businesses and manufacturers downstream, which they could then use to develop lightweight composite products, thereby spurring job creation and revenue streams.

2.3. PROCESSING OF THE SCRAP PREPREG INTERMEDIATE FORMS INTO FINAL PARTS

We describe three primary routes to process scrap prepreg. Hot pressing involves dispersing LCF within or laying RSRF plies between two tool plates or inside an open mold that is then cured under heat and pressure. This is shown schematically in Figure (2). Either a release film or a liquid release agent can be used to facilitate part ejection from the mold after curing, the latter being preferable for curved parts. Because the fixture is open, depending on the temperature ramp rate and compaction pressure, the chips can spread laterally and resin can bleed out of the laminate, resulting in irregular edges that must be machined off. These cured trims can then be recycled via fiber reclamation. This process is simple, rapid and efficient; requiring no expensive consumables, no protracted vacuum holds, and no debulk cycles. The cured parts can be removed or ejected without having to wait to cool completely, allowing a continuous operation with a heated press maintained at a fixed temperature. Because heat transfer occurs via conduction and not by air convection, aggressive ramp ups can be employed to minimize the overall cycle times. The maximum achievable part thickness
is limited with this approach, as thicker initial charges of scrap prepreg can result in material loss from the open edges during compaction.

Figure 2. Processing routes for scrap prepreg
The second primary manufacturing route involves compression molding with a closed mold. This process requires opening and closing a mold each cycle. However, the closed mold prevents resin bleeding and constrains lateral movement of chips during pressing, resulting in parts with net shape, smooth surface finish, and minimal cured trim waste. Thick parts can be made in this way. Both LCF and RSRF can be deposited inside the mold, and the process is shown schematically in Figure (2). An O-ring can be used around the male part of the mold or piston to provide a sealed mold cavity, and additionally, a vacuum port can be introduced into the lower female part of the mold to evacuate the cavity prior to compaction, although not required. A thin film of resin can bleed up the sides of the piston in the absence of an O-ring or with the presence of clearance between the two mold parts. Provided that a release agent has been applied to the fixtures, this cured resin film can be removed easily.

The final primary manufacturing route involves conventional OoA-VBO and autoclave processing using RSRF, as shown in Figure (2). With OoA-VBO processing, RSRF is laid up over a tool plate, vacuum bagged, and then heated in a convection oven. Because there are no continuous air evacuation paths in RSRF as there are in OoA prepregs, no vacuum holds are required. Debulk cycles are also unnecessary, as RSRF plies are already pre-compacted under pressure. The use of OoA-VBO processing with scrap prepreg is well suited to highly contoured parts or parts that are too large to fit inside a hot press because of the size constraints of the platens and capacity of the press. This demonstrates the versatility of intermediate product forms such as RSRF, since they can be processed much the same way as virgin prepreg.
2.4. APPLICATIONS, END PRODUCTS, AND DEMONSTRATORS FROM SCRAP PREPREG

Parts manufactured with scrap prepreg bring to bear the advantages of virgin composite materials, as well as additional advantages. Scrap prepreg is essentially a free raw material that is currently deposited in landfills. Even when upcycled into intermediate product forms, the starting raw material cost will always remain much lower than virgin CF/epoxy prepreg that can cost ~ US $99 per kg. This will lead to lower price points for scrap-based composite end products. In addition, costly infrastructure and consumables are not required, while low-skilled labor is sufficient to manufacture parts, further reducing cost. Therefore, upcycling scrap prepreg can potentially enable new products, applications, and markets for composites that were previously closed to virgin composites because of their high material and manufacturing costs.

