ABSTRACT

Out-of-autoclave (OOA) pre-impregnated (prepreg) materials are a prospective alternative to traditional autoclave processing, with the potential to reduce processing costs and build structures without size limitations imposed by the autoclave. Gas transport pathways in prepreg laminates play an important role in the removal of entrapped gases and volatiles during processing. Removal of gases by vacuum evacuation is essential in order to produce composite laminates with low final void content. Gas pathways are of particular importance in OOA prepregs where the maximum applied pressure during processing is 1 atm.

In this study, the gas transport of OOA prepreg MTM45-1/CF2426A (epoxy/carbon) laminates is examined. The gas permeability of laminates is carefully measured in the in-plane and through-thickness directions. The study examines the effect of the number of layers, the effect of internal ply terminations, and the effects of heat on laminate gas transport. Supplemental experiments such as laminate compaction, microscopy, and water visualization are conducted to gain additional understanding of laminate gas transport.

The study shows that gas transport is strongly directional for the studied prepreg with significantly higher permeability in-plane than in the through-thickness direction. Counter-intuitively, the permeability of MTM45-1/CF2426A is not found to be greater than autoclave prepreg when compared to carbon/epoxy Toray 3900-2 (plain weave). The physical nature of gas transport pathways in MTM45-1/CF2426A prepreg laminates is found to change with processing state. Debulking was found to decrease in-plane gas transport from its as-laid-up permeability. Laminate heating is found to affect laminate gas transport. In-plane permeability decreased with increasing temperature, while through-thickness permeability increased with increasing
temperature. Correlations between gas transport and laminate compaction is also evident. During debulking, laminate compaction is found to correlate to decreasing in-plane permeability. Additionally, laminate compaction is found to relate to the quality of edge breathing.
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1. INTRODUCTION

The use of carbon/epoxy composites in aerospace applications, primarily in the military sector, began in the 1970’s (Nutt & Boyd). Over the past few decades, the use of composites has become widespread in both military and increasingly in commercial applications. Currently, in various commercial aircraft, composite materials are commonly used for components like rotor blades, wing spars and vertical stabilizers. With improvements in composite design and manufacturing, composite materials are now being used for primary structures in the next generation of commercial aircraft. The Boeing 787 is the most notable example, where composites make up a majority of the aircraft, including key primary structures. Examples of these structures include: the fuselage, wings, wingbox and stabilizers.

Although infinite combinations of reinforcement fibres and resin systems exist for structural applications in the aerospace industry, the combination of interest in this work is carbon/epoxy. As this study involves aerospace composite material, for the remainder of this thesis, the words composite and prepreg refer to carbon/epoxy.

1.1 Composites Processing

A common method of building composite structures is to use pre-impregnated (prepreg) materials and build them up as laminate structures. Prepregs are a combination of reinforcement fibres impregnated with a resin (matrix). By pre-impregnating fibres with resin, the prepreg manufacturer insures an optimum fibre to resin ratio. Controlling the fibre to resin ratio is an advantage over manual composite wet lay-up which is highly variable. Typically, prepregs are supplied in the form of rolls to part manufacturers, which are then cut to the desired shape (Figure 1-1).
Cutting prepreg into sheets, the individual layers (often referred to as plies) are laid-up onto one another forming a laminate structure. Once laid into the desired part geometry, the composite is hardened by curing the matrix in a heating process. The typical manufacturing process flow is shown in Figure 1-2.

Figure 1-1. CFRP Prepreg Material Supplied in a Roll Form.

Figure 1-2. Typical Process Flow for a Composite Prepreg Laminate Structure.
Vacuum bag processing is done to both compact laminate plies together and to evacuate entrapped gases from within the laminate. Vacuum evacuation involves placing the laid-up laminate in a vacuum environment where air is then evacuated. The vacuum environment is achieved using a disposable vacuum bag set-up, consisting of a nylon bag and other consumables (Figure 1-3). An intermediate vacuum evacuation step, referred to as debulking, is often used prior to the cure process. In debulking, vacuum is periodically applied after laying down prepreg layers to compact the laminate plies together. Curved geometries require debulking in order to conform the laminate to the tool shape, and avoid wrinkling and bridging of the material at sharp geometry changes.

Figure 1-3. Schematic of a Typical Vacuum Bag Set-Up. Recommended Set-Up by Advanced Composites Group for MTM45-1 Processing.

The vacuum bag set-up is designed to have the laminate remain inside during the cure process where vacuum is usually pulled on the system. Whether vacuum is continually drawn throughout the cure process is dependent on the manufacture’s recommended cure cycle (MRCC) unique to each prepreg’s resin system. As the resin matrix increases in temperature during the heated cure cycle, its viscosity lowers allowing resin to flow and wet-out the reinforcement fibres. Resin flow stops when gelation occurs (i.e. where a 3-D polymer network is formed with the onset of
cross-linking between polymer chains). Once fully cured, the vacuum bagging consumables are removed and the cured laminate is pulled off the tool for finishing (cutting, sanding, painting, etc.).

In addition to applying heat, traditional processing of aerospace composite structures uses an autoclave for consolidation during the curing process (Thomas, Bongiovanni, & Nutt, 2008). The autoclave provides applied positive pressure usually around 7 atm. The positive pressure imposed by autoclave is greater than the vapour pressures of the entrapped gases, suppressing them into solution. Curing in this state creates a final cured laminate free of voids.

