

# Recyclability and reutilization of carbon fiber fabric/epoxy composites

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## Abstract

Solid carbon fiber/epoxy laminates manufactured by liquid resin infusion of twill fabric reinforcement are recycled in a bath of boiling sulfuric acid to separate the fibers from the matrix. The recycled reinforcement consists of long fibers arranged in a random, entangled mat. Using the same epoxy matrix and infusion materials and process, the recycled fibers are reutilized to manufacture solid laminates. The physical properties of the recycled laminates are evaluated by means of pulse-echo ultrasound, visual microscopy, and fiber volume content. The average fiber volume content of the recycled laminates is 33%, compared to the 62% of the twill laminates. The mechanical properties of the recycled composite include tension modulus and strength, compression modulus and strength, three-point bend flexure strength, and short beam shear strength. The properties are compared against the values of the twill reinforcement with quasi-isotropic stacking sequence. Results show that the recycled material offers promising elastic properties and strength values, similar to those of advanced carbon fiber sheet molding compounds, and therefore can be used as structural material.

## Keywords

recycling, liquid resin infusion, mechanical testing

## Introduction

A wide range of composite material forms are finding use in today's aerospace, automotive, and other transportation industry segments.<sup>1–5</sup> These materials are finally fulfilling the promise of providing manufacturers with a cost-competitive alternative to aluminum alloys. The Boeing 787 Dreamliner, due to join the world's active fleet by 2010, features over 50% carbon fiber reinforced polymers (CFRP) by structural weight.<sup>1</sup> Beside the direct benefits resulting from the greater specific mechanical properties, such as increased fuel efficiency, and reduced pollutant and acoustic emissions, other indirect advantages of a CFRP-intensive airframe are reduced maintenance requirements, and increased passenger comfort due to the superior fatigue- and corrosion-resistance characteristics of these materials. However, the introduction of composites in the primary structure of modern aircraft presents new challenges, such as meeting crashworthiness and lightning strike protection requirements, and developing a maintenance, inspection, and repair strategy. End-of-life considerations are also becoming increasingly important,

because of the large volumes of parts comprised of CFRP that are entering service. The vast majority of CFRP for aerospace and automotive structural applications consists of thermoset resins (typically epoxy and vinylester), which cannot be re-shaped or re-formed once processed. The chemical structure of thermosets, which imparts them the desirable attributes of high mechanical properties and environmental durability, is associated to an irreversible cross-linking process that takes place during curing. Hence, once a part is

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shaped and cured, it is not easy to physically or mechanically separate the matrix from the reinforcement. Even if careful effort is taken to reduce production waste during manufacturing, there is still a need to recycle CFRP material in order to avoid incinerating or disposing it in landfills.

Since the 1990s, industry has been investigating ways to recycle CFRP products.<sup>6,7</sup> Boeing and other industrial organizations have come together in 2006 to establish a common industry working group, Aircraft Fleet Recycling Association (AFRA), with a mutually shared commitment to improving older fleet asset management and fostering the recovery and reuse of aerospace materials. Boeing is working with AFRA-affiliated companies to develop technologies and processes capable of recovering CFRP parts after the end of their service life. The recycling of composites is normally a two-step procedure that involves first, a mechanical separation process of composites from other aircraft materials during an aircraft's retirement, and second, a recycling process that recovers fibers of sufficient quality so that they can be re-introduced as a materials source in manufacturing. To date, several recycling technologies have been suggested for aerospace-grade CFRP materials,<sup>8-10</sup> which can be broadly divided into mechanical, thermal, and chemical processing. In mechanical processing, the cured parts undergo crushing in a series of mills, through which the material is reduced to particles ranging in size from 50  $\mu\text{m}$  to 10 mm. In this process, all the constituents of the original CFRP appear in the resulting recycle, which is therefore a mixture of matrix, and reinforcement. These recycles are typically reutilized as powder fillers in new composites, not for structural applications. For example, they have been used to replace calcium carbonate in commercial-grade sheet molding compounds (SMC) or bulk molding compounds (BMC), the advantage of the recycles being that they are lighter than the materials they replace.

In thermal processing, it is possible to separate the fibers from the matrix, since the fibers have the most recoverable value in a CFRP. The bulk of the efforts to date have focused on pyrolysis and vacuum pyrolysis as the primary process to extract fibers for reutilization.<sup>8-12</sup> Heat is applied in the absence of oxygen, typically in furnaces using inert gases or under vacuum. Typical temperatures of 500°C and 600°C require several hours for the pyrolysis to take place. Single-fiber mechanical tests seem to show that contradicting results have been obtained, with some authors reporting more or less degradation in strength. Milled Carbon Ltd<sup>6-8</sup> uses a pyrolytic continuous flow process to burn off the resin and additives and separating the fibers, which can then be processed by chopping or milling for various applications. Pyrolysis was initially deemed a viable

technology but later discarded as the primary process because at a temperature of 500°C, the carbon fibers retain oxidation residue or char.

Another thermal process being proposed is catalytic conversion, whereby a depolymerizing catalyst is used to convert the cured resin into low-molecular-weight hydrocarbons.<sup>6-8</sup> Adherent Technologies Inc. uses a heat transfer fluid (e.g., phenol), which is heated between 150°C and 300°C, to remove the bulk of the resin and some contaminants. Vacuum pyrolysis is used as posttreatment to eliminate the remaining resin and contaminants, and achieve 99% purity. The process, though still in the early phases of development by Adherent Technologies, yields more pure carbon fibers than pyrolysis. However, challenges arise if the parts contain scrap contaminants (such as metal, paint, wires), which are typically present in all structures.

Within the framework of AFRA, shredded composite scrap from retired or damaged Boeing F/A-18 horizontal stabilizers was sent to both Milled Carbon and Adherent Technologies.<sup>6-8</sup> The reclaimed AS4 fibers underwent visual and X-ray surface quality assessment plus single-fiber mechanical property testing. The recycled fibers were chopped and successfully incorporated into SMC, BMC, or as resin injection molding feedstock, and incorporated into fiber preforms for the compression molding of an automotive component (Corvette C6 fender well).

A third type of thermal processing being proposed at the laboratory scale uses fluidized bed treatments or supercritical fluid treatments to successfully separate the matrix from the fibers. In the fluidized bed treatment,<sup>8,9</sup> the scrap composite is placed in a bed of silica sand fluidized with heated air at temperatures of 450–500°C. During this thermal process, the polymer matrix material volatilizes leaving recovered carbon fibers, which are separated through a cyclone from the gas stream. In the supercritical fluid treatment,<sup>13</sup> which is a thermal and chemical process, the pieces of material to be recycled (approximately 25 mm long) are infringed by a supercritical flow (e.g., propanol or glycol), at temperatures between 450°C and 500°C, pressure of 50 bars, and velocities up to 1 m/s, which removes the epoxy resin from the surface of the fibers. The recycled product is fluffy and random, and comprises individual fiber filaments with minimal contamination and mean length from 6 to 10 mm. Although processed in air, the carbon fibers seem to show negligible oxidation, and small reduction in surface oxygen content, indicating a good potential for subsequent bonding to a polymer matrix. The fibers exhibit a strength degradation of 20%, while the modulus remains unaffected. Advantages of the fluidized bed process is that it is suitable to process parts containing mechanical inserts, surface paints, and foam cores. The recycles

could be utilized as reinforcement for nonstructural composite applications such as BMC or nonwoven veils.