A variety of flat and contoured product forms can be readily produced from scrap prepreg by the routes described, and demonstrator parts support this claim. Figure (3a) shows an example of a contoured part, a prototype prosthetic ankle/foot produced from RSRF, while Figure (3b) shows an example of a common hat-stiffened panel. Additional demonstrator parts have been produced in our lab, including tubes, flat panels, and sandwich panels. The versatility of the materials and processes outlined indicate potential for a wide variety of products, including (1) medical parts and devices, (2) sporting goods and recreational equipment, (3) non-primary and non-critical aerospace and automotive structures, (4) construction, furniture, and shipping containers, (5) consumer and household goods, and (6) composite tooling.
Scrap prepreg can also be used to produce structural design elements that serve as building blocks for larger structures. These elements include thin and thick panels, cylinders, sandwich structures with honeycomb or foam cores, hat stiffeners, and even panels / veneers wherein scrap prepreg with multiple fiber types and resins have been mixed together in the same part, such as glass, carbon, and Kevlar fibers with different epoxies [22]. The resistance of CF/epoxy parts to moisture and insect infestation is appealing for use in indoor and outdoor furniture and as a basic construction material. Scrap prepreg can also find application in shipping containers currently made with denser materials that include aluminum, weathering steel, and wood. The light weight, durability, and FST properties are appealing qualities for inter modal freight containers for road, rail, and sea transportation, unit load devices in airports, and cargo containers for airplanes that would lead to reduced fuel costs. Scrap prepreg can be made into monolithic panels or into sandwich structures with a low density core to provide enhanced bending stiffness in these shipping containers. In addition, by virtue of the random placement and orientation of chips on the surface, parts produced from scrap prepreg possess an unusual aesthetic appearance. Incident light reflects differently off the randomly placed chips, creating patch-like reflectivity. Figure (3b) displays a hat-stiffened panel that exemplifies the surface finish of these parts. Further details of potential end uses and applications of CF/epoxy prepreg scrap can be obtained from Refs. [22,23].
Figure 3. Exemplary demonstrators made with scrap CF/epoxy prepreg (a) prosthetic foot (b) hat stiffened panel
3. MECHANICAL PROPERTIES AND MICROSTRUCTURE OF SCRAP PREPREG LAMINATES

3.1. MATERIAL, FABRICATION, AND TESTING METHODOLOGY

The prepreg material used in this study is a commercial OoA prepreg (Cytec CYCOM® 5320, ) with a 5-harness satin (5HS) fiber bed architecture (T650-35 6K tows). The typical resin content by weight for the virgin prepreg is 35-40% with a typical fiber areal weight of 370 g/m2. The particular stock of prepreg rolls used for this study was freezer stored for >3 years - well past the recommended freezer life of 12 months. The rolls had also been subjected to multiple freeze-thaw cycles over this period, exceeding the recommended tack life of 14 days and out-life of 21 days. Over time and with frequent handling, the sealed plastic storage bags used to store the individual prepreg rolls and stacks of pre-cut plies in the freezer had developed leaks. Thus the roll and plies of prepreg from which the scrap was sourced represented ‘worst-case’ scrap prepreg. The term ‘baseline’ used henceforth in this study refers to plies of prepreg cut from these dated, out-of-spec rolls and not virgin prepreg. Note that this prepreg line (CYCOM 5320) was discontinued by Cytec in favor of a newer prepreg system (5320-1). Thus, virgin 5320 prepreg material is no longer available to fabricate new panels. Consequently, the reader is encouraged to refer to the literature to compare the mechanical properties and microstructure reported in this study with that of virgin 5320 5HS prepreg laminates fabricated with various processing techniques, process conditions, and ply layups. (For additional information, see [24-26]).

Figure (4a) shows two types of chips cut from the scrap prepreg plies based on the orientation of the warp and fill tows within the chip: ‘0/90’ and ‘+/−’ where the latter refers to the 45° orientation of
the warp and fill tows with respect to the straight edges of the chip. ‘Mixed’ scrap refers to a 50-50 combination by weight of 0/90 and +/- scrap. In addition, a ‘Hybrid’ panel was produced using a 50-50 combination by weight of baseline plies and 0/90 scrap, i.e., a sandwich-like construction with the baseline plies on the surface and the scrap in the middle. Chip sizes on average were $7.6 \times 25.4$ mm. The scrap prepreg chips intentionally were not aligned when positioning the charge between the platens or when distributing the chips inside the compression mold before compacting, and thus the chips were randomly oriented within the resulting laminate. Laminates of in-plane dimensions $203.2 \times 203.2$ mm were fabricated using both OoA-VBO and hot pressing. The jagged edges of the open platen laminates were trimmed to yield a panel $177.8 \times 177.8$ mm. The cavity of the closed mold fixture was $228.6 \times 228.6$ mm. The vacuum bagged laminates consisted of a balanced, symmetric, quasi-isotropic layup using 8 plies of the baseline 5HS prepreg plies. The hot-pressed laminates were fabricated using either open platen or closed mold compression with two compaction pressures, 365 KPa (‘low’) and 731 KPa (‘high’).