Figure 1-4. Autoclave used for Autoclave Processing of Composites.
Despite autoclave processing producing high quality composites with low void content, there are several drawbacks: 1) Autoclaves represent a large capital cost investment. 2) The physical size of an autoclave limits the size of a composite structure that may be produced. 3) Autoclave processing typically involves a high cure temperature, requiring expensive tooling material.

1.2 Out-of-Autoclave Prepreg MTM45-1

Out-of-autoclave (OOA) prepregs represent an emerging class of prepreg materials designed specifically to be cured in an oven under a vacuum bag without positive pressure applied. In the aerospace industry, OOA prepregs are seen as attractive material systems for producing large primary structures. The reduction of expensive process equipment is seen as an opportunity to open up the market to smaller composite manufactures and to out-source to a larger subcontractor base. However, vacuum-bag-only low temperature cure prepregs traditionally exhibit high final void content (Repecka & Boyd, 2002). The absence of the consolidation pressure imposed by the autoclave puts a greater importance on gas extraction by vacuum only mechanisms. Gases not removed prior or during the cure process result in voids in the cured laminate. In autoclave prepregs, consolidation pressures of 7 atm during the cure process acts to reduce the volume or solution-ize entrapped gases that are not removed through vacuum evacuation. With OOA processing, the maximum effective applied pressure (consolidation pressure) is restricted to 1 atm. To counter this, the key design philosophy is to allow the prepreg laminate to ‘breathe’ and vent off entrapped gases and volatiles. Section 2.1 contains a more in-depth background of OOA prepregs and their processing.

The material system in this study is OOA prepreg MTM45-1/CF2426A (carbon/epoxy) produced by Advanced Composites Group (ACG). It consists of AS4 6k carbon fibre tows woven in a
five-harness satin weave partially impregnated with the MTM45-1 toughened epoxy matrix resin system. The MTM45-1 matrix resin system is a high performance toughened epoxy system optimized for low temperature vacuum-bag-only processing. A notable aerospace application using MTM45-1 is the Scale Composites built Virgin Galactic suborbital spacecraft and launch vehicle Spaceship 2 and White Knight 2 (Ridgard, 2010b). Designed to be highly versatile, the MTM45-1 resin system has cure temperatures ranging from as low as 80°C to up to 180°C. The processing parameters for ACG MTM45-1 system, including its variable cure cycle, is listed in Table 1-1.

Table 1-1. Recommended Initial Cure Cycle for MTM45-1 Resin System Prepregs. Source: Advanced Composites Group MTM45-1 PDS1205/11.07/3

<table>
<thead>
<tr>
<th>Ramp Rate</th>
<th>Vacuum Bag Processing</th>
<th>Autoclave Processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure</td>
<td>N/A</td>
<td>89-207inHg (3-7bar)</td>
</tr>
<tr>
<td>Vacuum Pressure</td>
<td>&gt;28inHg (0.95bar)</td>
<td>&gt;28inHg (0.95bar)</td>
</tr>
<tr>
<td>Cure Time</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20 hours @ 80°C (176°F) Or</td>
<td>20 hours @ 80°C (176°F) Or</td>
<td></td>
</tr>
<tr>
<td>4 hours @ 120°C (248°F) Or</td>
<td>4 hours @ 120°C (248°F) Or</td>
<td></td>
</tr>
<tr>
<td>2 hours @ 130°C (266°F)</td>
<td>2 hours @ 130°C (266°F)</td>
<td></td>
</tr>
<tr>
<td>2 hrs @ 180°C (356°F)</td>
<td>2 hrs @ 180°C (356°F)</td>
<td></td>
</tr>
</tbody>
</table>

The MTM45-1 resin system is designed for a low temperature initial cure. Maximum epoxy properties, such as mechanical strength and $T_g$ are achieved through a tool-free post cure (180°C). The low temperature initial cure permits the use of low temperature tooling materials. According to ACG, the viscosity of MTM45-1 is designed to be higher than that of traditional autoclave cure prepregs (Ridgard, 2010a). This design philosophy allows maximum time for
entrapped gas and volatile evacuation via gas permeation pathways prior to resin flow filling in these paths during the cure process (Mason, 2006).

1.3 Scope and Objectives of Thesis Work

The scope of this study was to measure gas transport in MTM45-1/CF2426A prepreg laminates and to observe how it is affected by various processing parameters. This study examined the effect of the number of laminate plies, debulking, and temperature on the laminate gas transport. To highlight how the findings from this study could be used in industrial applications, gas transport data was used as a model parameter in an existing gas transport model. From this, a guide for the evacuation times required for processing of MTM45-1/CF2426A prepreg laminates was generated.

Justification for this work is described by Repecka and Boyd (of Cytec Engineered Materials, Inc.) from their work in the 1980’s on Thick Laminate Prepreg (TLP) technology development. They concluded that the single major factor leading to voids in epoxy prepregs was a lack of gas permeability in the prepreg (Repecka & Boyd, 2002).

There were four individual objectives of this study.

Objectives:

- Characterize gas transport in different laminate directions: in-plane and through-thickness
- Investigate the effects of lay-up parameters: number of plies, debulking, and temperature
- Determine if there is a relationship between laminate compaction (during debulking) and gas transport
• Apply the measured data to an existing gas transport model to generate a guide for required evacuation times
2. BACKGROUND AND LITERATURE REVIEW

2.1 Out-of-Autoclave Composite Prepregs

To produce high-performance composite structures, there are typically three typical processing alternatives to traditional autoclave processing. 1) Vacuum-assisted resin transfer moulding (VARTM), 2) resin transfer moulding (RTM), and 3) out-of-autoclave prepregs (OOA prepregs). VARTM and RTM processes are resin infusion liquid moulding processes while OOA prepreg processing is simpler with regards to handling and manufacturing. Controlling the fibre to resin ratio, ease of handling and manufacturability, and typically higher performance resin systems are all advantages associated with prepregs. Because of these benefits, OOA prepregs are generally preferred and are receiving great interest for aerospace structural applications over the liquid moulding techniques for producing large structures. Described by Thomas and Nutt to be “a scant body of literature”, the development of OOA prepregs is not well documented (Thomas & Nutt, 2009). However, a brief overview of their differences from traditional autoclave processed prepregs, and those currently available (i.e. newer generation OOA prepregs suitable for structural applications) on the market is given here.