Microwave, thermal shock, and molten salt bath processes have also been used with partial success, but they have a tendency to produce recycled fibers with residues of resin or other contaminants on the surface.<sup>8,10</sup>

Limited effort has been directed toward chemical processing. Little success was achieved in Jody et al.<sup>10</sup> where a glycol/water solution at 240°C was used to treat composite scrap containing a urethane-based substrate. A more successful approach was conducted in ref.,<sup>14</sup> where neat epoxy resin and epoxy-based composites were dissolved in a solution of nitric acid at temperature 80°C and concentration of 4 M (mol/L). The extract was then neutralized and utilized subsequently for repolymerization. The study showed that it is possible to recover the epoxy resin following chemical processing, and the thermal and mechanical tests show that strength and glass transition temperature for the recycled resin are much higher than the pristine resin due to a higher density network structure. Another promising method of recycling of carbon fibers from carbon/epoxy composites with a nitric acid solution was demonstrated in Liu et al.<sup>15</sup> Three variables were controlled during the experiment: decomposition temperature (90°C), acid solution concentration (8 M), and ratio of specimen weight to acid solution volume (6 g:100 mL). The epoxy matrix in the carbon/epoxy composites was completely decomposed in 12 h. Then, the insoluble carbon fibers were separated from the solution and washed with acetone, and dried in an oven at 60°C. The carbon fiber surface was clean, with very little contamination. Gel permeation and gas chromatography showed that the epoxy resin could entirely decompose into low-molecular-weight compounds, with negligible contamination on the fibers. Electron probe microscopy showed no damage to the recycled carbon fibers, and single-fiber tension tests showed that the strength loss of the recycled carbon fibers was negligible.

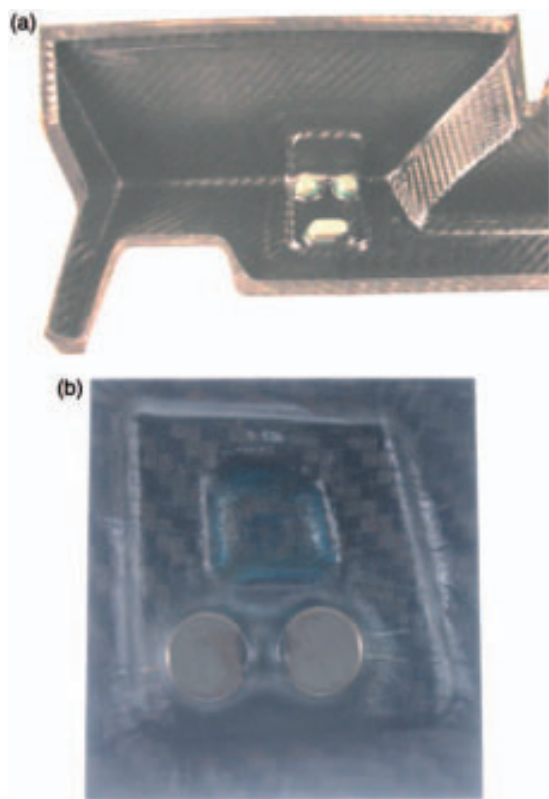
A summary of the studies performed to date on the possibility to recycle and reuse carbon fibers shows that the majority of the efforts call for chopping or milling of the recycled fibers, with a particular effort aimed toward fine particulates, whiskers, and nonwoven mats. These materials are not well suited for structural applications, but may be utilized for their high thermally or electrically conductive properties. This study focuses on assessing the possibility to use reclaimed fibers for structural applications. The reclaiming process is based on chemical solution of the matrix, which generates pure recycled fibers, without residues, contaminants, or surface modification, unlike many of the

currently used recycling processes. The precursor is a fabric, not tape as in the majority of the recycling studies performed to date, and the recycling process generates a mat of long, entangled, random fibers which can be used as reinforcement without the need for secondary chopping or milling. Finally, it proposes a direct comparison in terms of physical and mechanical properties between the composites made of pristine fiber fabric and those made of the reclaimed random mat. Using the same liquid resin and infusion process to manufacture the composite laminates with the pristine and recycled reinforcements, it is possible to extrapolate the immediate influence of the architecture of the reinforcement on the laminate-level properties. These tests yield significant information on the possibility to use the recycled fibers as reinforcement for structural applications.

## Experimental approach

### *Pristine material*

Parts are manufactured by vacuum-assisted resin transfer molding (VaRTM) at the Advanced Composites Research Center of Automobili Lamborghini S.p.A. in Sant'Agata Bolognese, Italy. The parts include embedded surface-treated aluminum inserts for bolted connections and are representative of typical primary automotive structures used by the super luxury car manufacturer (Figure 1(A) and (B)). Flat plates are also manufactured using the same method in order to obtain control specimens for comparative mechanical performance evaluation. Two different quasi-isotropic stacking sequences are investigated, yielding an upper bound and a lower bound for flexural strength. These are [(0/90)/(±45)]<sub>2s</sub> and [(±45)/(0/90)]<sub>2s</sub>. The reinforcement used is a Toray T700 carbon fiber, 12 k tow, 2 × 2 twill architecture, 380 gr fiber areal weight (FAW) (Figure 2(A)). The dry fabric reinforcement comes in a roll with a pre-applied binder, or tackifier, used for manual preforming or automated thermoforming (Figure 2(B)). The binder is present in a nominal content of 5% by weight. It should be noted that woven fabric comprises over 60% of Europe's carbon fiber waste.<sup>7</sup> The resin used is a Huntsman two-part liquid epoxy with XB 3518 BD resin and Aradur 22962 hardener, in a mix ratio of 100:26 parts by weight (Table 1). The untoughened resin is a low-viscosity system designed for infusion and has a pot life of over 3 h at room temperature but only 15 min at 60°C. The oven cure cycle calls for 1 h at 100°C with vacuum, followed by 2 h postcure at 140°C, giving a dry  $T_g$  of 135°C. The infusion process utilizes a distribution media, which is essential to achieve good resin distribution in the panel. The size of the distribution media, which

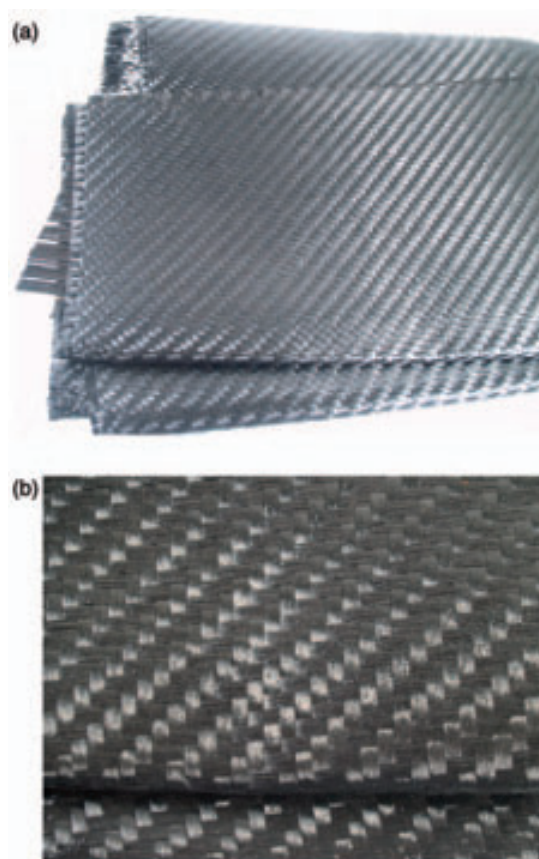


**Figure 1.** A, B. Example of parts manufactured by vacuum-assisted resin transfer molding (VaRTM) and containing aluminum inserts.

does not cover the entire preform, is determined by trial and error. The preform coverage by the distribution media is given by a complex interaction between preform permeability, resin viscoelastic characteristics, and part geometry, but unfortunately it can only be determined through experimentation. Other infusion disposable materials include breather, perforated and nonperforated barriers, vacuum bag, and sealant. A caul plate is not used to manufacture the panels with the pristine twill reinforcement.

