A UNIDIMENSIONAL FLOW AND CONSOLIDATION CHARACTERIZATION OF RECYCLED PREPREGS

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ABSTRACT

Aerospace composites manufacturers discard anywhere from 30-50% of their prepreg fabric during the ply-cutting phase of manufacturing. This waste is mainly attributed to nesting pattern inefficiencies caused by limited production schedules, fixed prepreg roll widths, limited material out-life, etc. By recovering high-value prepreg offcuts directly from the ply cutter and converting them into long discontinuous fibre moulding compounds, it may be possible to produce complex geometry parts capable of serving as secondary or tertiary aircraft structures. Unfortunately, prepreg materials are tailored to exhibit high levels of percolation, or bleeding flow, during processing. This is in contrast to typical compression moulding compounds which are required to deform in shear, where fibre and matrix move together in order to properly fill a mould cavity. If recovered prepregs are to serve as an alternative to virgin compression moulding compounds, an understanding of the relationship between material flow and processing parameters such as temperature, pressure, and strain rate is needed. The work presented here is an attempt to use a one-dimensional compaction test, previously used to characterize the flow behaviour of prepreg tapes, to study the flow of two recycled materials: a recovered autoclave prepreg fabric and a bulk moulding compound made with recycled carbon fibre. Non-uniform shear flow was observed in tested specimens via optical microscopy, which led to a modified treatment of the strain analysis. The inextensibility of the fibre reinforcement was found to restrict shear deformation and mask any dependence on resin viscosity when oriented close to the flow direction.

1 INTRODUCTION

1.1 Background

The amount of prepreg wasted during the ply cutting phase of manufacturing varies from company to company and it is driven by a combination of production schedule length, production volume and diversity, roll width, ply orientations, and software nesting capabilities. In the last several years, multiple industrial aerospace sources have estimated overall ply-cutter waste at 30 – 50%, all of which is currently sent to the landfill.

The literature regarding the recycling of prepreg materials is substantial; however, it focuses on fibre reclamation through pyrolysis, mechanical comminution, or chemical solvolysis [1-4] and relegates the matrix to low value chemical feedstock, or filler. Compression moulding of recovered prepreg strands into highly featured parts is an alternative recycling method, which makes use of both fibre and matrix. It is a process which has been studied at length using virgin precursors with both thermoset and thermoplastic matrix materials [5-16]. In most cases, unidirectional tapes have been preferred, as they exhibit better formability; however, fabrics must be considered a priority in the context of recycling, as they represent the majority of the waste generated [16, 17]. With this restriction, comes two obvious processing challenges. First, inextensible fibres in both 0° and 90° directions will promote fibre locking and hinder bulk material flow through shear. Second, relatively low resin viscosities, a hallmark of autoclave prepregs, promotes resin transport through percolation and may lead to excessive resin loss and high void contents. Both of these challenges are showcased in Figure 1.

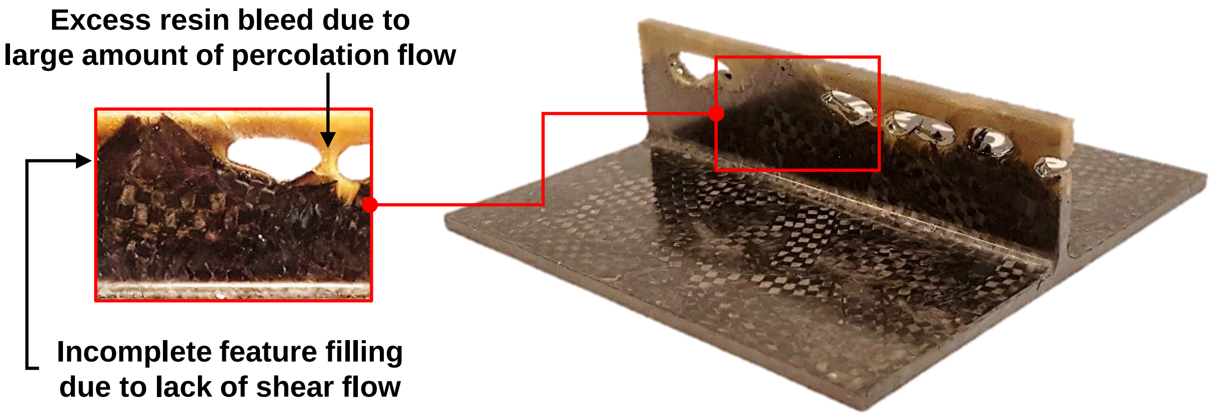

Figure 1. Rib-stiffened compression moulding panel made from recycled prepreg strands having experienced excessive resin loss through percolation and insufficient filling of the rib feature with fibre through shear.