For all scrap panels, two starting material states were considered. The first featured no additional out-time (‘+0 days’), i.e., scrap chips were cut from baseline plies that as noted earlier were already dated and beyond specification. The second featured an additional 14 days of out-time (‘+14 days’). Baseline plies were additionally aged by leaving them exposed for 14 days in a closed, no-window laboratory with temperature maintained between 22-24°C. The relative humidity fluctuated between 40-65% depending on the local weather conditions. A temperature and humidity data logger was used to record the laboratory environment over an extended period of time during the Fall (August), resulting in an average room temperature of 22 °C, relative humidity of 60%, and dew point of 14°C approximately. The laboratory had no windows, therefore the plies were not exposed to direct
sunlight. The baseline plies were aged with the backing paper still intact, and stacked on top of each other without a sealed bag around them, simulating conditions of a typical shop floor. Figure (4b) shows one of the manufacturer-recommended cure cycles (Cure Cycle B) for 5320 prepreg, consisting of a temperature ramp at 1.5 °C/min to 121°C followed by a 2-hour hold, then a ramp down to room temperature. VBO laminates with and without a 4-hour vacuum hold at room temperature (pre-cure) were fabricated. All VBO panels were subject to a free-standing post-cure cycle at 177°C for 2 hours. For the open platen hot-pressed laminates, two temperature ramp profiles were considered - a ‘Fast Cycle’ of ~40°C/min ↑ and ~15°C/min ↓, and a ‘Slow Cycle’ of ~4°C/min ↑ and ~1.5°C/min ↓ with a dwell at ~135°C. For the closed-mold, hot-pressed laminates, ramp rates of ~15°C/min ↑ and ~2.5°C/min ↓ were achieved. The slower ramp resulted from the greater thermal mass of the mold fixture. Hot pressed laminates were fabricated both with and without the 2-hour 177°C free-standing post-cure cycle. Figure (4c) compares the cure cycle schematics of the open platen slow cycle and closed mold cycle shown in Figure (4b) with actual thermocouple data from two exemplary runs. For the open platen processed panel, the thermocouple was attached to the panel, while for the closed mold processed panel, the thermocouple was attached to the outer (exposed) side of the mold fixture. Because of the size and mass of the closed mold fixture, it had much higher thermal inertia than the open platens, which are essentially two thin tool plates. No thermally insulating material was placed around the closed mold fixture during the run, and there was some lag before it reached the peak cure temperature.
Figure 4. (a) Cut scrap 5HS CF/epoxy prepreg chips (arrows indicate warp and fill fiber directions) (b) Schematics of all the cure cycles (c) Exemplary cure cycle temperature profiles using thermocouple data
In-plane tensile tests were performed on coupons cut from the fabricated panels following ASTM D3039, while compression tests with a combined loading compression (CLC) fixture were performed following ASTM D6641. Fiber volume fractions were measured following ASTM D3171 that also yielded the densities and 3D or volume void contents of the panels. After each test, micrographs of the failed surfaces and cross-sections were recorded to assess the failure morphologies. From each fabricated panel, the center coupon was polished on one side along its entire length and used to assess the 2D or area void content. Three sets of images were recorded from the left, center, and right sides of the coupon and stitched together to represent a length of approximately 50.8 mm, out of the total coupon length of 177.8 mm. A script (Matlab®) was then implemented to compute the area void content. Glass transition temperatures (Tg) of the fabricated panels were measured using Dynamic Mechanical Analysis (DMA, TA Instruments Q800) following ASTM E1640 using a temperature ramp rate of 5°C/min. Modulated Differential Scanning Calorimetry (mDSC, TA Instruments Q2000) runs were conducted on uncured scrap prepreg plies using a temperature sweep from -90°C to 250°C with a ramp rate of 3°C/min and modulating ±1°C/min every 60 sec, followed by equilibration at 25°C for 5 min and then a second ramp up to 250°C also at 3°C/min. The Tg was then periodically determined for 5HS scrap prepreg plies with additional out-times of 1-33 days. Note that while ASTM standards were used to guide the characterization, new testing approaches and standards may ultimately be required specifically for these chip-based composite materials.