Unlike autoclave prepregs, OOA prepregs are designed to be vacuum-bag-only processed and cured in an oven. Designed specifically for oven cure, the resins impregnated into OOA prepregs are different than autoclave cure resins. One major difference between OOA prepregs and autoclave prepregs is that the latter relies on a combination of vacuum evacuation and high applied pressure to suppress entrapped gases and volatiles. However, the absence of an autoclave limits the compaction pressure on a laminate to 1 atm, which is a particular challenge for OOA compared to autoclaving (Tavares, Michaud, & Månson, 2010). In OOA processing, entrapped
gases and volatiles entrapped within the laminate cannot be dissolved into solution under the low compaction pressure. Consequently, producing void free laminates is achieved by removing entrapped gas and volatiles by vacuum mechanisms through engineered gas extraction pathways in the prepreg. The development of the newer OOA prepreg systems has been aided by a better understanding of mechanisms of air extraction and the material system’s ability to ‘breathe’ during processing (Ridgard, 2010a). The measurement of this ability, permeability, is the focus of this thesis.

Sometimes referred to as vacuum-bag-only systems, traditional first generation out-of-autoclave prepreg systems were designed for non-structural applications. Instead of design being driven by mechanical performance, the emphasis was placed on reducing processing costs. To reduce processing costs, these early OOA prepregs were designed for low temperature cure to allow the use of inexpensive tooling materials. After the initial cure, matrix properties were enhanced by a tool-free post-cure (Ridgard, 2010a). Typically, these systems produced laminates with high porosity and inferior mechanical properties compared to autoclave processed prepregs (Bond, Griffith, & Hahn, 2008). As a result, early OOA prepregs were primarily used for prototyping and limited production parts where applications were less structurally demanding (Ridgard, 2009). The early development of OOA prepregs for structural aerospace applications was carried out by McDonnell Douglas, Lockheed/Martin and the US Air Force in the 1990’s (Ridgard, 1997). Today’s newer generation OOA prepreg systems are designed for mechanical performance to be on par with autoclave processed systems. Other design considerations are: glass transition temperature $T_g$ (therefore affecting service temperature), out-life, and tack-life. Using autoclave prepregs as the benchmark, the newer OOA prepreg systems are being designed
to match if not exceed all of the aforementioned areas without the need of the applied pressure provided by the autoclave.

The development of the newer generation, sometimes referred to as second generation out-of-autoclave prepreg systems, began in the early 2000’s (Ridgard, 2010a). These new OOA systems are claimed to be able to achieve void free autoclave part quality (ACG, 2007; Cytec, 2009). Table 2-1 summarizes the currently available OOA prepregs suitable for structural applications. Prepreg manufacturer Toray is missing from the list, as they do not officially have an OOA prepreg system currently available on the market. Other OOA prepreg systems, resin film infusion (RFI) systems, and resin film adhesive layers exist but were left off. Only structural suitable prepregs are listed.


<table>
<thead>
<tr>
<th>Prepreg System</th>
<th>Target Market</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTM44-1</td>
<td>Aerospace</td>
<td>(ACG, 2010a)</td>
</tr>
<tr>
<td>MTM45-1</td>
<td>Aerospace</td>
<td>(ACG, 2007)</td>
</tr>
<tr>
<td>MTM46</td>
<td>Aerospace</td>
<td>(ACG, 2010b)</td>
</tr>
<tr>
<td>LTM24ST</td>
<td>Motorsport, semi-structural (e.g. race car bodywork)</td>
<td>(ACG, 2006)</td>
</tr>
<tr>
<td>CYCOM 5320</td>
<td>Structural</td>
<td>(Cytec, 2009)</td>
</tr>
<tr>
<td>CYCOM 5215</td>
<td>Automotive, Marine, Tooling</td>
<td>(Cytec, 2002)</td>
</tr>
<tr>
<td>HexPly M56</td>
<td>Aerospace</td>
<td>(Hexcel, 2010)</td>
</tr>
<tr>
<td>HexPly M9</td>
<td>Marine, wind energy</td>
<td>(Hexcel, 2007)</td>
</tr>
</tbody>
</table>
In order to produce void free laminates, entrapped air and other volatiles must be removed prior to, or early in, the cure cycle. The key design parameter for OOA prepregs is their ability to remove entrapped gases and volatiles by vacuum only mechanisms. Among each prepreg manufacturer, the design philosophy of how to ‘breathe’ the laminate is different.

Another secondary design consideration for OOA prepregs mentioned in the literature is controlling the amount of off-gassing due to the resin curing reaction (Thomas & Nutt, 2009). One approach is to focus on resin chemistry. For thermoset resin systems, two polymerization reactions exist: addition polymerization, and condensation polymerization. The use an addition-cured resin, which does not give off volatiles during the curing reaction, rather than a condensation cure (where volatiles result from polymerization and create a potential void source) may be beneficial for OOA prepregs (Thomas & Nutt, 2009). However, because the focus of this thesis is on measuring a laminate’s ability to breathe, resin particulars involving curing reactions will not be discussed any further in this thesis.