A simple and effective way to characterize the flow behaviour of these materials is needed if complex parts are to be manufactured within a reasonable cycle time and with optimal mechanical performance. To this end, the authors draw on the work of Hubert [18] who, while studying prepreg tapes, proposed that laminate consolidation involves two coupled and competing flow mechanisms: percolation, where resin flows relative to a static fibre bed, and shear flow, where fibre and resin flow together as a viscous fluid. Hubert [18] carried out compaction tests on prepreg laminates with two different resin systems and for a variety of layup configurations and cure temperatures (Figure 2). While these tests shed some light on the particular importance of resin viscosity and fibre orientation to the prevalence of each flow mechanism, they were not able to determine a comprehensive understanding of this relationship.

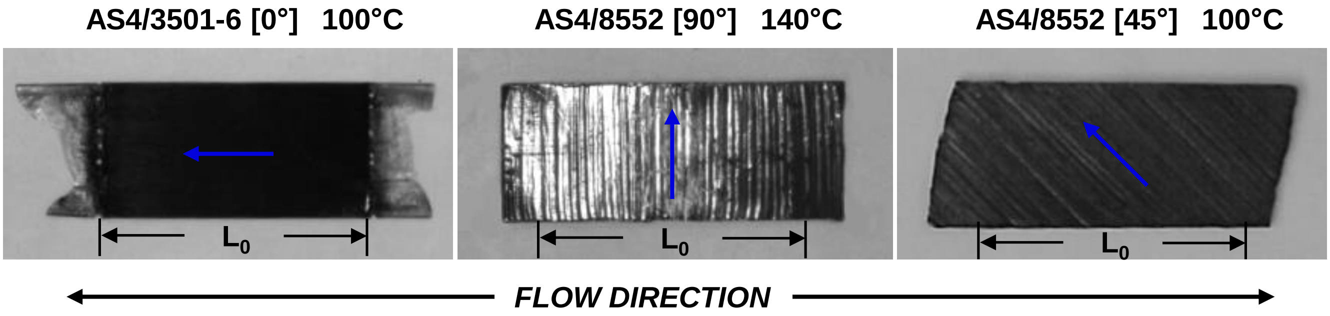

Figure 2. Prepreg tape samples tested using a 1-D compaction channel. Different resin systems, testing temperatures, and layup orientations showcase different degrees of percolation and shear flow (adapted from Hubert, et al. [19]).

1.2 Objectives

The work presented here attempts to address this gap in the processing-related literature for two recycled materials: A recovered autoclave prepreg fabric and a bulk moulding compound (BMC) consisting of a commercial epoxy and recycled carbon fibre (rCF) reinforcement. This papers objectives include:

- Apply compaction experiment in [18] to recycled prepreg and BMC.
- Determine technique suitability for prepreg fabric and BMC characterization and propose modifications.
- Determine the effect of moulding temperature on shear and percolation flow of recycled materials.

2 EXPERIMENTAL METHODS

2.1 Apparatus

The compaction fixture used in this research is shown below in Figure 3 on the left hand side. It consists of a steel channel, heated copper platens, insulating blocks and steel base plates and has been mounted on a tabletop mechanical testing frame (Insight 5kN, MTS Corporation). This system is able to reproduce temperatures up to 200 °C, compaction forces up to 5000 N, as well as a broad range of displacement speeds. As mentioned in the previous section, specimens are restricted by the walls of the steel channel, which simplifies the deformation analysis to one dimension. This configuration, as well as the two distinct flow mechanisms to be observed, are illustrated schematically on the right-hand-side of Figure 3. Notice that both percolation and shear flow cause a reduction in specimen height; however, only shear flow cause an expansion in the x-direction. This assumption is important and will be revisited in later sections.