3.2. GLASS TRANSITION TEMPERATURES AND EFFECTS OF POSTCURING

Figure (5a) shows the Tg values (from DMA) of various 0/90 scrap and baseline hot pressed panels with and without a post-cure cycle, and baseline VBO panels. These dry Tg values remain
approximately at 160°C and 180°C for the non-post-cured and post-cured panels, respectively. The dry Tg of virgin prepreg (CYCOM 5320) is ~210°C. Note that Tg is indicative of the degree of cure (DoC), and thus the scrap and baseline panels are apparently not reaching a full DoC with the recommended 177°C post-cure cycle. To investigate why the post-cured panels were not reaching the full Tg value, two additional trials were conducted. Select 177°C post-cured scrap and baseline panels were subjected to an additional post-cure cycle at 200°C for two hours. In addition, non-post-cured panels were subjected to a single post-cure cycle at 200°C. As Figure (5b) shows, with these elevated post-cure cycles, the panels now reach the expected Tg of 210°C indicating full cure. A plausible explanation for this behavior is that the combination of expired rolls (freezer life), out-time beyond specification, moisture absorption with each freeze-thaw cycle and exposure to the ambient environment has chemically altered the resin behavior. Otherwise, the 177°C dwell should have been sufficient for the scrap material to achieve the full Tg and DoC.

**Figure 5. DMA glass transition temperatures of scrap 5HS CF/epoxy prepreg panels (a) comparison of scrap and baseline panels (b) effect of post curing**

Figure (6) shows Tg values obtained from DSC analysis using scrap prepreg stored under lab ambient conditions for five weeks. Day 0 corresponds to when the scrap pieces were first removed from the freezer (i.e., already with some unknown previous out-time). Note that because the day 0
Tg value is positive, the scrap prepreg has an initial DoC greater than that of virgin prepreg. Typically the Tg values of fresh or virgin prepreg are negative, indicating B-stage Tg values. Using the heat of reaction, we can compare this DSC data with that of virgin 5320 5HS prepreg to estimate the initial DoC of the scrap prepreg (note that virgin samples are no longer available for this prepreg). While the preferred approach is to conduct DSC studies directly on neat resin samples (i.e. sourced directly from virgin and aged / exposed B-staged resin films), prepreg suppliers rarely make the prepreg resin film available (for proprietary reasons). With increasing out-times, the Tg values of the scrap prepreg increases in approximately linear fashion, reaching almost 55°C after 33 days of additional out-time (i.e., over and above the unknown previous out-time of this scrap prepreg), at which point the scrap is unusable and can no longer be re-processed because the resin has become powdery and brittle. For reference, the tack life and out-life of virgin prepreg are also indicated in Figure (6).

Plots such as Figure (6) provide useful data to determine how long scrap prepreg can be left out before it must be re-frozen, in turn indicating how quickly it must be collected at the source. It is also useful to qualify or screen incoming scrap prepreg to determine initial DoC and B-stage Tg. Once a sufficient database of properties has been compiled for scrap prepreg materials that are at various initial conditions and once process-property correlations are generated, one can readily determine whether the scrap can still be reused, and if so, the required process parameters and expected mechanical performance. In some instances, prepreg must be diverted to the cured scrap stream for fiber reclamation.
Figure 6. Effect of additional out-time on the B-stage glass transition temperature of scrap 5HS CF/epoxy prepreg

3.3. EFFECTS OF COMPACTION PRESSURE AND OUT-TIME ON OPEN PLATEN HOT PRESSED SCRAP PANELS AND COMPARISON WITH VBO PANELS

Figures (7a) and (7b) compare the 2D (area) and 3D (volume) void contents of scrap and baseline panels fabricated with open platen hot pressing for two compaction pressures (365 KPa and 731 KPa) and two out-times (+0 and +14 days) using the fast cure cycle. Both 2D and 3D void contents exhibited similar trends and magnitudes in the mean void contents. Higher compaction pressures and additional out-time resulted in slightly lower void contents than counterpart panels, i.e., lower compaction pressures and no additional out-time, respectively. Increased out-time corresponded to reduced tack, facilitating uniform distribution of scrap chips. The particular orientation of fibers within the chips had negligible effect on the void contents. The baseline hot-pressed panels showed the lowest void content of all, much lower than the scrap based panels. Figures (7c-e) compare the
material density, fiber volume fraction, and panel thickness of the scrap and baseline panels. Higher compaction pressures resulted in lower panel thickness and higher fiber volume fractions. For each case, the baseline panels demonstrated the highest material density and fiber volume fraction and lowest panel thickness.