2.2 Gas Extraction and Void Relationship

The presence of voids and porosity are a major concern in the processing of composite materials where voids in the final cured laminate are detrimental to mechanical performance. Voids are defined as empty cavities in the laminate whereas porosity is defined as the volume of voids with respect to the overall volume of the laminate. The formation and growth of voids is thought to be primarily due to entrapped volatiles (Mallow & Campbell, 2000; Xu, Repecka, Mortimer, Peake, & Boyd, 2002). The final void content of a laminate is described to be directly related to its void sources and void sinks (Arafath, Fernlund, & Poursartip, 2009). Inherently from the prepreg lay-up process, the presence of voids is inevitable (Boey & Lye, 1992). The ability to remove the
entrapped gases and other volatiles is crucial in producing cured laminates with a low fraction of voids. This in particular has been problematic in thick composites parts, as the removal of entrapped gases under vacuum is highly time dependent (Nam, Seferis, Kim, & Lee, 1995). The removal of entrapped gases and volatiles is achieved through a variety of mechanisms, some of which are not applicable in out-of-autoclave processing. In autoclave processing, gases are removed through a combination of applied pressure and vacuum evacuation mechanisms, while out-of-autoclave processing relies solely on vacuum evacuation. Thorfinnson and Biermann (1986; 1987) investigated what processing factors affect laminate void content and found that controlling the degree of impregnation of a prepreg was fundamentally important in allowing laminates vent out entrapped gases. Partial resin impregnation of the prepregs creates gas transport pathways within the laid-up laminates (Thorfinnson & Biermann, 1986, 1987).

### 2.2.1 Gas Extraction Pathways

The removal of entrapped gases from prepreg laminates through vacuum only mechanisms is achieved physically by transporting gases through purposely engineered gas flow pathways. The gas evacuation process is driven by the pressure gradient that exists from within the laminate (recalling that initially gas entrapped is at 1 atm), and the pressure outside of the laminate within the bag set-up as illustrated in Figure 1-3 (close to 0 atm under hard vacuum conditions). The physical nature of these pathways is specific to each prepreg system. They may consist of purposely dry areas or channels in the impregnated resin, dry fibre tows, or a ‘vascular network’ of interconnected voids (i.e. a porous resin) which permit gas transport. By carefully controlling partial resin impregnation or using perforated resin film, gas extraction paths may be created in the prepreg. (Thomas et al., 2008; Thomas & Nutt, 2009). By design, these pathways are present after lay-up but are then filled-in and closed as the resin flows due to a drop in viscosity during
Chapter 2 Background and Literature Review

the heated curing process. Resin rheology is a significant consideration and can be tailored to maximize the time for air and volatile removal. For example, prepreg manufacturer AGC believes in having a high resin viscosity to maximize gas extraction (Ridgard, 2009).

Gas transport within prepreg laminates is reported in the literature to be highly anisotropic in nature occurring in two distinct flow directions; in the laminate plane (in-plane) and through the laminate thickness (through-thickness). With regards to in-plane gas transport, gas travel can occur both inter-laminar (between individual plies) and intra-laminar (within each individual ply) as is illustrated in Figure 2-1.

![Figure 2-1. Gas transport in the in-plane laminate direction.](image)

Comparing these in-plane flow paths, a number of studies have found that gas transport occurs predominately inter-laminar (Ahn, Seferis, Price, & Berg, 1991; Nam et al., 1995; Putnam & Seferis, 1995; Shim & Seferis, 1997). However, this is different for each prepreg system as in-
plane gas flow is affected by how the reinforcement fibres are impregnated with resin and the reinforcement fibre architecture. Additionally, orthogonal difference in gas flow in-plane may also result dependant on fibre orientation. The alternate direction of gas travel is the through-thickness laminate direction shown in Figure 2-2.

![Diagram of gas transport in the through-thickness laminate direction.](image)

**Figure 2-2. Gas transport in the through-thickness laminate direction.**

For gas transport in the laminate through-thickness direction, gas must physically travel through resin layers. This may be aided with porous resin films, or resin film dry spots which are present in some prepreg systems.

Particularly for OOA prepreg systems, prepreg manufacturers put a large emphasis on edge breathing during processing. This suggests that by design, in-plane gas transport is more dominant than the through-thickness transport.
2.3 Permeability Studies

Permeability studies on composite materials have been carried out on and off over the past 20 years. The importance of this work and its relationship to void reduction in cured composite laminates was first emphasized in the work of Thorfinnson and Biermann (Thorfinnson & Biermann, 1986, 1987). They described permeability as the property of prepreg that is of the most interest to describe the laminate’s ability to remove entrapped gases. However, the first permeability studies of composite prepregs in the literature did not appear until a decade later. These studies investigated the permeability of composite prepregs and its relation to producing void free autoclave processed laminates. After these initial studies, prepreg gas permeability appears to have largely been ignored until recently when OOA prepregs and their processing have once again begun to raise interest in the topic. Additionally, today, permeability studies have been expanded to include (constituting a bulk of the current literature) liquid permeability (resin) in resin film infusion (RFI) and liquid composite moulding (LCM) processes.