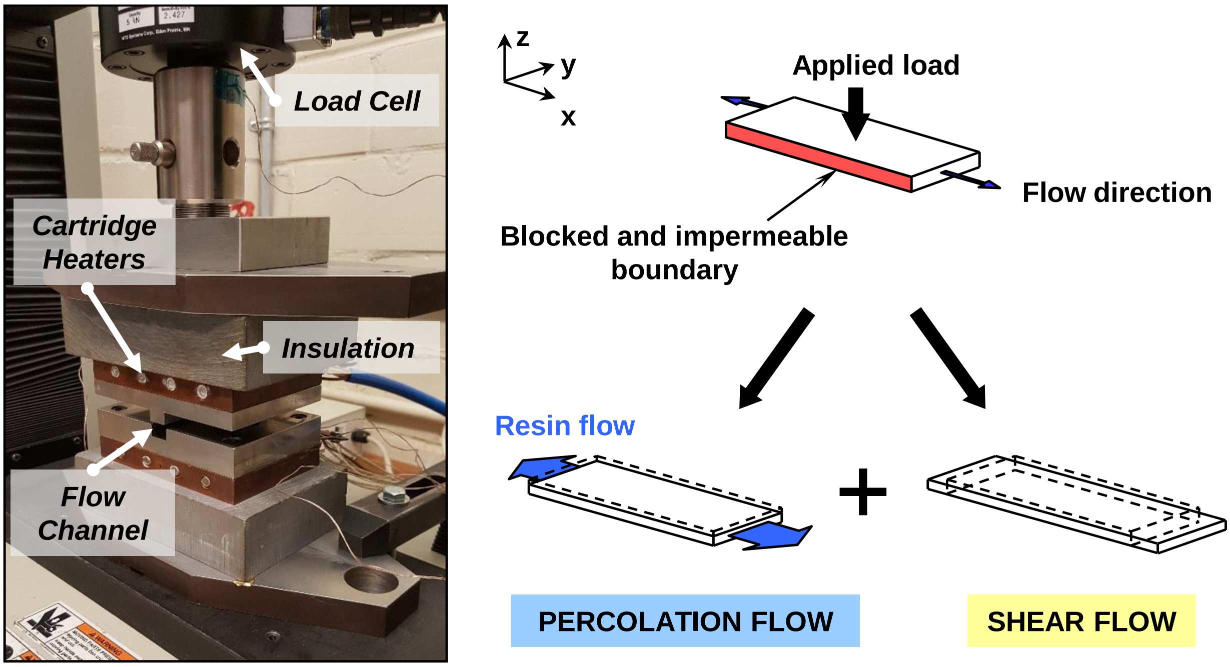

Figure 3. Compaction fixture (left) and specimen deformation schematic adapted from [18] featuring decoupled flow mechanisms (right).

2.2 Materials & Specimen Preparation

The first material studied here is an autoclave prepreg recovered directly from the ply-cutter of an aerospace manufacturer and is within its specified out-life and shelf life. The material itself is produced by Cytec Solvay Group with a matrix of Cycom®5276-1 and a recommended cure temperature of 177 °C (350 °F). The reinforcement consists of a THORNEL® T650 PAN-based fibre with a 3k tow size woven into a 371 gsm eight harness satin (8HS) architecture. The resin content, room temperature out-life, and cured resin density are specified as being 42%, 15 days, and 1.29 g/cc respectively.

The second material studied is a BMC consisting of the commercially available epoxy system Dow DER-331 combined with CVC Omnicure BC-120 hardener and rCF reinforcement from Carbon Conversions (formerly MIT-RCF, LLC). For each batch, 100 parts resin (DER-331), 8 parts hardener (BC-120), and 100 parts rCF were combined by weight. The rCF reinforcement was specified as having a 6.35 mm (0.25 in.) fibre length; however, the process by which the fibres were recycled is unknown. The recommended moulding temperature for this system is 140 °C (284 °F).

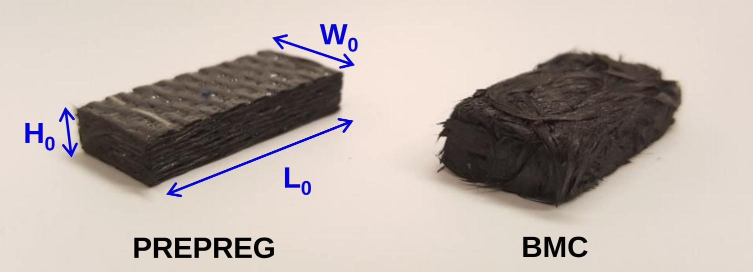

Figure 4. Laminated prepreg stack (left) and BMC preformed charge (right) shown prior to testing.

All prepreg specimens tested were 25.5 ± 0.5 mm x 12.5 ± 0.1 mm x 5.0 ± 0.1 mm (L_0 x W_0 x H_0) with a layup of $[0_8]_T$. Each BMC specimen was preformed to approximately the same dimensions using a 1.85 g charge of material and a custom-made aluminum jig (Figure 4). This jig was left in the freezer at approximately – 18 °C for a minimum of one (1) hour, so that samples, once extracted, would maintain their shape prior to testing. It is important to note that these specimens were not sealed in an airtight bag while in the freezer and some condensation was observed prior to testing.