Figures (7f-g) compare the specific tensile modulus and strength for the same panels, i.e., the modulus and strength normalized by the respective panel material density (Figure 7c) to allow direct comparisons between each. The scrap panels exhibit \(\sim 70\%\) retention in tensile modulus, while the hybrid panel shows a slightly greater tensile modulus than the baseline. The scrap panels exhibit \(\sim 15\text{–}20\%\) retention in tensile strength, while the hybrid panel shows \(\sim 60\%\) retention, despite the 50-50 combination of scrap chips and baseline plies. Neither the fiber orientation within the scrap chips nor the compaction pressure and additional out-time had any noticeable or consistent effect on the mean tensile modulus and tensile strength as indicated by the overlapping error bars. As mentioned earlier, the ‘baseline’ here does not refer to a virgin prepreg laminate, but instead to a laminate comprised of plies cut from the expired and out-of-spec prepreg rolls (i.e., scrap rolls).
Figure 7. Effects of compaction pressure and out-time on scrap 5HS CF/epoxy prepreg panels

(a) area void content (b) volume void content (c) material density (d) fiber volume fraction

(e) panel thickness (f) specific tensile modulus (g) specific tensile strength

Figure (8) shows typical hot-pressed panels. Resin edge bleeding occurred for baseline panels cured with a fast cure cycle, as shown in Figure (8a). The scrap panels pressed between open heated platens show irregular edges caused by the redistribution of chips during compaction, while the closed mold compression panels show well-defined edges (Figure (8b)).
Figure (9) shows the 2D and 3D void content, material density, fiber volume fraction, panel thickness, and specific tensile modulus and strength values of VBO processed baseline panels for two out-times (+0 and +14 days) and two room temperature vacuum hold times (0 and 4 hours). For the panels with no 4-hour vacuum hold prior to curing, the +14 day panels showed much lower void contents than the panels with no additional out-time. The lower void content in these panels is attributed to the reduced tack of the plies, which facilitated evacuation of inter-ply trapped gases under vacuum applied during the cure cycle. For the baseline panels with a 4-hour vacuum hold, the void contents were small, regardless of ply out-time. The VBO baseline panels showed similar tensile moduli, as shown in Figure (9f). The tensile strengths of the panels with a 4-hour vacuum hold were similar to those with no hold, an unexpected finding, given the much lower void contents (near-zero when considering area void content), and the expectation that tensile strengths and void contents are considered to be negatively correlated. Similarly, panels with a +14 day out-time showed lower tensile strength values in spite of lower void contents than panels fabricated with no additional out-time. This behavior is attributed to the fact that the baseline plies are well beyond their specification and potentially degraded. Comparing Figure (7) with Figure (9), the baseline hot pressed panels demonstrate increased tensile moduli and strengths compared to the VBO processed baseline panels. This finding is not unexpected, given the significantly higher compaction pressures achieved by hot pressing, in spite of the resin edge bleeding. Note that the overall cycle time for the hot pressed laminates was shorter than for VBO panels.
Figure 9. Effects of RT vacuum hold time and out-time on 5HS CF/epoxy baseline VBO prepreg panels (a) area void content (b) volume void content (c) material density (d) fiber volume fraction (e) panel thickness (f) specific tensile modulus (g) specific tensile strength