### Extraction of fibers from the matrix

The automotive parts are received at the Automobili Lamborghini Advanced Composite Structures Laboratory at the University of Washington, Seattle, WA, where they are cut into smaller sections ( $200 \times 200 \text{ mm}^2$ ) for ease of processing. This step is not required, but it is used due to the constraints of the experimental setup that is described below. For industrial applications, this step would be avoided, which would streamline the operation and yield longer fibers. The procedure used in this study to extract the carbon fibers from the CFRP parts is loosely based on the principles of acid digestion,<sup>16</sup> which is a



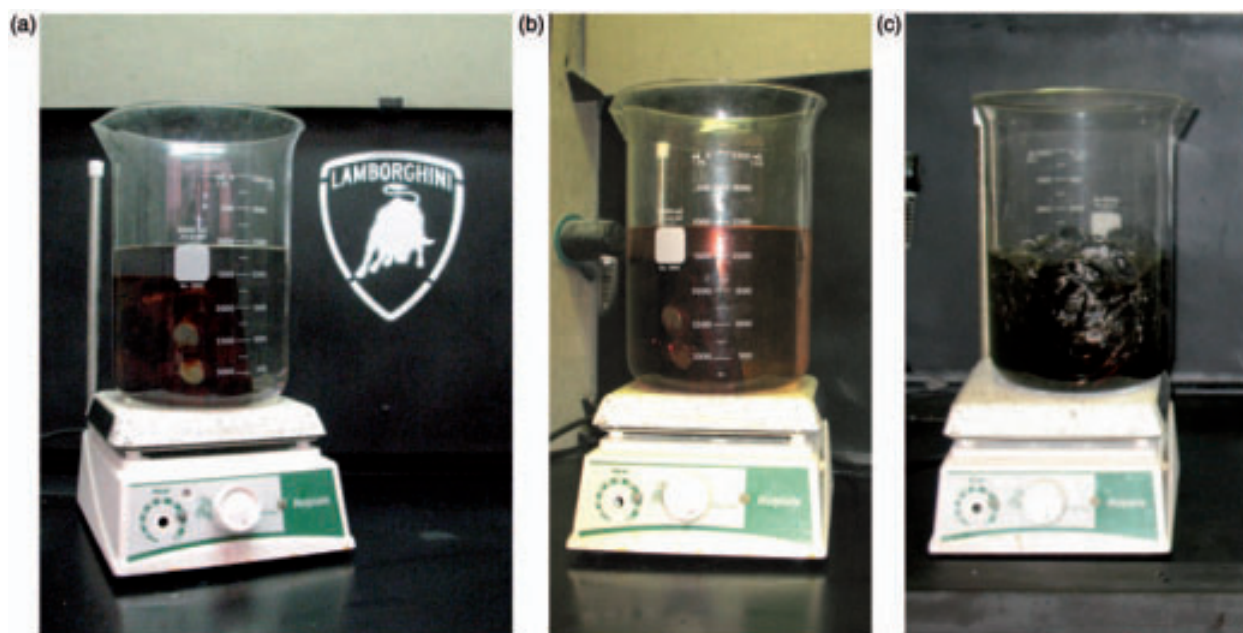
**Figure 2.** A, B. Dry fabric reinforcement and detailed view of thermoset binder.

test method utilized to determine the fiber volume content  $V_f$  of a composite material. The specimen is placed in a glass acid- and heat-resistant container (i.e., a large beaker); then sulfuric acid is added and the solution heated on a burner to  $110^\circ\text{C}$ . When the solution starts to boil, hydrogen peroxide is added to accelerate the reaction and oxidize the matrix. The process takes place under a fume hood to remove the harmful gases. Mechanical stirring throughout the duration of the entire process is essential. If the parts are left in boiling acid without stirring, the separation of the matrix from the fibers does not take place, and only the surface of the matrix gets removed. The process typically takes several hours from beginning to end. When the digestion is complete, the container is removed from the burner in order to cool down, at which point the solution is separated from the reinforcement through a series of filters. A vacuum pump is used to provide the aspiration force for the liquid to go through the series of filters. Unlike the actual acid digestion standard, which requires filters in the order of a few microns, for this purpose, it is possible to use much coarser filters, since the objective is to recover only the large fibers for subsequent reutilization. The



**Table 1.** Summary of test families

Family	Fiber	Architecture	Stacking sequence	Processing	Epoxy resin/ hardener
A	T700 12k	2 × 2 Twill	[(0/90)/(±45)] <sub>2s</sub>	VaRTM	XB35 18BD/ 22962
B	T700 12k	2 × 2 Twill	[(±45)/(0/90)] <sub>2s</sub>	VaRTM	XB35 18BD/ 22962
R	T700 12k	Long random fiber mat recycled	Quasi-isotropic	VaRTM	XB35 18BD/ 22962

**Figure 3.** Separation of fibers from resin and insert by acid digestion. The solution progressively changes color from transparent (A), to brown (B), and to black (C).

filtration process requires approximately 30 min. After filtration, the fibers are washed with acetone and distilled water to remove traces of sulfuric acid as well as other contaminants. They are subsequently dried in an oven at 100°C for 30 min.

During the digestion, the transparent solution of sulfuric acid and hydrogen peroxide changes color from light brown to dark brown, and eventually to 'pitch' black (Figure 3(A)–(C)). This suggests that not only the resin, which is transparent, but also other materials are dissolved during the process. These may include the binder (also known as tackifier) or carbon fiber dust (microparticles of carbon fibers resulting from fiber processing). It should be noted that the binder has an opaque, white color, and hence it should not be responsible for the black coloration. The carbon fibers are insoluble in acid; so they do not contribute to the black coloration by mass loss. A test is performed to measure the weight loss of the dry fabric reinforcement, which also includes the binder and the sizing. Following the steps of ASTM D3171,<sup>16</sup> it is possible to measure

the weight loss at 5.7%, as an average of three measurements. The nominal content of binder is 5% by weight, but a ±1% variation is expectable according to the manufacturer. The coloration of the mixture following the acid digestion is black, as it was in the case of the cured panels. Based on these results, the black coloration can be attributable to a minor loss of carbon fiber dust/powder that is left around the fibers after processing.

The result of the separation process is a mass of entangled carbon fibers, with a random distribution. The mass of fibers is fluffy and fuzzy, with the majority of filaments maintaining the form of single tows (Figure 4). The aluminum inserts are preserved and can be reutilized in other applications, though the protective coating will likely need to be reapplied prior to embedding in the CFRP. The 2 × 2 twill architecture is mostly lost, but it remains preserved in few areas (Figure 4). The fibers are continuous and approximately 200 mm long, which is the size of the composite specimen panel from which they were extracted.