2.3 *Test Matrix*

The test matrix carried out as part of this investigation is summarized below in Table 1. As an initial benchmarking of the 1-D compaction method, the matrix focuses exclusively on the impact of moulding temperature on the degree of percolation and shear flow.

Material	Temperature	Applied Load	Closure Rate	Repetitions
T650/5276-1(8HS)	100, 120, 140, 160, 180 °C	4000 N	0.025 mm/s	3
rCF/DER-331(BMC)	90, 100, 110, 120, 130 °C	4000 N	0.025 mm/s	3

Table 1. Compaction study test matrix

While past studies have indicated that 70 atm may be adequate to properly fill a highly featured mould using prepreg tapes [20], prepreg fabrics have been shown to flow much less in general. For this reason, the applied load was selected to correspond to an initial consolidation pressure of approximately 120 atm given by Equation 1. Finally, the closure rate was made intentionally slow to ensure test stability.

$$P_{applied_i} = \frac{F_{applied}}{A_{sample_i}} = \frac{F_{applied}}{L_0 \times W_0} \tag{1}$$

2.4 *Benchmark Deformation Analysis*

Hubert [18] chose to quantify percolation as a volumetric loss of resin, measured by weighing a specimen before and after testing, having manually removed excess resin from the specimen edges. This volumetric strain is described in Equation 2, where L_0, H_0, W_0, and M_0 are the specimen initial dimensions and mass. M, L, and ρ_{resin} are the final mass, final length, and cured resin density respectively.

$$\varepsilon_v = \frac{1}{\rho_{resin}}\left(\frac{M - M_0}{L_0 H_0 W_0}\right) \tag{2}$$

Shear flow was assessed using measured specimen displacements and the Green strain tensor evaluated in the x-direction (Equation 3). For additional detail and derivation, see Karasudhi [21].

$$e_x = \frac{1}{2}\left[\left(1 + \frac{L - L_0}{L_0}\right)^2 - 1\right] \tag{3}$$

The contribution of each flow mechanism to a specimen's total vertical deformation, denoted e_z, was approximated by applying the assumptions listed below to the volumetric strain of a specimen described by Equation 4, as well as physical constraint imposed by the fixture walls such that $e_y \approx 0$. This provides a description of both percolation and shear flow contributions in Equation 5 and Equation 6 respectively.

Percolation Flow Assumptions

1. Resin movement occurs relative to the fibre bed
2. Non-zero resin mass loss ($\varepsilon_v \neq 0$)
3. No deformation of the fibre bed occurs in the x-y plane ($e_x \approx 0$ and $e_y \approx 0$)

Shear Flow Assumptions

1. Resin and fibre bed move together
2. No resin mass loss (material is assumed to be incompressible and $\varepsilon_v = 0$)
3. Non-zero deformation of the fibre bed occurs in the x-y plane ($e_x \neq 0$ and $e_y \neq 0$)

$$\frac{\Delta V}{V_0} = \varepsilon_v = \left(e_x + e_y + e_z\right) + \left(e_x e_y + e_x e_z + e_y e_z\right) + e_x e_y e_z \tag{4}$$

$$e_z^P = \varepsilon_v \tag{5}$$

$$e_z^S = -\frac{e_x}{1 + e_x} \tag{6}$$

2.5 *Analysis Modification*

Unlike the unidirectional tape specimens shown in Figure 2, the fabric and BMC materials tested here deformed heterogeneously, making it difficult to discern resin from composite in certain regions and precise length measurements were not possible using traditional calipers or micrometers. Instead, specimens were imaged on a flatbed scanner at a resolution of 600 dpi and with a strong backlight to highlight the contrast between resin and composite. These images were then fed into a code written in Matlab® (Mathworks), which performed a series of alignment, trimming, and thresholding operations to isolate and measure the average specimen length associated with shear flow (outlined in yellow in Figure 5). A potential shortcoming of this scanning method is the assumption that the specimen thickness remains constant along its entire length, which contradicts observations of a plug-like flow made by Picher-Martel [11]. This assumption was tested using optical microscopy of specimen cross-sections in the x-z plane taken at $y = \frac{W}{2}$.