Initial gas permeability studies of prepreg composites were conducted by a research group led by Seferis in the 1990’s in the Polymeric Composites Laboratory in the Department of Chemical Engineering at the University of Washington. The group published a series of studies on autoclave prepreg systems investigating gas permeation. They examined the affect of applied compaction pressure, and a limited investigation into the effects of the heated cure cycle on laminate gas transport (Ahn et al., 1991; Nam et al., 1995; Putnam & Seferis, 1995; Shim & Seferis, 1997). Their early studies measured gas permeation by imposing a pressure differential on opposing sides of a laminate and then monitoring the rate of pressure change between the two sides. Seferis et al. (1991; 1995) derived a model to describe gas permeation based on a Darcy’s law flow model using a pressure-decay approach, which related the rate of pressure change as a
function of the laminate’s ability to transfer gas (permeability). Rather than quantify it as permeability (independent of geometry), permeation was described as function of laminate thickness (i.e. number of layers) (Ahn et al., 1991; Nam et al., 1995; Putnam & Seferis, 1995). In later studies, Seferis et al. (1995; 1997) modified their approach by using a steady-state Darcy’s law equation, modeling gas permeation as permeability. In the steady-state approach, permeability was determined by measuring the volumetric rate of gas flow through a laminate (Nam et al., 1995; Shim & Seferis, 1997). Seferis et al. found that in-plane, in particular interlaminar, permeability was largely dependent on fibre architecture. For example, due to how the prepreg layers nest with respect to each another; uni-directional, plain weave, and five-harness satin prepregs each exhibited differing gas permeability.

After the work by Seferis’ group in the 1990’s, no gas permeability studies were published in the literature again until the late 2000’s. Recent gas permeability studies performed on prepregs materials were carried out at Ecole Polytechnique Fédérale de Lausanne (EPFL) (Tavares, Caillet-Bois, Michaud, & Månson, 2007; Tavares, Michaud, & Månson, 2009; Tavares et al., 2010). For use in curing of sandwich structures, these more recent investigations studied the through-thickness permeability of an OOA prepreg (ACG VTM 264). To determine gas permeability, Darcy’s law was also used to model gas flow. Like the early studies by Seferis et al., permeability was calculated using a falling pressure (pressure-decay) approach.

More recently, permeability tests similar to those performed at EPFL were carried out by Kratz at McGill University. Kratz performed his tests on OOA prepreg MTM45-1/CF2426A, the same materials system as this thesis study (Kratz, 2009). The study briefly looked at the role of gas permeability in relieving internal sandwich structure core pressure during the heated cure process. Kratz’s study was performed under the same joint collaborative project umbrella as this
thesis study. It should be noted that although Kratz's study refers to permeability characterization, the work represents an ‘effective’ permeability in sandwich structure processing, whereas this study measures permeability as an intrinsic material property for given processing states.

Liquid permeability for resin infusion processes (rather than gas permeability) represents a majority of the more recent composite permeability studies. A number of studies look at the impregnation of fibres with flowing resin during heating in the RFI process (Shim, Ahn, Seferis, Berg, & Hudson, 1994; Thomas et al., 2008; Thomas & Nutt, 2009). In the RFI process, a resin film is placed on top of dry fibres, and infiltrates the fibres when heated. This process is conceptually similar to the fibre wet-out that occurs during heating of a partially impregnated prepreg. Other liquid permeability studies look at the permeability of fibre performs in LCM techniques such as RTM and VARTM (Choi, Lee, Chang, & Lee, 1998; Gebart & Lidström, 1996; Hoes et al., 2002; Michaud & Mortensen, 2007; Schell, Deleglise, Binetruy, Krawczak, & Ermanni, 2007; Verrey, Michaud, & Månson, 2006; Zingraff, Michaud, Bourban, & Månson, 2005).

For liquid flow in fibrous performs, there are two flow conditions: saturated flow and unsaturated flow. In saturated flow, the fibre tows are already wetted. Resin flows to replace existing resin already in the fibres. In unsaturated flow, resin flows while simultaneously wetting the dry fibres. This wetting adds additional capillary effects. For saturated flow, the flow model for liquid resin assumes Darcy’s law. For unsaturated flow, additional terms are added to account for the capillary effect. With the capillary effect, the resin flow front velocity is slowed because of a localized pressure drop at the vicinity of the flow front due to surface tension (Schell et al., 2007; Verrey et al., 2006; Zingraff et al., 2005). At low fluid flow velocities, the capillary
pressure effect becomes more important (Hoes et al., 2002). Upon accounting for these different resin flow conditions in Darcy’s law, liquid permeability is determined. However, despite permeability being an intrinsic property, there are some differences between measured liquid permeability and measured gas permeability.

Differences between liquid permeability and gas permeability exist in cases where permeability is low and fluid flow is slow. This is because of the Klinkenberg effect, otherwise known as slip flow. In this scenario, gas molecules at the capillary walls move, which is in disagreement to the laminar flow condition. Instead of the fluid velocity being zero at the walls due to friction, gas molecules at the walls slip and move. Due to the over exaggerated gas velocity resulting from the slip flow phenomenon; gas permeability is greater than liquid permeability. Converting between gas and liquid permeability is done with the experimentally determined Klinkenberg correction factor (Gas Transport in Porous Media, 2006). The correction factor is dependant on both the specific gas being transported and temperature. In rock hydrology, for rocks with similar permeability as composite prepgs, the differences between gas (such as nitrogen) and liquid (such as water) permeability is described to be at most one order of magnitude (Tanikawa & Shimamoto, 2006).