3.4. EFFECTS OF CURE CYCLE ON OPEN PLATEN HOT PRERESSED SCRAP PANELS

Figure (10) shows the effects of cure cycle on 0/90 scrap and baseline hot-pressed panels compacted at 365 KPa. The relatively short cure cycle resulted in much lower 2D and 3D void contents for both scrap and baseline panels compared to the fast cure cycle, as shown in Figure (10a). However, for each panel, the specific tensile strength values for both cure cycles were similar, as shown in Figure (10d). A similar trend was apparent for the specific compressive strengths, and these trends led to key observations. First, there appeared to be no correlation between void contents and strengths for these scrap and baseline panels, behavior first noted in Figures (7) and (9). Possible explanations include the operation of flaw mechanisms other than voids, such as non-uniform resin curing and embrittlement at the chip edges that could initiate failure before the effect of voids. Second, the cure cycle had little effect on the measured strengths. The specific tensile modulus for the fast cycle was greater than the slow cycle for both scrap and baseline panels. The temperature ramp rate affects the
resin viscosity, with higher ramp rates leading to lower minimum resin viscosities. With the fast cycle and a 365 KPa compaction pressure, significant resin bleeding was observed from the edges of the baseline panel, while little bleeding occurred for the slow cycle (see Figure (8a)). Excessive resin bleeding can result in dry spots and increased void content in the laminate, although as noted earlier, this has little effect on measured mechanical properties. On the other hand, resin bleeding can drive up the fiber volume fraction, as shown in Figure (10b), where the fast cure cycle resulted in a higher fiber volume fraction. For the 0/90 scrap panels, the fast cure cycle and lower resin viscosity led to more spreading of the chips under compaction, resulting in a panel with a slightly larger in-plane area than the slow cure cycle (see Figure (8b)). The jagged edges of the fast cure cycle panel contained ‘dryer’ chips that were not fully impregnated, whereas the slow cure cycle yielded a more impregnated set of chips along the periphery. In either case, these edges are ultimately trimmed and discarded.
Figure 10. Effect of cure cycle on scrap 5HS CF/epoxy prepreg panels

(a) area and volume void content (b) material density and fiber volume fraction
(c) specific tensile and compressive modulus (d) specific tensile and compressive strength

3.5. COMPARISON OF OPEN PLATEN VERSUS CLOSED MOLD COMPRESSION

Figure (11) compares open-platen to closed-mold hot pressing of scrap 0/90 laminates under two compaction pressures (365 KPa and 731 KPa), two additional out-times (+0 and +14 days), and with and without a post-cure cycle. The fast-cure cycle was used for all panels, noting that the closed-mold fixture featured a slightly different cure cycle as per Figure (4b). Both the 2D and 3D void contents were generally lower for the closed-mold scrap panels than for the open-platen panels. Compaction pressures had negligible effect on the void contents for the open platen panels. However, for the closed mold panels, the higher compaction pressure led to slightly lower void contents. Figure (11d) shows the fiber volume fractions, and no particular trend is apparent. The total volume of the composite is comprised of the fiber, the resin, and the void volumes. By summing the fiber and void volume fractions in Figure (11), the resin volume fraction of the closed-mold
panels is greater than the open-platen panels, as expected. There are competing effects at play. In open-platen processing, the chips have greater ability (mobility) to re-distribute. However, resin can bleed from panel edges, which can result in dry spots and large voids inside the scrap panel. While resin bleed can drive up the fiber volume fraction, increases in voids and dry spots can drive it back down as the void volume fraction increases.

Note that OoA prepregs are designed to be net-resin prepregs. Because the resin is not fully contained, it generally is not hydrostatically loaded during bagging. Therefore, for the same applied compaction pressure, the resin pressure in open-platen processing will always be less than that in closed-mold processing. Voids can form when the partial pressure of the gases or volatiles in the resin is sufficient to resist the resin pressure – otherwise, the voids collapse under the resin pressure. In contrast, the chips have limited mobility in closed-mold compression, and the resin is pressurized to greater extents, as it is contained within the mold cavity. Thus, in closed-mold compression, the void content will generally decrease with increasing compaction pressure, and the void content will typically be lower than open platen processing. However, the fiber volume fraction is not necessarily increased because the resin remains in the cavity and there is no mechanism for excess to bleed out. This factor was part of the rationale for not using an O-ring during the closed-mold processing, thus allowing a thin film of resin to bleed up the sides of the piston and into the O-ring groove. However, this in turn may lead to reduced resin pressure and potentially the re-occurrence of voids based on the relative pressures, thereby resulting in an additional competing effect.