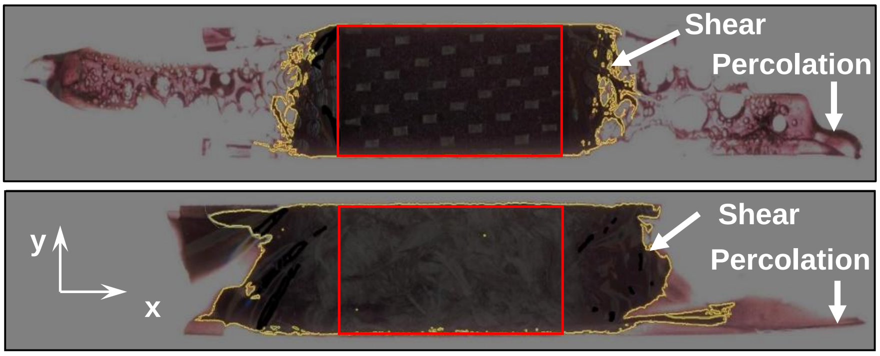

Figure 5. Strain analysis representation for both prepreg (top) and BMC (bottom) specimens. Initial specimen size and shear flow front are highlighted in red and yellow respectively.

These micrographs revealed the existence of a transitional region between the composite and resin portions of each specimen. BMC specimens showed a gradual decrease in fibre volume content at the specimen edge as is shown at the top of Figure 6, as well as a rotation of fibres toward a 90° orientation, which is consistent with transverse shear flow behaviour (see A & B). Prepreg plies, on the other hand, showed very little evidence of shear deformation. That being said, a small amount of 90° fibres at the specimen edge were seemingly pulled away from the ply stack in what is being called fibre jetting (Figure 6 – D).

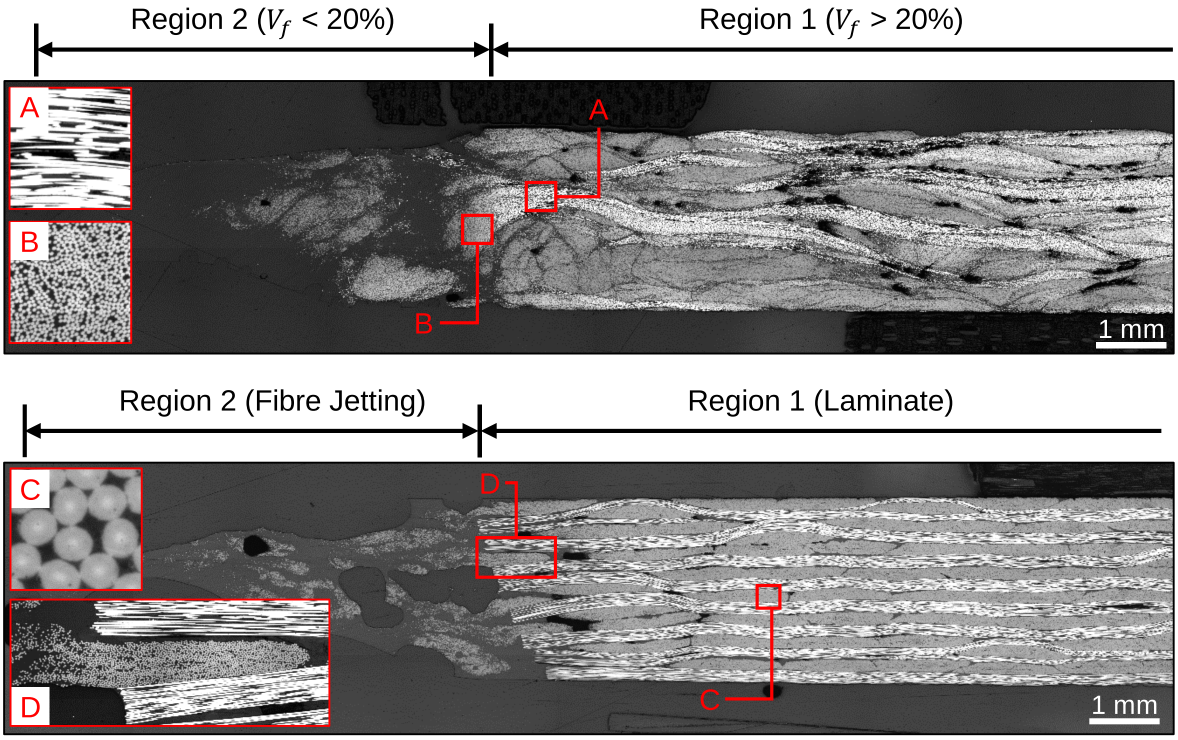

Figure 6. Representative micrographs of rCF/DER-331 BMC (top) and T650/5276-1 8HS (bottom) specimen mid sections. Fibre alignment due to shear flow observed in all BMC specimens (A+B). Fibre diameter measurements made on 90° fibres used to determine number of fibres present in prepreg jetting flow region (C+D).