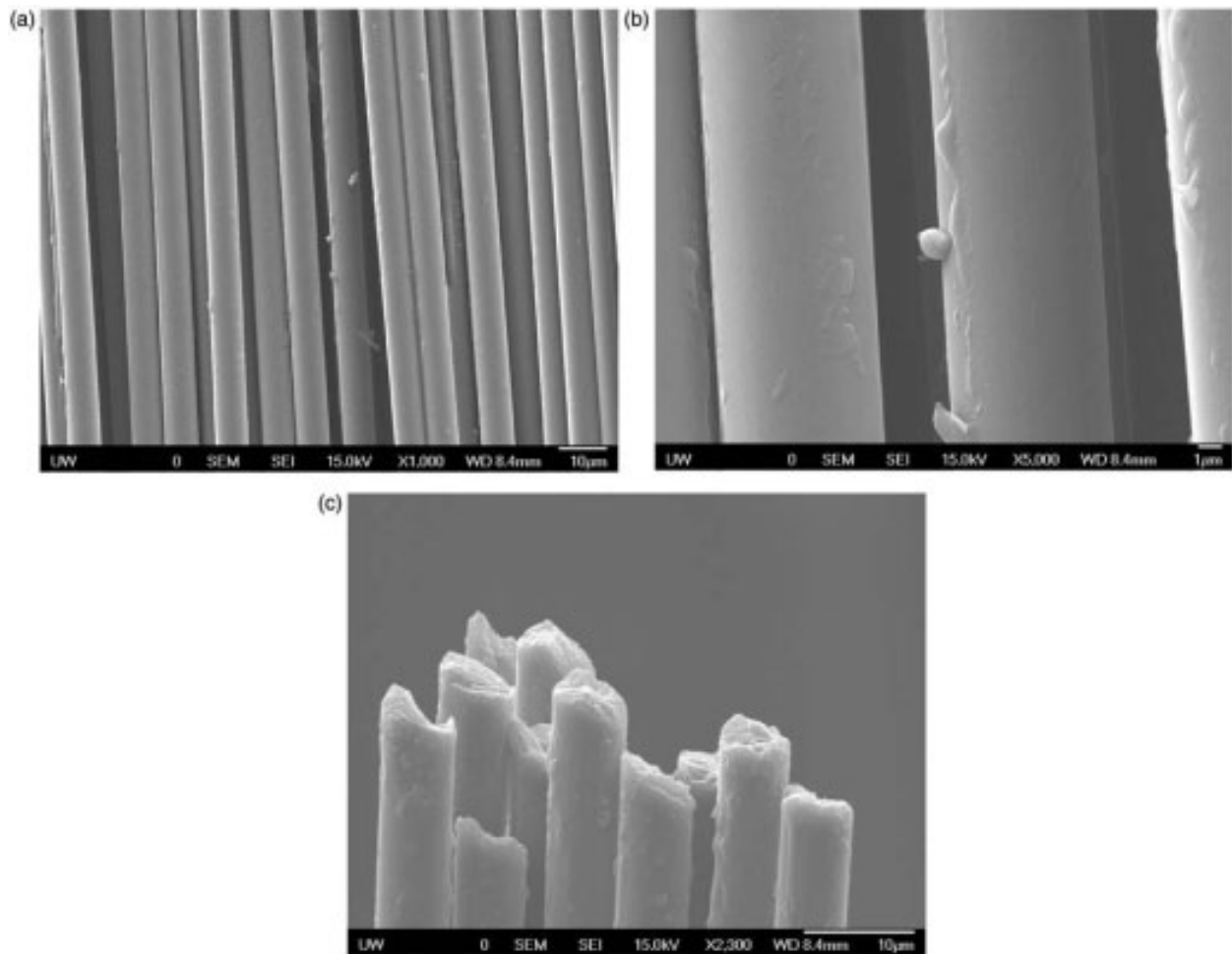
**Figure 4.** Carbon fiber reinforcements after acid digestion (a) and separation of aluminum insert (b); the fibers appear mostly as single tows, but in a few places, the twill architecture is still visible.

Using a scanning electron microscope (SEM), it is possible to evaluate the surface purity of the recycled fibers, and compare them to the pristine ones. The pristine fibers of the dry twill fabric exhibit a rough, irregular surface at  $\times 1000$  (Figure 5(A)), which upon greater magnification (Figure 5(B) and (C)) reveals to be associated to a liquid or spray coating, most likely the fiber sizing or the binder. The recycled fibers are free of resin, and exhibit a much more regular and smooth surface (Figure 6(A)–(D)). Very few traces of micron-sized resin residue can be seen in few regions on the fiber surface, but overall it can be confirmed that chemical processing yields very pure fibers with little or no resin residue.<sup>15</sup>

### Reutilization of fibers

The recycled reinforcement is used, without additional processing, as the preform for VaRTM infusion. The same resin, infusion process, and materials are utilized as in the case of the pristine twill architecture material (Table 1). This ensures that the only difference between pristine and recycled specimens is the reinforcement. It is acknowledged that the binder is lost during this operation, and for industrial purposes the recycled fiber mat would have to be reapplied with the binder prior to utilization. The infusion procedure for the recycled random mat is nearly identical to the one used for the pristine twill reinforcement, with the difference that a semi-rigid perforated caul plate is used to obtain panels of constant thickness (Figure 7(A)). The random mat is

not homogeneous, since the FAW varies from point to point. The variation in fiber density results in different ‘preform’ thicknesses when subjected to vacuum pressure. After infusion and curing, these regions result in large thickness variations on the bag side. A 2-mm thick aluminum caul plate is used to reduce the thickness variation. The caul plate is perforated with 6.35 mm holes to allow for the vacuum to distribute evenly across the preform, to facilitate resin flow, and to impart flexibility to the caul plate itself. However, since the FAW of the preform remains variable from point to point, the resulting fiber volume content of the laminate varies accordingly. The setup under vacuum prior to infusion also shows line ports for resin injection and aspiration (Figure 7(B)). A total of three plates for each family (A, B, and R) are manufactured by VaRTM, having dimensions  $300 \times 300 \text{ mm}^2$  and nominal thickness 4.0 mm. The families are described in Table 1, and they include the two quasi-isotropic stacking sequences described above (families A and B) and the recycled mat (family R). The average measured thickness for families A and B is 4.0 mm ( $\pm 0.2$  mm), without the use of a caul plate. For family R, the average thickness measured thickness is 4.3 mm ( $\pm 0.4$  mm), with the use of a caul plate. A representative surface of the mold side of a composite panel of family A is shown in Figure 8(A) and exhibits the familiar twill pattern. A representative mold side surface for the recycled composite panel, family R, is shown in Figure 8(B), and shows the long random fiber pattern with some evidence of the original twill architecture.



**Figure 5.** Scanning electron microscope (SEM) images (at different levels of magnification) of pristine carbon fibers show the presence of a distributed fiber coating as well as discrete larger particles, both of which can be attributed to either the binder or the sizing: (A)  $\times 1000$ , (B)  $\times 5000$ , and (C)  $\times 2300$ .

## Results

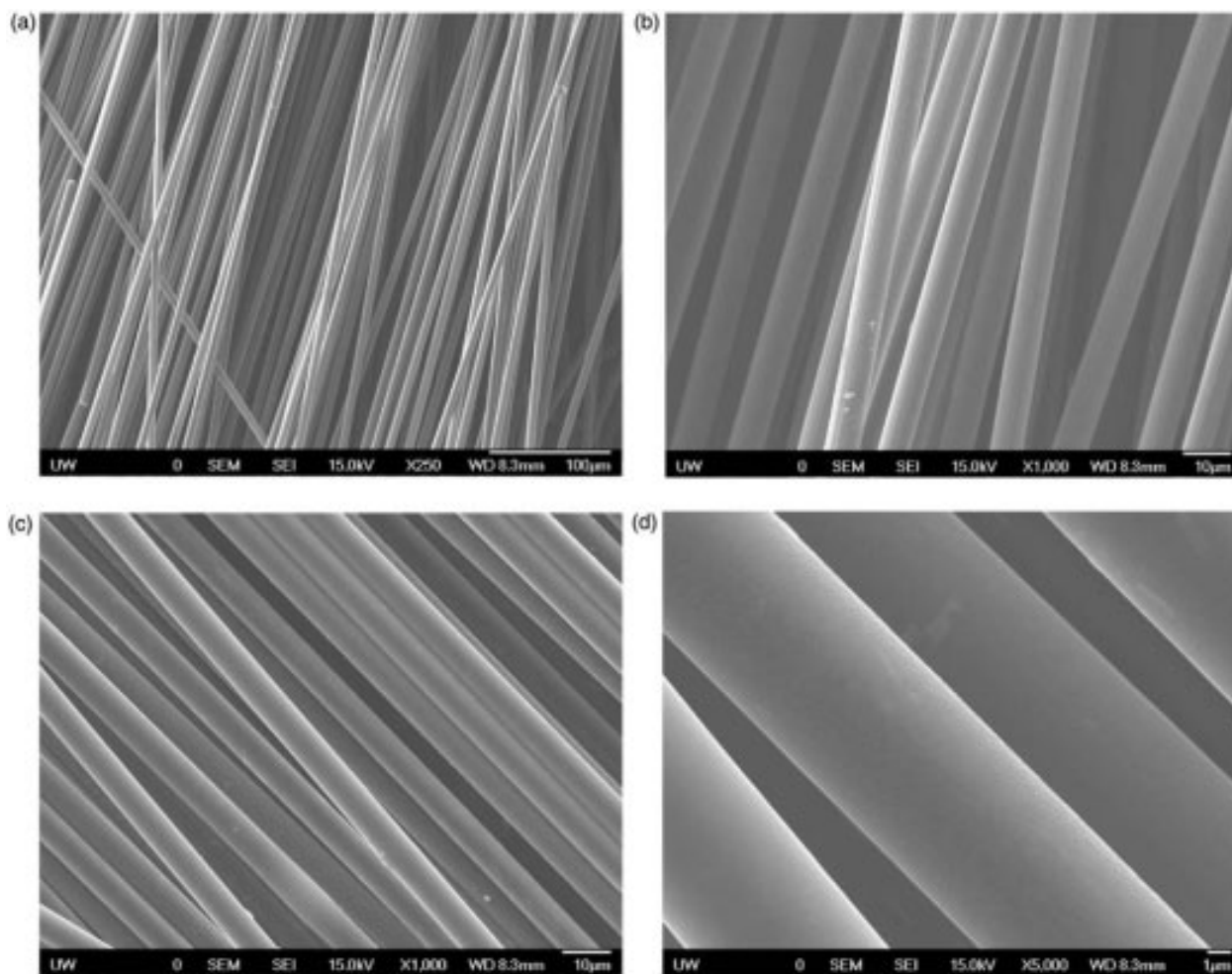
### *Comparative evaluation of the physical properties*

