CHARACTERISATION OF THE PROCESSING PROPERTIES OF OUT-OF-AUTOCLAVE PREPREGS

Chris Hickey\textsuperscript{1}, cmdhickey@gmail.com
Simon Bickerton\textsuperscript{2}, s.bickerton@auckland.ac.nz

Abstract. Out-of-autoclave prepregs are the construction material of choice for high performance marine craft, due to their high material properties, and relatively low processing costs compared to traditional autoclave prepregs. Because of the low compaction pressure, voids are not collapsed by external pressure; therefore air must be removed from the laminate prior to cure to ensure a low void-content, high quality laminate is produced. This work presents the development of experimental techniques to accurately measure the as-laminated void content, compaction response and in-plane and through thickness air permeability of two prepreg materials. The material models developed were implemented into an air removal model, and compared against experiments. Excellent correlation was observed for the cloth material, whereas the unidirectional material exhibited complete closing of air pathways, which was not captured well by the current model.

1. INTRODUCTION

Composites manufactured from pre-impregnated reinforcement, commonly known as prepreg, cured inside an autoclave is the current pinnacle of part quality and mechanical performance in composites manufacturing, and hence is the mainstay of the aerospace industry. Plies of pre-impregnated reinforcement are laid into the mould then covered with perforated release film, breather and vacuum bag, the bag evacuated, and the part cured under elevated temperature and pressure in an autoclave (pressurised oven).

An autoclave is a pressure vessel, therefore increasing part size and hence increased autoclave diameter necessitates an increase in autoclave wall thickness resulting in a large increase in cost. In order to restrict manufacturing cost, numerous class rules specify the maximum pressure to be used in manufacture, particularly for hull and decks, thereby restricting manufacture to ‘out-of-autoclave’ or ‘oven-cure’ processes.

As an example the AC62 class rule specifies “Hulls and their internal structure shall not have pressure applied at any time during construction that exceeds 1.0 atmosphere, but this limitation shall not prohibit building methods including the use of clamps or mechanical fastenings, wrapping, and winding, etc.”\textsuperscript{[1]}

Curing outside of the autoclave therefore offers advantages of reduced production cost, and less restrictive part size constraints. However because of the use of vacuum alone, without the application of autoclave pressure (typically 6 bar), any entrapped air in the laminate results in voidage in the part, reducing mechanical properties, particularly compressive and shear strength\textsuperscript{[2]}.

Key to producing a high quality laminate without autoclave pressure is the removal of entrapped air before the cure cycle commences. Prepreg manufacturers have developed materials with air pathways, to improve the evacuation of air from the laminate stack, and therefore reduce voids in the final laminate. These techniques generally employ a prepreg material that is semi-impregnated, thereby creating pathways for air to escape, unlike traditional autoclave prepreg, where the reinforcement is fully impregnated with resin. Various manufacturers have developed a range of techniques to physically create these air evacuation pathways, including semi-impregnating the tows with resin, applying the resin in a film, or applying the resin in discontinuous stripes. These techniques create materials with varying air flow resistance both in-plane and through thickness.

To date there is no clear method to correlate these prepreg morphological parameters, to their processing requirements in terms of strategies and time required to sufficiently remove air from a laminate. This work presents the development of a method to characterise the processing properties of two out-of-autoclave prepregs, in order to develop processing guidelines.

2. MATERIAL CHARACTERISATION

2.1 Porosity by Gas Pycnometry

In order to develop accurate predictions of the air removal time of out-of-autoclave prepreg materials, an accurate knowledge of the quantity of air entrapped in the laminate is required. Traditional methods for measuring porosity of cured laminates by gravimetric methods are incapable for measuring the porosity of uncured prepreg, due the network of porosity that may or may not be infiltrated by the test fluid.