To adapt the benchmark analysis to suit these new observations, new specimen percolation and shear flow regions were identified as follows. The final length used to compute e_x* for prepreg specimens was taken at the edge of 0° tows as illustrated in Figure 6 by Region 1 (Laminate). BMC specimens were treated in a similar fashion; however, the shear flow region was limited by the point past which the fibre volume content became less than 20%. This bound was chosen as the lower end of fibre volume fractions attainable for compression moulded BMC parts [22]. For both prepreg and BMC, the remaining resin area was used to adjust the percolation strain ε_v*.

3 RESULTS & DISCUSSION

The shear and percolation strains measured for both prepreg and BMC systems are summarized in Figure 7 and Figure 8 below. Strains computed using specimen lengths measured with the flatbed scanner are shown in blue and yellow for both materials. These are denoted as e_x and ε_v and are referred to as original strains, as they were meant to emulate the original technique used in [18]. Strains computed using the micrograph analysis are shown in green and magenta for the prepreg and BMC specimens respectively. These are referred to as the modified strains and are denoted as e_x* and ε_v*. The original strains significantly over predict the amount of shear flow experienced by each specimen. Not surprisingly, they also under predicted the amount of percolation flow for a given specimen, as they do not account for resin found in the transitional regions. Given these shortcomings, it is assumed that the modified strains are a more precise representation of the real specimen deformation and only these results will be discussed further.

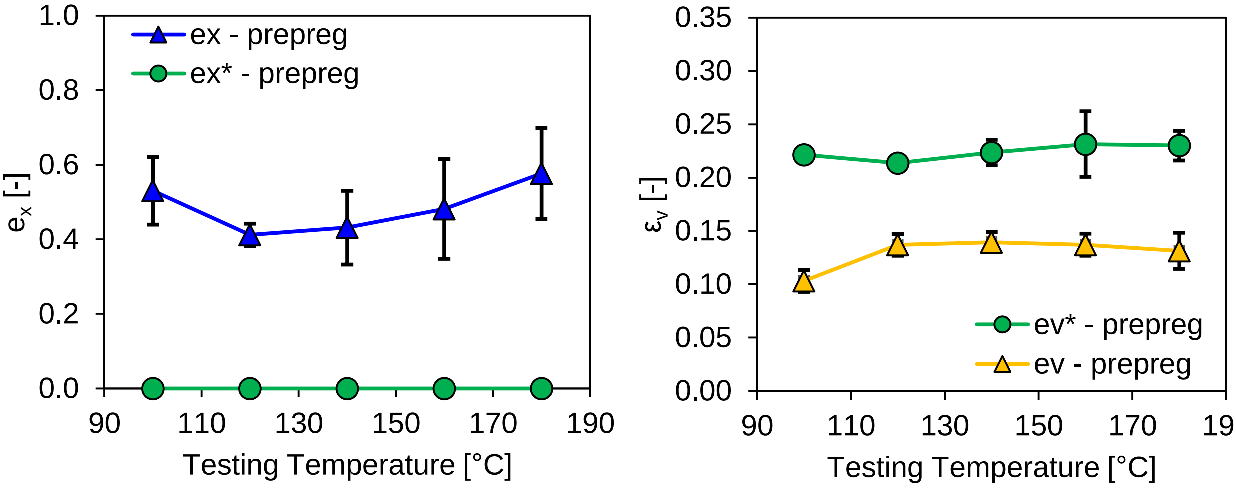

Figure 7. Original (e_x,ε_v) and modified (e_x*,ε_v*) shear strains (left) and percolation strains (right) for all prepreg specimens.