Nondestructive inspection is performed on 100% of the plates (Table 2) by means of pulse-echo ultrasound using a C-scan system with a 5 MHz sensor. Typical amplitude images are shown in Figure 9(A) and (B) for families A and R, respectively. The multichromatic scale ranges from red, which indicates no signal loss, to blue, or complete loss of signal. The signal threshold is set at 6 dB loss, which corresponds to 80% of the emitted signal. Family A panel indicates a very homogeneous structure, with yellow/orange/and red coloration (Figure 9(A)). Family R panel is comprised of a diffused mix of colors (Figure 9(B)), which indicates a highly heterogeneous structure. The signal is noisy and difficult to interpret compared to family A. With ultrasonic inspection, any discontinuity in material properties, such as the presence of a defect, thickness transition, or change of modulus is characterized by an

attenuation of the signal. For this material form, there are point-to-point variations in material properties due to the random fiber distribution and the presence of resin-rich areas, which generate signal attenuation often not corresponding to the presence of a traditional defect (void or porosity). Therefore, it becomes difficult to isolate the presence of a small defect from the background noise. These observations are consistent with the ones observed by the authors for chopped fiber systems for compression molding.<sup>17,18</sup>

Two micrographic coupons are extracted for each family (Table 2) in order to better understand the quality of the infused panels as well as the overall meso-structure. The microscopic specimen is first vacuum-impregnated with the epoxy, then pressure-cured to minimize formation of air bubbles. The specimen is polished with a six-step process: 180 grit, 600 grit, 1200 grit, 9-micron, 3-micron silk, and 3-micron nonnap polyester, ending up with 1 h polishing with a nonnap polyester cloth and 10% alumina solution.<sup>19</sup> A representative





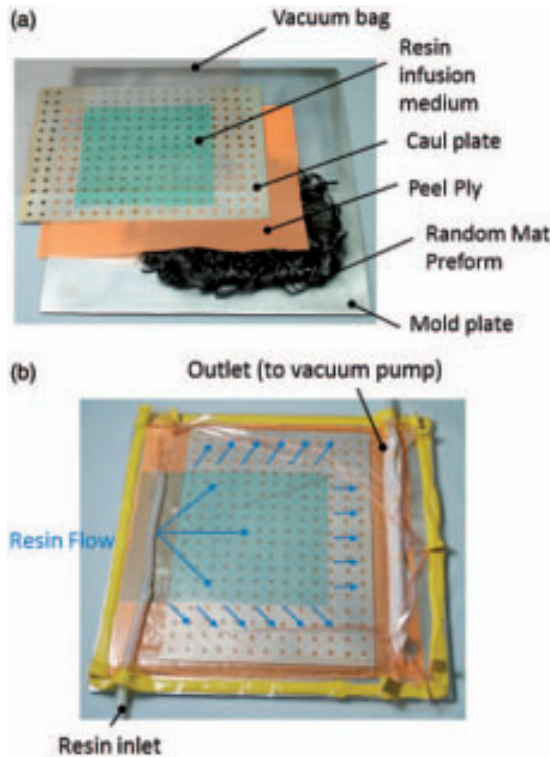
**Figure 6.** Scanning electron microscope (SEM) images (at different levels of magnification) of recycled carbon fibers show a clean and smooth surface, with very limited traces of resin or coating residue: (A)  $\times 250$ , (B) and (C)  $\times 1000$ , and (D)  $\times 5000$ .

cross-section of a panel of family A (Figure 10(A)) shows an ordered distribution of the plies and tows, tight ply compaction, low resin content, as well as absence of macro-voids (entrapped air). The cross-section of the panel of family R (Figure 10(B)) shows uneven distribution of fibers and resin, a relatively planar distribution of fibers (two-rather than three-dimensional), odd-shaped groupings of carbon fibers, and a much higher resin content. On the other hand, there are very few voids, of small size, indicating a successful infusion process.

From the composite panels, specimens weighing only 1 g are evaluated for fiber volume content by means of ASTM standard D3171 for acid digestion<sup>16</sup> (Table 2). After measuring the volumes of the composite specimens by immersion in distilled water, the specimens are subjected to a procedure essentially identical to the one used to recycle the fibers. The residue is cooled down in cold water and filtered in 15  $\mu\text{m}$  ceramic filters using a vacuum pump to separate the liquid,

which is collected in a catcher. After desiccation, the left over fibers are weighed to measure the remaining volume. The difference between the composite specimen volume before acid digestion and the volume of the leftover fibers gives a measure of the resin volume content. It should be emphasized that this process is very slow and lengthy, and selection of the appropriate filters as a compromise between process time and measurement accuracy is key. The average values for the four combined specimens extracted from families A and B indicate a fiber volume content  $V_f = 62 \pm 1\%$ , while the average of the five measurements for family R specimens shows a fiber volume content  $V_f = 33 \pm 2\%$ . The results are summarized in Table 3. The volume content for the recycled panel is extremely low, and future research should aim at improving the fiber volume content. This can be achieved by first disentangling and rearranging the mass of fibers after the recycling operation with a series of rakes, thus generating a more



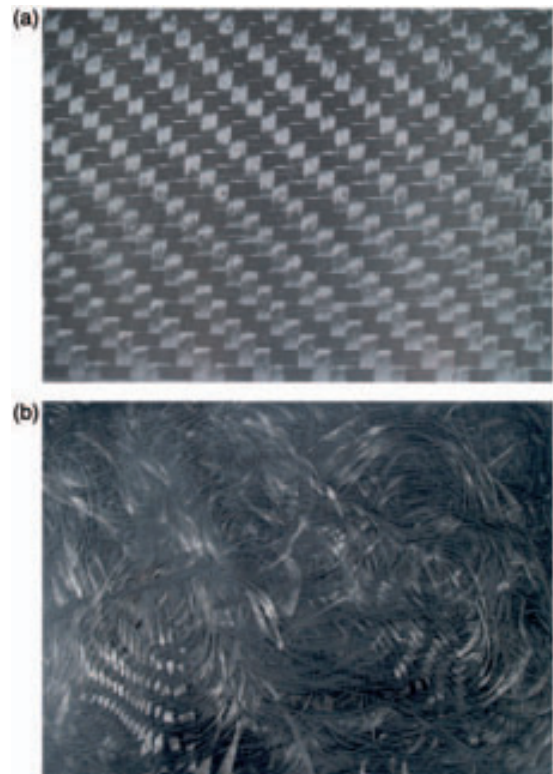


**Figure 7.** Vacuum-assisted resin transfer molding (VaRTM) materials utilized in this study (A), and setup under vacuum at infusion (B), also showing a sketch of the progression of flow.

uniform mat. Second, during the preforming operation, by reapplying a binder (tackifier) and thermoforming, the recycled mat in a heated press. These preforming operations will lead to a higher fiber volume content and more uniform panel thickness distribution after curing.