Figure (11e) shows the panel thickness values. The thickness remains insensitive to the compaction pressure for closed-mold compression, as the chips are already tightly packed within the closed
volume. However, open-platen compression results in an appreciable reduction of panel thickness with increasing compaction pressure, as the chips have greater mobility and can redistribute within the platens (with reduced tack increasing this effect). Note that we do not attempt direct comparisons of absolute thickness between the open-platen and closed-mold panels, as the initial volumes of chips used were different for both cases, as were the in-plane dimensions, as reported earlier. Figures (11f) and (11g) show the specific tensile moduli and strengths of the panels. No distinct trends are apparent, although a few observations can be made. Post-curing the panels at the recommended 177°C did not seem to affect the tensile modulus and strength values of the scrap panels - the tensile strengths were statistically indistinguishable from one another, as seen from the overlapping error bars in Figure (11g), where the error bars indicate variability in tensile strength. This unexpected behavior can be attributed to the panels not reaching a full DoC with a 177°C post-cure cycle, as shown previously in Figure (5b). Additional scrap panels were not available for post-curing at 200°C and then conducting additional mechanical tests to evaluate the effect on modulus or strength. Note also the similarity in tensile strength values between the open-platen and closed-mold panels shown in Figure (11g), despite the much greater void contents for the open-platen scrap panels, as shown in Figures (11a) and (11b). This finding further supports the conclusion that there does not appear to be a correlation between void content and tensile strength for the scrap prepreg panels. This result is attributed to the presence of other flaw mechanisms that either supercede the effect of voids and/or dominate the failure mechanism. Further studies are required to determine the exact cause of this unusual trend.
3.6. MICROSTRUCTURE, FAILURE AND VOID MORPHOLOGIES

Figure (12) shows failure morphologies of the scrap and baseline hot-pressed panels processed with the fast cure cycle after tensile and compression testing. The failed scrap panels show multiple tortuous crack patterns that follow the boundaries between chips. Thus, the failure in scrap panels appears to be matrix-dominated. The baseline panels in contrast exhibit inter-laminar crack paths with significant fiber failure in tension. The failure is also more catastrophic for the baseline panels, as the elastic strain energy built up during tensile testing releases suddenly. Failure in the baseline panels is fiber-dominated because of the continuous plies.
Figure (13) shows cross-sectional views of open-platen and closed-mold scrap panels for two compaction pressures (365 KPa and 731 KPa) and two additional out-times (+0 and +14 days). The open-platen panels are distinguished by larger and highly irregular void shapes and in some cases dry spots. The void morphology of the closed-mold panels on the other hand show fewer voids that are smaller and more regular in shape, and more evenly distributed through the cross-section. Voids in continuous-ply prepreg laminates can be inter-laminar (within the matrix between the plies), inter-tow (within the matrix interstitials between the tows), or intra-tow (in-between the fibers within the tow cross-section). Less tack between plies or plies with longer out-times generally helps reduce inter-laminar voids that form from air trapped between two resin layers. The pre-cure room
temperature vacuum hold and recommended controlled temperature ramp rate during cure help minimize inter-tow and intra-tow voids by ensuring that tow fiber bundles are not fully impregnated by the resin flow before the air is fully evacuated through the tows.
Figure 13. Micrographs of scrap SHS CF/epoxy prepreg panels showing the void content and microstructure
The mechanisms affecting void formation are slightly different in the scrap panels. The initial chip distribution is important because tacky chips readily agglomerate, creating empty pockets in the initial charge. For open-platen processing, where the resin can bleed from the edges and is not pressurized to the full extent by the applied compaction pressure, this can lead to larger voids and dry spots. For closed-mold panels, this problem can be mitigated by using chips with minimal tack, or by using the RSRF or scrap roll that is already pre-compacted. Such chips can be dispersed more uniformly and have increased mobility during compaction. This is illustrated in Figure (7), where the +14 day 0/90 scrap panels typically had lower void contents. Voids in scrap panels are typically more prevalent at the chip edges, as opposed to between the chips (analogous to inter-laminar voids) or inside the chips (analogous to intra-tow voids). Recalling that these scrap panels do not have air evacuation channels like continuous-ply prepregs and are not subject to vacuum during curing, voids will form wherever trapped gas is present and the partial pressure of the trapped gases is sufficient to withstand collapse by the surrounding resin, which is more likely at the chip edges. Alternative techniques can be applied to minimize porosity in the scrap panels, such as applying vacuum during hot pressing or adding compatible resin to the initial scrap charge prior to pressing. However, as shown earlier, there is no clear correlation between void content and tensile strengths. Therefore, unlike continuous-ply virgin prepreg laminates, void content is not a strong indicator of performance for scrap panels.