\textsuperscript{1} Centre for Advanced Composite Materials, Department of Mechanical Engineering, University of Auckland, now Southern Spars
\textsuperscript{2} Professors, Centre for Advanced Composite Materials, Department of Mechanical Engineering, University of Auckland
A gas pycnometer is a device for measuring volume of a sample, allowing the skeletal density of a substance to be calculated, based on a measured pressure decrease and ideal gas law. A gas pycnometer is typically used to measure the density of powders, foams and other porous substances. A gas pycnometer suitable for the measurement of prepreg samples was designed and manufactured with a schematic shown in Figure 1 and photographs of the final device in Figure 2 and Figure 3.

The procedure was as follows:

- Pressurise reference chamber.
- Record steady state pressure.
- Open Valve V2.
- Record steady state pressure.

The skeletal volume could then be calculated using Equation 1:

$$V_{\text{skeletal}} = \frac{P_{\text{high}} - P_{\text{equilibrium}}}{P_{\text{atm}} - P_{\text{equilibrium}} V_{\text{samplechamber}}}$$  \hspace{1cm} (1)

The two materials analysed in this work were a 300gsm cloth material, and a 150gsm unidirectional material as described in Table 1.

<table>
<thead>
<tr>
<th>Material Specifications</th>
<th>Cloth</th>
<th>Unidirectional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fibre Areal Weight (g/m²)</td>
<td>300</td>
<td>150</td>
</tr>
<tr>
<td>Fibre Architecture</td>
<td>Plain Weave</td>
<td>Unidirectional</td>
</tr>
<tr>
<td>Resin System</td>
<td>SE70</td>
<td>SE70</td>
</tr>
<tr>
<td>Resin Weight (%)</td>
<td>42</td>
<td>37</td>
</tr>
<tr>
<td>Fibre Density (kg/m³)</td>
<td>1808</td>
<td>1800</td>
</tr>
<tr>
<td>Uncured Resin Density (kg/m³)</td>
<td>1160</td>
<td>1160</td>
</tr>
</tbody>
</table>

It was noticed that with increasing pressure, the total measured skeletal volume decreased, indicating some compressibility of one of the phases. Of the fibre, resin and entrapped air phases, it is assumed this compressibility can be attributed to the entrapped air alone, which allows the entrapped porosity to be calculated along with the open-cell porosity. Figure 4 presents the closed cell (not open to edge of sample) and open cell (open to edge of sample) porosity of each material at a range of nominal fibre volume fractions. Different practically achievable nominal fibre volume fractions were achieved by altering the number of layers in the 10mm deep pycnometer cavity. This correlated to fibre volume fraction ranges of 33-42% and 48-53% for the cloth and unidirectional materials respectively.

![Figure 1: Schematic of pycnometer](image1.png)

![Figure 2: Photo of pycnometer, pressure control and data acquisition](image2.png)

![Figure 3: Photograph of pycnometer internal geometry with carbon prepreg sample in left chamber](image3.png)

![Figure 4: Measured sample porosity](image4.png)

2.2 Compaction Response

In order to establish the relationship between applied pressure and laminate thickness and hence porosity, the compaction response of both materials at varying numbers of layers was quantified. Techniques traditionally employed in the characterisation of dry
reinforcements were used, where a sample is compressed between two parallel platens in an Instron Universal Testing Machine, as shown in Figure 5. Load and cavity thickness are measured continuously, allowing applied pressure and void fraction respectively to be calculated.

![Figure 5: Schematic of fixture used for compaction and in-plane permeability measurements.](image)

![Figure 6: Photograph of fixture used for compaction and in-plane permeability measurements.](image)

Figure 7 and Figure 8 present the measured compaction response of both material types at varying number of layers.

![Figure 7: Compaction response of cloth material](image)

![Figure 8: Compaction response of unidirectional material](image)

Generally both materials showed significant non-recoverable plastic deformation. For both materials, significant nesting behaviour was observed, in that for increasing numbers of layers, a lower level of porosity was reached. Due to the more optimum packing of the unidirectional material, lower levels of porosity were reached, compared to the cloth.