### Comparative evaluation of the mechanical properties

The panels obtained from recycled fibers have a random fiber distribution, which imparts them in-plane quasi-isotropic properties similar to the chopped fiber systems<sup>17</sup> used for compression molding. Although this assumption has not been verified experimentally in this study, the following discussion refers to tests performed on coupon machined in the 0 direction, but applies to all four principal directions. From the molded panels, tensile specimens are machined to the ASTM D3039<sup>20</sup> straight-sided rectangular geometry with dimensions  $304.80 \times 38.10 \text{ mm}^2$ . Since glass/epoxy fabric tabs of length 50.8 mm are bonded to the specimen using 3M Scotch-Weld film adhesive, the effective gage length is reduced to 203.2 mm. Compression specimens have the same geometry, but are left untabbed and placed in the ASTM D6484



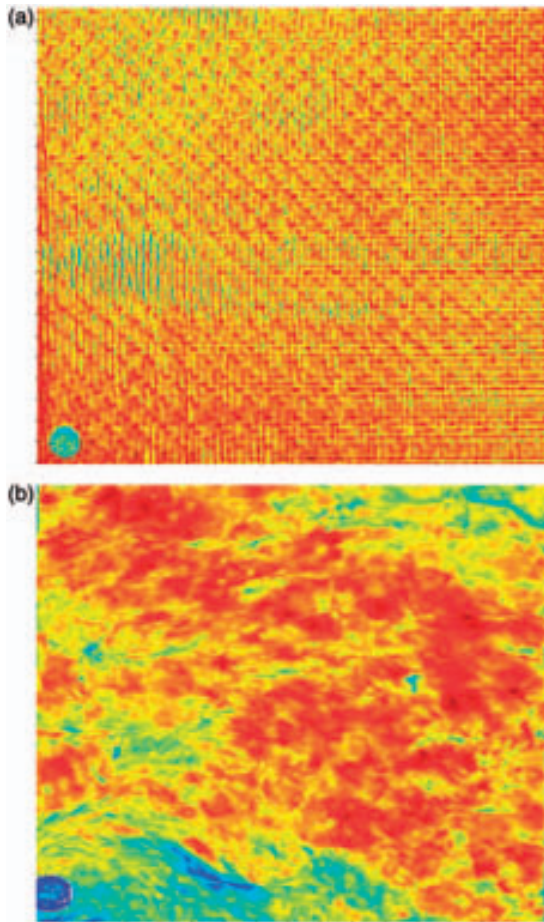
**Figure 8.** Visual aspects of the mold side of a VaRTM panel manufactured using the pristine twill reinforcement (A) and the one using the recycled long random fiber reinforcement (B).

compression fixture,<sup>21</sup> typically used for open-hole compression tests but used by Boeing for unnotched compression tests as well.<sup>19</sup> Flexure specimens are machined to ASTM D790<sup>22</sup> three-point bend flexure tests specifications. It should be noted that the support span  $s$  for the three-point bend fixture is determined to be  $s = 40 \times t$ , where  $t$  is the specimen thickness. While for traditional composites, the error introduced by keeping the span constant based on the panel average thickness, for the recycled panel this assumption could lead to gross errors. It is therefore necessary to painstakingly change the span for each specimen in order to maintain the constant  $s/t$  ratio. Finally, the short beam shear specimen is machined to the specifications of the ASTM D2344 standard,<sup>23</sup> using an  $s/t$  ratio of 4.0. All specimens are loaded at a rate of 1.3 mm/min in a 2-grip Instron hydraulic tension/compression test frame. Modulus is measured using a 25.4 mm gage extensometer. A total of five repetitions per family per test type are tested (Table 2).

Tensile modulus results show that family R retains approximately 55% of the modulus of the quasi-isotropic pristine twill material (Figure 12(B)). Specimens

**Table 2.** Summary of tests performed and repetitions

Family	C-scan	Microscopy	Acid digestion	Tension	Compression	Three-point bend flexure	Short beam shear
A	3	2	2	5	5	5	5
B	3	2	2	5	5	5	5
R	3	2	5	5	5	5	5



**Figure 9.** C-scan images of a vacuum-assisted resin transfer molding (VaRTM) panel manufactured using the pristine twill reinforcement (A) and the one using the recycled long random fiber reinforcement (B).

of family R fail in tension by shear of the intertwined tows, which pull out from the matrix (Figure 11(A) and (B)). This resin-dominated failure mechanism is typically observed in discontinuous fiber systems, such as the chopped carbon fiber SMC discussed by the authors in ref. 17. Some fiber breakage is observed, but the main failure mechanism is not fiber-dominated. After failure, the specimen retains partial integrity and remains somewhat intact. On the other hand,

specimens of families A and B fail by traditional brittle fiber fracture, with a clean, more defined fracture front which divides the specimen in two halves. Tensile strength results, reported in Table 3 and Figure 12(A), show that family R retains approximately 25–30% of the tensile strength of families A and B. Although the as-measured results indicate that the recycled material form is much less performing than the pristine form, these results should be interpreted carefully. First of all, the failure mechanisms are completely different between family R and families A and B. For the recycled fibers, failure is matrix-dominated, while for twill fibers it is fiber-dominated. Second, the fiber volume content of family R specimens is nearly half that of families A and B. With these considerations in mind, it becomes important to understand that with improved preforming operations, it is possible to obtain a more homogeneous recycled fiber mat, with fewer thickness variations and higher fiber volume content. If the results for family R ( $V_f = 33\%$ ) and families A and B ( $V_f = 62\%$ ) are normalized<sup>24</sup> to a nominal fiber volume content of 50%, the results for the recycled material become more appealing, with a tensile strength of approximately 50% (Figure 12(A)) and a modulus of over 100% (Figure 12(B)) that of the pristine material. Unfortunately, the normalization process in this context cannot be used in such straightforward manner. While tension and compression properties are fiber-dominated for families A and B, they are not so in the case of family R. Therefore, it is not correct to use the standard normalization process, in the same way that it cannot be used for other nonfiber-dominated properties, such as flexure, or short beam shear.<sup>24</sup> However, it is expected that increasing the fiber volume content and improving the homogeneity of the preform will result in fewer and smaller resin-rich areas, which are known to be the locations of failure initiation for chopped fiber systems.<sup>17,18</sup> With that in mind, and based on the experience of the authors, it is more likely that we can expect a tensile strength of 240–270 MPa and a modulus of 38.0–40.0 GPa if a fiber volume content of 50% is achieved. Compression failure consists of a similar shearing mechanism as the one seen in tension (Figure 13), but takes the advantage of the higher resin compressive strength over the resin's tension and shear strengths.