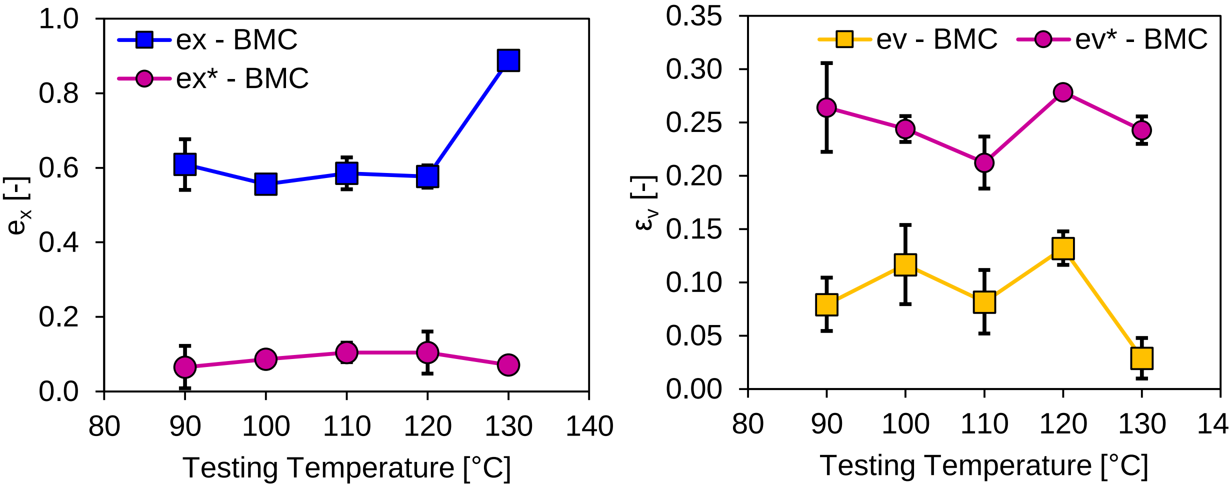

Figure 8. Original (e_x,ε_v) and modified (e_x*,ε_v*) shear strains (left) and percolation strains (right) for all BMC specimens.

Prepreg specimens exhibited no appreciable shear flow across all testing temperatures (Figure 7). Some fibre jetting was observed on all micrographs; however, counting the total number of fibres expelled by the laminate revealed that no more than eight (8) 3k tows were ever displaced. Given that each specimen layup is $[0_8]_T$, it is safe to assume that these fibres belong exclusively to the very first 90° tow in each ply and that the rest of the laminate remains unmoved, locked in place by the 0° tows running along its entire length. The amount of percolation observed for prepreg specimens was fairly insensitive to testing temperature with ε_v^* varying slightly between 0.20-0.25.

Non-zero shear and percolation strains were observed for all BMC specimens; however, once again, no trend was observed in terms of the testing temperature. These observations are reinforced by plotting the contribution of each flow mechanism to the total specimen deformation, which is shown in Figure 9. Note that these contributions were not plotted for the prepreg specimens, as percolation constituted 100% of e_z in all cases.

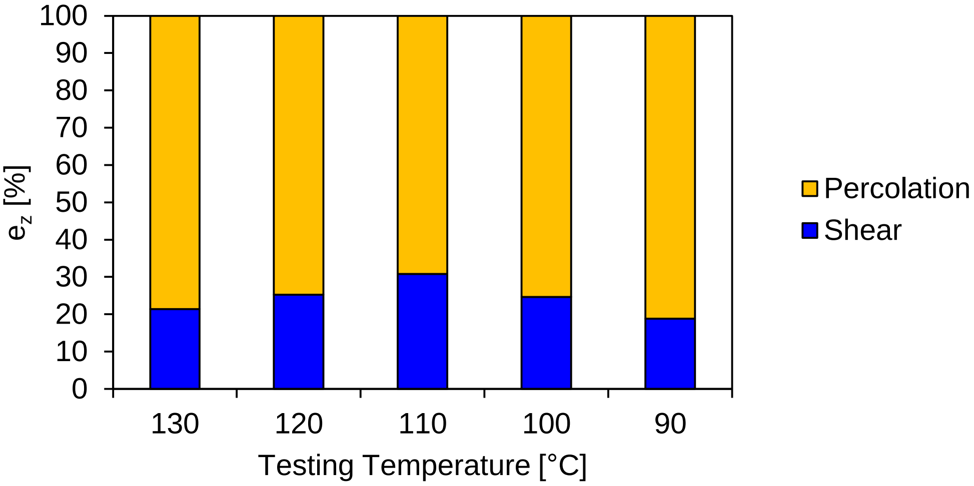

Figure 9. Flow mechanism contributions for rCF/DER-331 BMC calculated using modified strains.

To better understand the temperature insensitivity observed, the percolation and shear strains were compared with those obtained by Hubert [18] for Hercules AS4/3501-6 and AS4/8552 tape laminates. The comparison for shear strain and percolation strain are shown in Figure 10 and Figure 11 respectively.