2.3 Through Thickness and In-Plane Air Permeability

Permeability tests were performed over the range of porosities found from the compaction tests in Section 2.2. In plane permeability was performed using the same test fixture as for compaction shown in Figure 6. Air passes through a flow meter, via a pressure sensor, and flows radially outwards in the circular sample. In-plane Darcy permeability was calculated using Equation 2.

\[
K = \frac{Q_{\text{high}} \cdot \mu \ln \left( \frac{r_o}{r_i} \right)}{\pi h \left( \frac{P_{\text{high}}}{P_{\text{low}}} \right)^{\frac{2}{3}}}
\]  

(2)

Figure 9 and Figure 10 present the measured in-plane permeability for the cloth and unidirectional material respectively, along with model fit. For the cloth material, a simple exponential model was used, and for the unidirectional material a model based on the work by Gebart [ref] was used, in order to capture the rapidly decreasing permeability approaching zero porosity.
Through-thickness permeability was measured using the fixture shown in Figure 10 and Figure 11. The key features of this fixture are the air-permeable machined porous aluminium platens, and the exterior channel around the sample in which silicone grease was injected to prevent air from passing around the prepreg sample.

Of note is that the through-thickness permeability of the unidirectional material was immeasurable, equivalent with that of a vacuum bag sample used in a leak check of the system. This equates to a permeability of less than $2\times10^{-22}$ m$^2$. This has significant implications in the processing of this unidirectional laminate.

Figure 13 presents the result of the through thickness permeability tests across a range of thicknesses and hence porosities.

### 3. MODEL APPLICATION

The previously developed material models were implemented into a one-dimensional Darcy flow air removal model, including compressibility effects, in order to predict the air removal time for laminates of varying dimensions.

For the cloth material this was a through thickness model, as for most practical geometries, flow would be predominately through thickness, based on the measured permeabilities. Figure 14 presents a schematic of the test.
Figure 14: Schematic of through thickness permeability model experimental validation test.

Figure 15 presents model comparisons for 16 layers of cloth material, for first vacuum application, and in Figure 16 when vacuum is removed, as would be typical in a debulk process.

Of note is the short time (approximately 30s) required to initially remove the air from the laminate, and the long time for air to re-enter the laminate after vacuum is removed. (Approximately 1 hour), showing the significance of the plastic deformation that occurs during compaction and the strong affect this has on permeability behaviour. Generally the fit of the model is very good.

Figure 16: Pressure versus time after vacuum is removed debulk of 16 layers of cloth material

For the unidirectional material, flow is solely in-plane so a 1-dimensional in-plane model was used. Figure 17 presents a schematic of the experiment, with 3 sides sealed and one open to vacuum.

Figure 17: Schematic of the unidirectional in-plane permeability test.

Figure 18 presents an example model comparison for 2 layers of unidirectional material. Here a relatively poor model fit can be seen, indicating there is some closing off of air paths that does not appear to be well captured by the model. An improvement of the model could be to include spatially varying porosity and hence permeability, induced by the spatially varying air pressure and hence compaction stress on the prepreg. This could potentially capture the low/no permeability behaviour at the edge of the laminate.

Figure 15: Pressure versus time for first debulk of 16 layers of cloth material
4. CONCLUSIONS

Out-of-autoclave prepreg materials find favour in the marine industry, due to their relatively high performance, yet low processing costs when compared to autoclave cure materials.

Because of the lower compaction pressure applied, air removal is the key method of void mitigation, rather than void collapse as occurs with autoclave processing.

In order to measure the widely varying processing properties of prepreg materials, a methodology has been developed to accurately measure the open and closed cell porosity, compaction response and through thickness air permeability of prepreg materials.

The material models found were implemented into a one-dimensional air removal model, and compared against experiments. Very good agreement was found for the cloth material, whereas unidirectional material exhibited close off behaviour, and requires further investigation and model development.

This combined material characterisation and modelling technique enables the quantity of air entrapped, the ease at which it can be removed to be quantified, enabling the prediction of required consumable layouts and vacuum times, in order to produce void free laminates, without the use of an autoclave.

References