2.4 Gas Permeation Flow Models

Gas transport through porous media consists of both diffusive and momentum mass transport. However, when considering the time-scales involved for gas diffusion, its effect is negligible. Tavares et al. reported that the diffusion coefficient of nitrogen in epoxy resins is between $10^{-11}$ m$^2$s$^{-1}$ and $10^{-9}$ m$^2$s$^{-1}$. This equated to 0.8mm of diffusion transport distance in 5 hours (Tavares et
al., 2009). Based on this consideration, modeling gas transport assuming only momentum transport is sufficient.

Momentum transport of a fluid through a porous medium can be characterized as permeability, an intrinsic material property describing a porous medium’s ability to transport fluids. Permeability was used in this study as a convenient way to normalize gas permeation data from the test geometry. This allowed the ability of a prepreg laminate to vent entrapped gases and volatiles to be quantified. However, it should be noted that this ability is directly dependant on the microstructure of the laminate’s internal gas flow pathway network, which is a function of temperature, pressure and processing history. Therefore, the measured permeability for a laminate is not a true intrinsic material property, but instead a value for the given process state of the composite. In this study, the processed state of the prepreg was designed to mimic processing conditions in industry.

Darcy’s law, the most standard model used to describe permeability and its extensions follow next.

### 2.4.1 Darcy’s Law

Darcy’s law is a standard approach to characterize porous materials (Chen & Penumadu, 2008). For gas-phase advection transport, the Darcy’s law flow model is generally used and considered to work well for low velocity gas transport (Gas Transport in Porous Media, 2006). In the majority of gas permeation studies on composite prepreg materials, the use of this model has been applied (Ahn et al., 1991; Arafath et al., 2009; Arafath, Louis, Fernlund, & Poursartip, 2010; Nam et al., 1995; Putnam & Seferis, 1995; Shim & Seferis, 1997; Tavares et al., 2009, 2010). Originally derived to describe water flow through pipes, it can be used for gas transport...
given the fluid flow is slow (laminar in nature), which is represented by a very small Reynolds number. Darcy’s law states that the rate of fluid flow velocity is proportional to the pressure differential driving the fluid flow, the permeability of the media, cross-sectional area of the porous media, and length of distance traveled. Equation [1] is a simplified generic form of Darcy’s law (Gas Transport in Porous Media, 2006).

$$\bar{u}_g = -\frac{k_g}{\mu_g} \nabla P_g$$

[1]

Where Darcy fluid velocity ($\bar{u}$) is related to material permeability (k), fluid dynamic viscosity ($\mu$), and pressure gradient ($\nabla P$). The subscript “g” denotes gas as the transport fluid.

Permeability may be determined by Darcy’s law using one of two approaches: a steady-state method, or a non-steady-state method (Gas Transport in Porous Media, 2006). In the steady-state approach, permeability is determined by measuring the constant volumetric fluid flow rate through the porous medium. In the non-steady-state approach, a pressure-decay is used to determine permeability; where a pressure differential is set to either side of a porous medium, and both sides are let to equilibrate to one another. Permeability is then determined by the rate of pressure equilibration between the two sides. In this study, the more direct steady-state approach is used to measure permeability of prepreg laminates. The derivation of the steady-state equation from Darcy’s law is shown later in the experimental section 3.1.

2.4.1.1 Carman-Kozeny

Relating permeability K to the effective pore size (fluid channel path) is the Carman-Kozeny equation. Popular in liquid permeability studies of composites, this relationship is valid for slow seepage conditions through porous media. This equation relates permeability to the effective path
fluid travels through the porous medium using a geometric term. Permeability parameter ‘k’, from the Darcy equation, is related to the effective pore hydraulic radius $r_{\text{eff}}$ and geometric term $F$ (Equation [2]) (Pape, Clauser, & Iffland, 1999).

$$k = \frac{r_{\text{eff}}^2}{8F}$$  \[2\]

This geometric term $F$, relates tortuosity ($T$), or the amount of winding, the fluid flow path takes, to the porosity ($\phi$) of the medium (Equation [3]) (Pape et al., 1999).

$$F = \frac{T}{\phi}$$  \[3\]

The geometric term $F$ is usually determined experimentally, and was not done for the MTM45-1/CF2426A prepreg laminates evaluated in this thesis. The Carman-Kozeny equation and its geometric term are mentioned for discussing results obtained for through-thickness laminate permeability later in this thesis.

Several extensions of Darcy’s law are mentioned next, accounting for laminar shear flow at the boundary wall and possible turbulent fluid flow. Application of these extensions is typically only of significance in specific scenarios. Previous gas permeation studies on composite prepgs have ignored these extensions and will also be disregarded in this study.

### 2.4.2 Brinkman

An extension to Darcy’s law, the Brinkman extension takes into account the frictional effects at the boundary wall. Shown in Equation [4], a shear term is added (see the right hand side of the
equation) to the Darcy equation to account for a boundary shear effect on the fluid flow velocity (Gas Transport in Porous Media, 2006).

\[ \nabla P_g = -\frac{\mu_g}{k_g} \overline{u}_g + \overline{\mu} \nabla^2 \overline{u}_g \]  

[4]

The shear term is only considered significant for regions near the shear boundary. In the case of gas transport, the threshold of the application of this extension is considered when the boundary thickness is of the order of the square root of the permeability, \( k^{1/2} \) (Gas Transport in Porous Media, 2006).