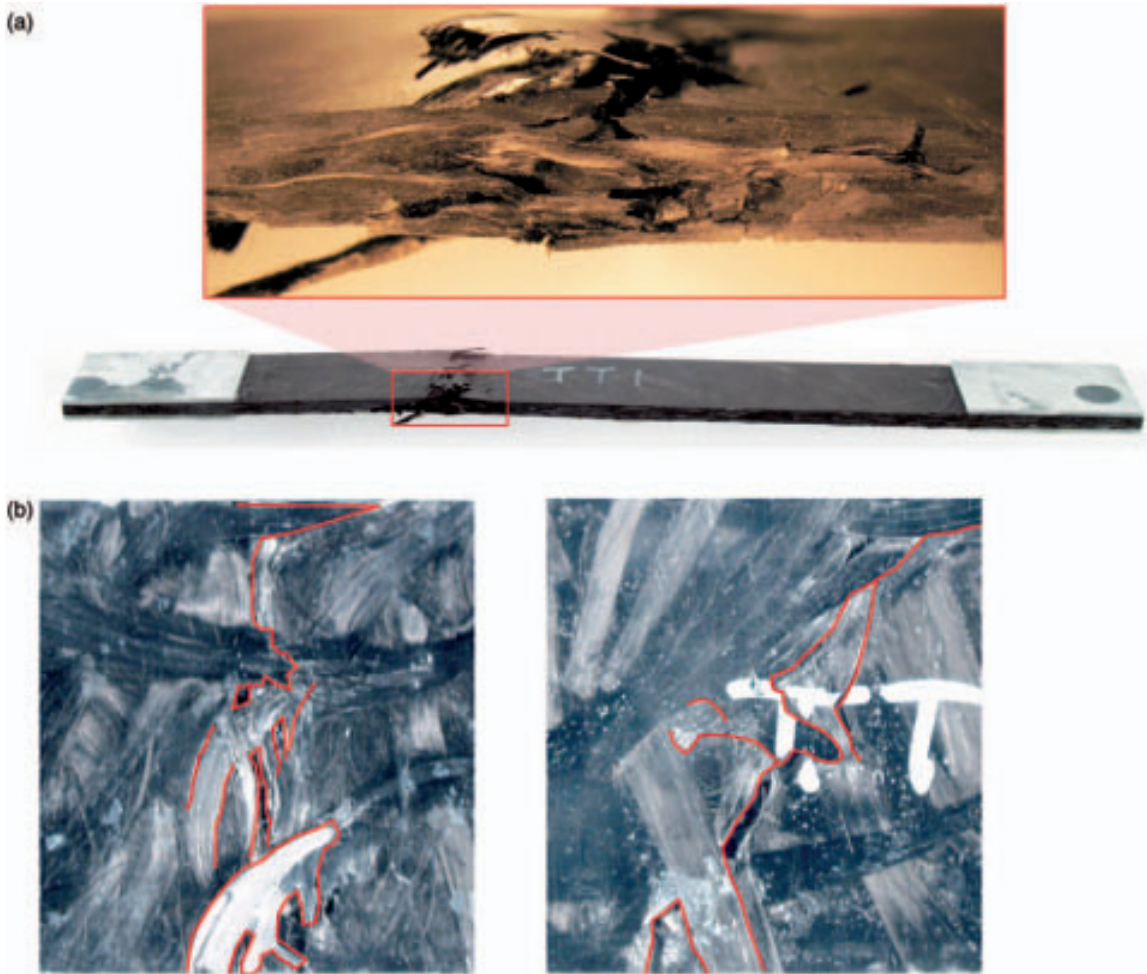


**Figure 10.** Optical microscopy images of a vacuum-assisted resin transfer molding (VaRTM) panel manufactured using the pristine twill reinforcement (A) and the one using the recycled long random fiber reinforcement (B).

**Table 3.** Summary of test results – unnormalized

Family	Test type	Average thickness (mm)	Average fiber volume (%)	Strength (MPa)	CoV (%)	Modulus (GPa)	CoV (%)
A	Tension	4.0	62	758.4	0.7	51.0	1.5
	Compression	4.0	62	450.9	1.1	46.2	3.1
	Flexure	4.0	62	697.0	3.3	–	–
	Short beam shear	4.0	62	50.3	6.3	–	–
B	Tension	4.0	62	715.7	1.7	49.6	0.9
	Compression	4.0	62	422.6	4.0	45.5	2.3
	Flexure	4.0	62	522.6	3.6	–	–
	Short beam shear	4.0	62	52.4	4.7	–	–
R	Tension	4.3	33	196.5	3.2	29.0	15.8
	Compression	4.3	33	190.3	5.9	27.6	9.5
	Flexure	4.3	33	365.4	16.0	–	–
	Short beam shear	4.3	33	37.2	3.4	–	–





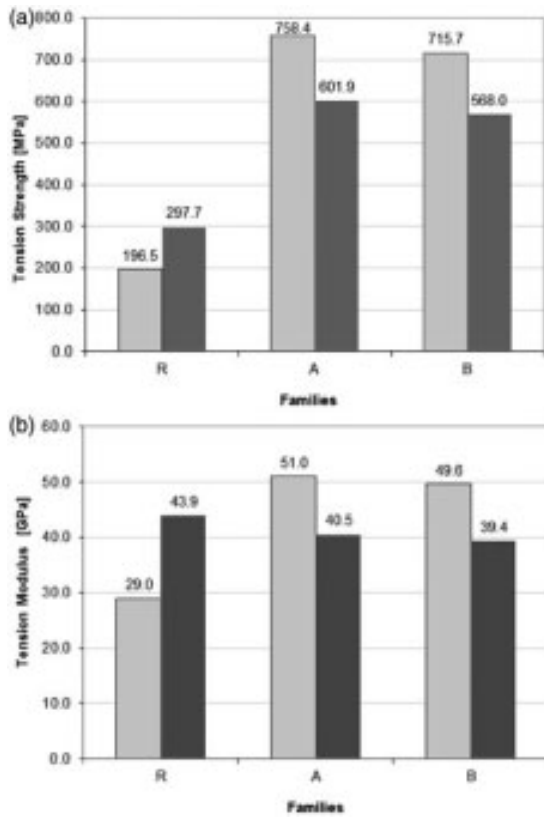
**Figure 11.** (A) Postfailure image and details of tensile specimen extracted from a vacuum-assisted resin transfer molding (VaRTM) panel manufactured using the recycled long random fiber reinforcement and (B) surface details showing failure morphology by shearing and pull-out of tensile specimen.

As-measured compression strength results, reported in Table 3 and Figure 14(A), show that family R retains approximately 42% of the compressive strength of families A and B. It is typical for resin-dominated material systems, such as chopped carbon fiber SMC, that compressive strength is equal or greater than tensile strength,<sup>17,25</sup> which is in direct contrast with the typical behaviors observed for unidirectional tapes or fabrics, whereby tension strength always greatly exceeds the compressive strength. Compressive modulus results show that family R retains approximately 60% of the modulus of the quasi-isotropic pristine twill material (Figure 14(B)). Normalization to a nominal fiber volume content  $V_f = 50\%$  would suggest that the compressive strength for family R would reach 80% of the strength of families A and B, while the modulus would exceed 100% of the pristine twill (Figure 14(A) and (B)). Following the same considerations, discussed for tension, it is expected that a more realistic value for

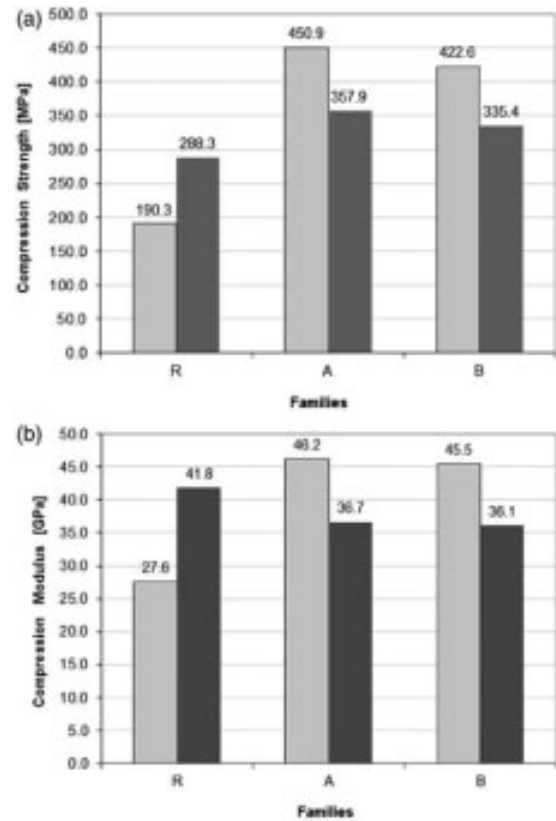
compressive strength for this material following an improved preforming operation would lead to a compressive strength of 250–280 MPa and a modulus of 38.0–40.0 GPa if a fiber volume content of 50% is achieved.