4. CONCLUSIONS

A technologically viable technique to reuse uncured scrap prepreg material and upcycle it to end-products was demonstrated. The process involved converting scrap prepreg into one of four
intermediate product forms, identified as LCF, RCF, RPRF, and RSRF, then processing these intermediate forms into final end-products through hot pressing and compression molding. A fifth form, CCMat, consisted of fully cured composite ‘lumber’. The low cost of the starting raw material coupled with simple, efficient, and rapid processing routes should translate into lower price points for commercial end-products while maintaining relatively high performance expected of composite materials. In addition, the demonstrator parts based on scrap prepreg can be aesthetically appealing, featuring a smooth and distinctive surface appearance by virtue of the random orientation and placement of the chips. While the majority of the uncured scrap thermoset prepreg in the US is currently sent to landfills because of convenience and the relative immaturity of reuse and recycling technologies, the methodologies and ideas presented here provide impetus to industrial suppliers to adopt more sustainable manufacturing practices to better utilize uncured scrap prepreg. Potential applications and industries that could utilize scrap prepreg to manufacture end-products include medical parts and devices, sporting and recreation equipment, non-critical aerospace and automotive structures, shipping containers, composite tooling, and household and consumer products.

We investigated the properties and performance of scrap prepreg based laminates under different starting material states and processing conditions, leading to several conclusions. The conventional rationale behind certain processing techniques and performance metrics used for virgin prepreg composites did not necessarily apply to scrap prepreg composites. For example the relatively high void contents (>2%) of the scrap panels did not translate to decreased performance. In fact, the void contents showed little correlation with tensile strength, in distinct contrast with virgin prepreg panels, pointing to the prevalence of other mechanisms. While prepreg tack is considered essential for virgin ply layups, tack in the scrap chips actually inhibited uniform dispersal and redistribution, resulting in higher void contents. Furthermore, the mechanical performance of scrap panels
comprised of chips with some tack were similar to those without any tack. While virgin prepreg materials are processed with relatively long cure cycles and slow temperature ramp rates (e.g. 0.5-3°C/min), the scrap panels in the present study were cured with aggressive cure cycles with ramp rates as high as 40°C/min. The scrap prepreg did not require lengthy pre-cure vacuum hold and debulk cycles, significantly reducing the cycle times and offering the prospect of increased production rates. In addition, scrap panels processed under slow and fast cure cycles resulted in similar mechanical performance despite the latter showing higher void contents and in the case of open-platen processing, excess resin edge bleeding. The manufacturer-specified post-cure cycle for virgin prepreg composites raised the DMA-based Tg value, although not to the value expected for a fully-cured composite. Therefore, this post-cure cycle did not fully cure the scrap prepreg panels, pointing to a thermochemical change in the resin structure brought about by a combination of the prolonged freezer life, out-life, and moisture uptake from the ambient environment with the repeated freeze-thaw cycles. Moreover, this post-curing of the scrap panels did not result in a statistically significant increase in the tensile strength of the scrap panels, in sharp contrast with the effect of post-curing on virgin prepreg panels. However post-curing at temperatures higher than manufacturer-recommended resulted in a further degree of cure, as evident by the increased DMA-based Tg values. The corresponding effect on mechanical properties however is unclear, as there was insufficient material available for testing. Scrap prepreg panels exhibited unusual stiffness retention in both tension and compression compared to baseline panels comprised of continuous plies of the same prepreg material in the same initial state, and in some cases were near 100% retention. Again, note that at the time of this study, virgin 5320 prepreg material was no longer available to fabricate panels and measure mechanical properties; therefore the reader is referred to the literature to make comparisons with the properties reported here.
The scrap prepreg panels also demonstrated partial retention of tensile and compressive strength. The high variability in the reported microstructural and mechanical properties motivates the need for further processing and characterization studies of reused scrap prepreg laminates. The mechanical properties reported here were based on a scrap prepreg in an excessively aged and diminished state, and are meant for informational and illustratory purposes. In reality, we expect commercial reuse efforts to source prepreg scrap close to the site of generation and process it quickly into intermediate forms which will then be properly stored in freezers until they are fully processed into parts. The strength and stiffness properties of such parts, including consistency and quality, are therefore expected to exceed the values reported here.

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