2.4.3 Forchheimer

In scenarios where gas flow is turbulent, the Forchheimer extension is used (Equation [5]) (Gas Transport in Porous Media, 2006). The application of this equation is only considered valid when the Reynolds number is in the range of 1 to 10. Using the experimentally obtained permeability in this study, the Reynolds number in these experiments was found to be in the range of \( 10^{-5} \ll 1 \) (the full calculations of the Reynolds number can be found in Appendix C). With a low Reynolds number, using Darcy’s law on its own is considered sufficient.

\[ \nabla P_g = -\frac{\mu_g}{k_g} \overline{u}_g - C_F \frac{k_g^{-1/2}}{\rho_g} |\overline{u}_g| \overline{u}_g \]  

[5]

In the equation, \( C_F \) is an experimental constant that is a function of the porous medium. In turbulent flow conditions, it ranges typically from 0.1 to 0.55. In the case of non-turbulent flow, Darcy’s law, \( C_F = 0 \).
3. EXPERIMENTS

To characterize a MTM45-1/CF2426A prepreg laminate’s ability to vent gases and other volatiles, several experiments were developed. Permeability testing was done to quantify the internal gas transport pathways. Laminate behaviour under vacuum evacuation, where entrapped gas is removed via these pathways, was observed through a series of compaction tests. Microscopy and a developed water visualization test allowed the nature of these gas transport pathways to be visualized.

3.1 Experimental Methods

3.1.1 Permeability Testing Using Darcy’s Law

Permeability is a measure of the ability of a porous material to transport fluids and gases. In this study, gas permeability was measured using a steady-state gas flow test with data reduced using a Darcy’s law 1-D gas transport model. Air flow through laminate samples was induced by imposing a pressure differential over opposing sides of the laminate. 1-D gas flow can be idealized as a tube analogy illustrated in Figure 3-1.

Illustrated as atmospheric $p_a$ and vacuum $p_v$, a pressure differential is induced on either end of a tube of a permeable material by drawing hard vacuum on one end and pulling in atmospheric conditioned air through the other. Because $p_v$ is lower in pressure than $p_a$, gas flows through the
tube in the direction of \( p_v \). The rate of gas transport through the tube is a function of the tube cross-sectional area, the length of gas transport, and the ability of the porous medium to transport gas. This later property is known as permeability. Adapting this analogy to measure the permeability of a composite laminate, the tube analogy for 1-D gas flow is extended as shown in Figure 3-2.

![Figure 3-2. 1-D Gas Flow Through a Laminate (In-Plane Direction).](image)

For permeability tests, the pressure differential needed to drive air flow was created as previously described by drawing hard vacuum \((p_v = 30 \text{ inHg})\) on one side of a laminate and pulling in ambient pressured air \((p_a)\) through the other side.

Under steady-state flow conditions, gas permeability was calculated for tested laminates by measuring only a few parameters. From Darcy’s law of fluid flow, the derivation for 1-D fluid flow through a porous medium at atmospheric pressure is as follows (Arafath et al., 2009):
Chapter 3 Experiments

\[ Q = -\frac{AK}{\mu} \frac{dp}{dx} \]  \hspace{1cm} \text{[6]}

Using the assumption of ideal gas law:

\[ pQ = nRT = C \text{ (constant)} \]  \hspace{1cm} \text{[7]}

where the variables are defined:

\[ n = \text{molar flow rate of air [mol/s]} \]

\[ R = \text{gas constant [8.314 J/Kmol]} \]

\[ T = \text{absolute temperature of the air [K]} \]

Inserting Equation [6] into Equation [7]:

\[ \frac{C}{p} = -\frac{AK}{\mu} \frac{dp}{dx} \]  \hspace{1cm} \text{[8]}

Rearranging and integrating both sides:

\[ \int_{0}^{L} Cdx = -\int_{p_{a}}^{p_{v}} \frac{AK}{\mu} pdp \]  \hspace{1cm} \text{[9]}

\[ CL = -\frac{AK}{2\mu} p_{v}^{2} - p_{a}^{2} \]  \hspace{1cm} \text{[10]}
Chapter 3 Experiments

\[ C = \frac{AK}{2\mu L} \left( p_a^2 - p_v^2 \right) \]  \[11\]

At atmospheric conditions (indicated by subscript a):

\[ p_a Q_a = C \]  \[12\]

\[ p_a Q_a = \frac{AK}{2\mu L} \left( p_a^2 - p_v^2 \right) \]  \[13\]

\[ Q_a = \frac{AK}{2\mu L} \left( \frac{p_a - p_v^2}{p_a} \right) \]  \[14\]

where the variables are defined as follows:

\( A \) = cross sectional area of the laminate sample [m\(^2\)]

\( L \) = length of the part [m]

\( K \) = permeability [m\(^2\)]

\( \mu \) = dynamic viscosity [Pa·s]

\( p_a \) = atmospheric pressure [Pa]

\( p_v \) = vacuum pressure [Pa]
\(Q_a\) = volumetric gas flow rate measured at \(p_a\) [m\(^3\)/s]

The dynamic viscosity of gas (in this case air) \(\mu\) is considered constant at ambient conditions (1.82E-5 Pa·s). Therefore, by applying the Darcy’s law and using Equation [14], the permeability could be calculated by measuring the following:

**Table 3-1. Measured and Calculated Parameters for 1-D Steady-State Darcy's Law Flow.**

<table>
<thead>
<tr>
<th>Measured:</th>
<th>Calculated:</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A =) cross sectional area [m(^2)]</td>
<td>(K =) permeability [m(^2)]</td>
</tr>
<tr>
<td>(L =) length of sample [m]</td>
<td></td>
</tr>
<tr>
<td>(p_a =) atmospheric pressure [Pa]</td>
<td></td>
</tr>
<tr>
<td>(p_v =) vacuum pressure [Pa]</td>
<td></td>
</tr>
<tr>
<td>(Q_a =) volumetric flow rate measured at (p_a) [m(^3)/s]</td>
<td></td>
</tr>
</tbody>
</table>

The calculation of permeability from Equation [14] is based on several simplifying assumptions: that the cross-sectional area and the volume of the vascular network (gas transport paths) remain constant during the test, that the gas flow through the prepreg is assumed to obey Darcy’s law, and that the humidity and any related off-gassing due to partial vacuum pressure is assumed negligible.
Due to the fibrous and usually oriented nature of composites, their permeability is highly anisotropic (Gebart & Lidström, 1996; Hoes et al., 2002). For permeability tests, gas flow was isolated to 1-D flow in the two specific flow directions so that they could be independently studied; in-plane (Figure 2-1) and the through-thickness (Figure 2-2).