Flexural failure manifests on the tensile side, and propagates as a shear crack up through the thickness toward the compressive side (Figure 15). Three-point bend flexure strength results for family R are 70% and 52%, respectively, of the flexural strengths of families A and B, respectively (Figure 16). The flexure strength of the stacking sequence with the (0/90) ply on the outer surface is expectedly higher than the one with the ( $\pm 45$ ) ply. Normalization cannot be performed for flexure strength, according to MIL-HDBK-17 guidelines,<sup>24</sup> but the authors expect that flexure strength will increase significantly if the fiber volume content is increased. It is expected that an average flexural strength of approximately 480–500 MPa should be

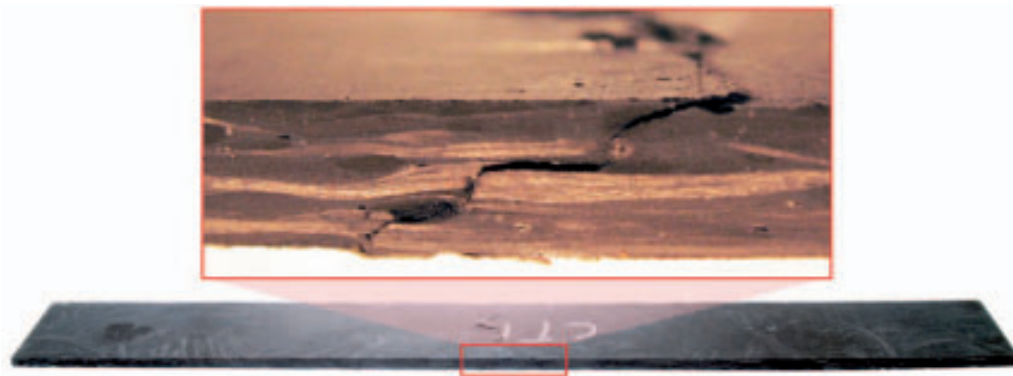




**Figure 12.** Tension strength (A) and modulus (B) results for all three families. For each family, the light bar represents the as-measured values, while the dark bar represents a ‘normalized’ value based on a nominal fiber volume content of 50%.



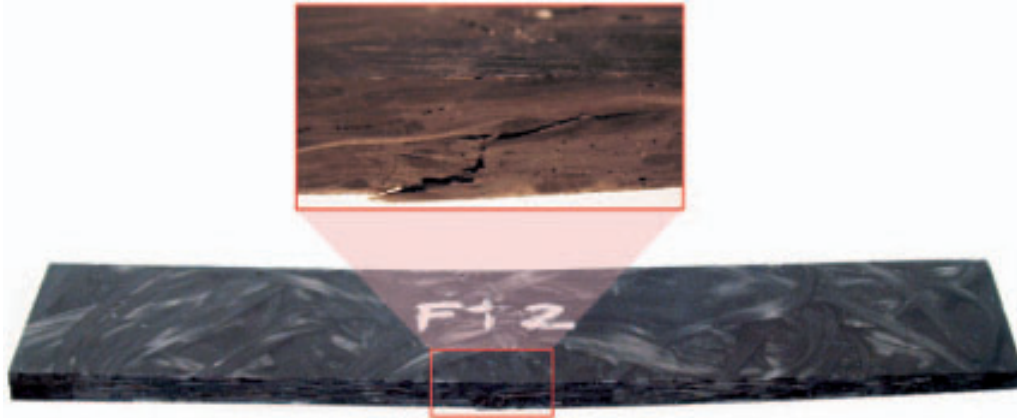
**Figure 14.** Compression strength (A) and modulus (B) results for all three families. For each family, the light bar represents the as-measured values, while the dark bar represents a ‘normalized’ value based on a nominal fiber volume content of 50%.



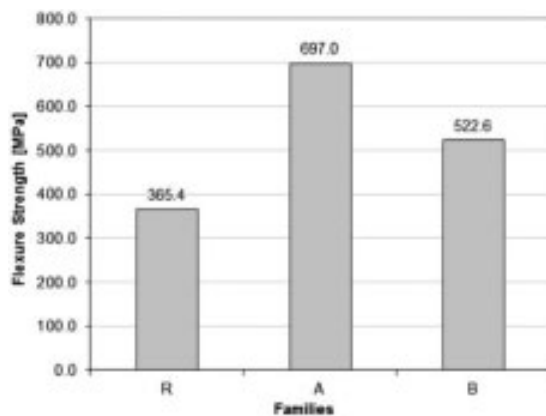
**Figure 13.** Postfailure image and details of compressive specimen extracted from a vacuum-assisted resin transfer molding (VaRTM) panel manufactured using the recycled long random fiber reinforcement.

achieved with a fiber volume content of 50%. Interlaminar shear strength for family R (Figure 17) is 70% of the strength of families A and B. It is typical of random fiber systems to have short beam shear strength lower than the continuous fiber system made

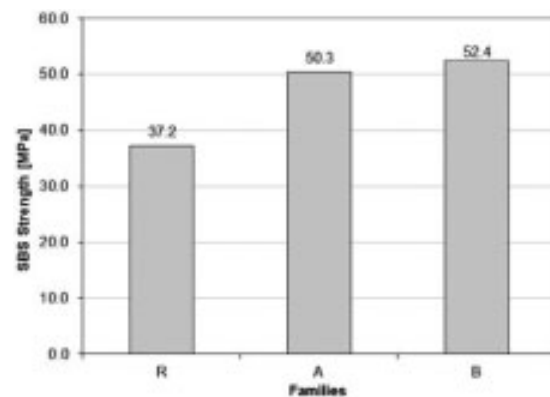
of the same fiber and resin system.<sup>17,25</sup> This is justified by the absence of well-defined interlaminar planes, as well as the pre-existing kinking of the ‘plies,’ which leads to premature slipping of the ‘plies’ with respect to each other. Increasing fiber volume content will



**Figure 15.** Postfailure image and details of compressive specimen extracted from a vacuum-assisted resin transfer molding (VaRTM) panel manufactured using the recycled long random fiber reinforcement.



**Figure 16.** Measured flexure strength results for all three families.



**Figure 17.** Measured short beam shear strength results for all three families.

likely not improve the short beam shear strength, but a more homogeneous distribution of the reinforcement (more flat and well-arranged recycled fiber mat) may lead to some improvements.

## Conclusions

A recycling process that allows for the separation of carbon fibers from the epoxy matrix was demonstrated. The process is based on immersion in boiling sulfuric acid, similar to the ASTM acid digestion standard test method. The carbon fibers, which originally featured twill architecture, take the appearance of a fuzzy, entangled, random, long-fiber mat following the recycling process. VaRTM liquid resin infusion is used to manufacture panels of both the pristine twill fabric as well as the recycled fiber mat. The panels manufactured using the recycled mat exhibit high thickness variations, and a fiber volume content of 33%, much lower than

the twill at 62%. The preform has lower permeability, and is more difficult to infuse using the VaRTM process, but the end quality of laminates is good in terms of void content. Both physical properties (ultrasonic and micrographic images) and mechanical properties (tension, compression, flexure, and short beam shear) of the recycled panels show trends and traits similar to those of advanced carbon fiber SMC. However, with further development efforts, it is possible to realign the recycled long fibers and re-establish their original performance. Furthermore, by optimizing the degree of compaction and homogenization of the recycled fiber mat, it will be possible to increase the fiber volume content to values around 45–50%, and dramatically reduce the thickness variation. Finally, though VaRTM is a suitable process for liquid resin infusion of the preform, a pure RTM process will lead to further increases in the quality of the laminate thanks to the higher injection pressures and control of both inner and outer mold lines.

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