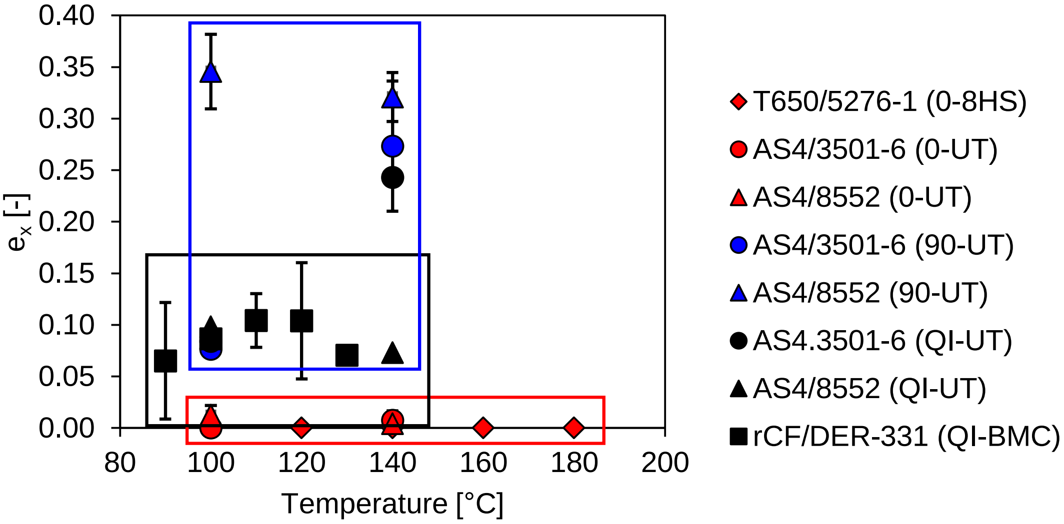

Figure 10. Comparison of shear strain data collected here with previous tests by [18].

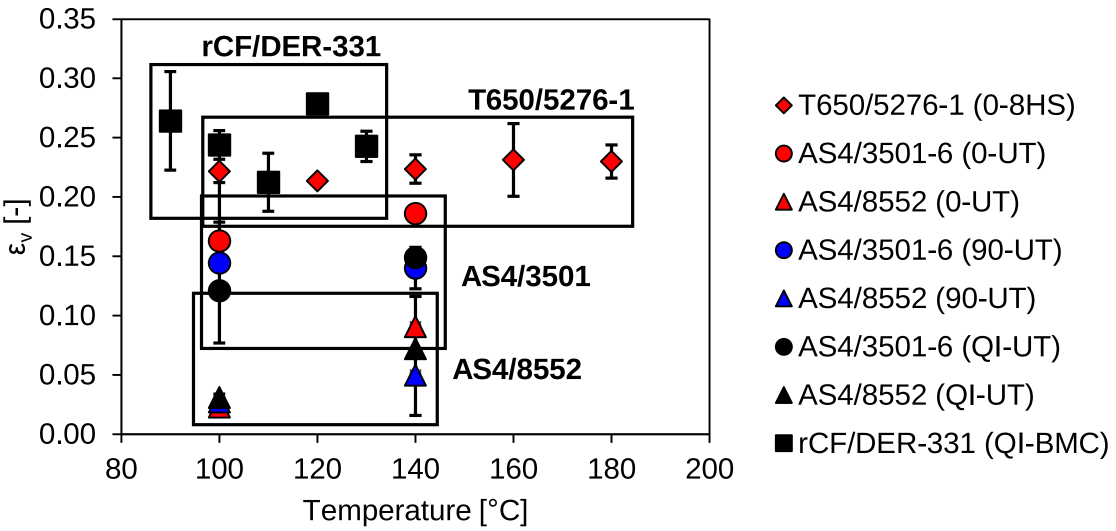

Figure 11. Comparison of percolation strain data collected here and by [18].

In each of these plots, resin systems are assigned a specific marker shape and reinforcement architectures/orientations are assigned a specific colour for clarity. Notice that, in general, fibre orientation limits shear strain as it approaches 0°. Also notice that, as this happens, the effect of temperature and material type, and by extension resin viscosity, are diminished. This is observed as a vertical shortening of the bounding boxes surrounding each orientation group. This is to say that the effect of resin viscosity may only be evident in situations where inextensible fibres are not restricting flow.

4 CONCLUSIONS

In summary, benchmarking was performed on two recycled materials using a one-dimensional compaction characterization methodology. It was revealed that prepreg fabrics and BMCs cannot be studied in the same way as prepreg tapes, due to the non-uniformity of their shear flow behaviour. BMC specimens, in particular, displayed large variations in fibre volume content along their length. Optical microscopy was used to account for this phenomenon and modified deformation results were compared with the prepreg tape results in [18]. This comparison revealed that the relationship between resin viscosity and shear flow may be overshadowed by the presence of inextensible fibres oriented in the direction of flow. This was especially evident in the behaviour of the prepreg 8HS tested, as percolation was found to be dominant and shear flow non-existent regardless of testing temperature. Finally, this fibre-locking observation would also suggest that the rib filling observed Figure 1 is due to interply slippage as opposed to intraply deformation.

5 ACKNOWLEDGEMENTS

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