Under real processing conditions, in-plane gas transport can occur in two directions as shown in Figure 3-3. However, for use of the 1-D Darcy flow model, permeability was measured with gas flow restricted to only one of the two available flow directions. Due to symmetry in the five-harness satin weave, gas flow is expected to behave similar in the 0° and 90° directions (illustrated in Figure 3-3 as 1, 2).

Additionally, gas flow was not isolated to inter-laminar or intra-laminar flow paths as shown in Figure 2-1. As tested, the calculation of gas permeability using Darcy’s law cannot determine where gas flow is occurring. Instead, the laminate is treated as homogenous porous medium.

In the through-thickness gas flow direction, any effect of in-plane gas transport is also ignored. Similarly to the in-plane gas flow direction, the laminate is treated as a homogenous medium. The experimental set-ups are described below.
3.1.1.1 In-Plane

The experimental set-up to measure in-plane gas permeability was based on work by Shim and Seferis (1997) and by Arafath et al. (2009; 2010). Ambient condition in-plane permeability testing was performed using the set-up in Figure 3-4.

Sealant tape was used to form a three compartment test set-up. Each compartment was isolated so that different pressure states could be applied. By continually venting the vacuum port, the vent chamber (left compartment as pictured) was under atmospheric pressure conditions.
Connected to a vacuum pump, the vacuum chamber (right compartment as pictured) was under full vacuum. Air pressures were measured using a vacuum gauge on a vacuum regulator (Torr Technologies, Inc. MODEL No. 490200) and a barometer on the atmospheric side (Oregon Scientific MODEL No. BAR898HGA), respectively. The middle chamber was used to ensure air flow through the laminate was 1-D. A vacuum gauge monitored vacuum pressure to ensure that the sides, top, and bottom of the laminate sample were sufficiently sealed. If insufficiently sealed, any escaping air from the sample would have been detected by a loss in vacuum pressure in the middle compartment.

The laminate test sample was placed in the centre chamber with ends extending into the vent and vacuum chambers to act as an air bridge between the two. The set-up was sealed so that the only connection between these two chambers was the porous laminate. Therefore, by creating a pressure differential between the vent and vacuum chamber, air flowed between the two compartments by traveling through the laminate in the in-plane direction. Figure 3-5 illustrates the manner in which 1-D gas flow through a laminate sample was performed.
Figure 3-5. 1-D In-Plane Gas Flow Through a Laminate Sample for In-Plane Permeability Tests.

Air was drawn into a laminate from the left compartment where it flowed in the in-plane direction for a known length into the right compartment. Hard vacuum \( p_v \) (usually 30 inHg) was applied on one end while the other side was exposed to atmospheric conditioned air \( p_a \). Sealant tape (AT-200Y, Airtech advanced materials group) was used to cover the top, bottom, and side regions of the laminate so that no air escaped the laminate. Within this sealed region of the laminate, all gas transport, was known to be 1-D in the in-plane direction. Knowing the cross-sectional area and length over which 1-D gas flow occurs, and parameters such as pressure differential, permeability was calculated by simply measuring the gas flow rate through the sample.

Laminate samples tested were of nominal size \( L = 101.6\text{mm}, W = 101.6\text{mm} \) (4in x 4in). Within the sealed region, the sample test area was assumed to be \( L = 50.53\text{mm} \times W = 101.6\text{mm} \) (2in x 4in). Samples were laid-up and then cut with a knife to ensure uniform size. Laminate samples 4-
plies or greater were intermittently debulked every 4-ply for 7 minutes (Advanced Composites Group recommends debulking for 5-10 minutes every 3-4 plies). Laminate samples less than 4-ply were not debulked. Once prepared, samples were placed and sealed in the test set-up described.

To test that each chamber was independent from one another (Figure 3-4), a simple system leak test was performed prior to taking permeability measurements. The procedure for this is found in Appendix A. Once the set-up was verified to have no air leaks, in-plane permeability testing was performed using the procedure in Appendix B. Air flow through the laminate was measured with digital mass flow sensors on loan from Convergent Manufacturing Technologies (Vancouver, Canada). The sensors were placed at both the vent and vacuum ports. If the experiment set-up had no leaks, the measured mass flow of air would be the same for both sensors. Air flow was monitored using the mass flow sensors until a steady-state flow condition was reached. Typically, this was achieved in less than 5 minutes. With the steady-state gas flow, permeability for the laminate calculated using Darcy’s law (Equation [14]).

The length L of 1-D gas transport was assumed to be the sealed region of 50.8mm (2in) in length. However, as illustrated in Figure 3-5, the actual laminate sample extended out 25.4mm (1in) as tabs on either side of the sealed area. The entry and exit points for air flow within these tabs was unknown (Figure 3-5). In these regions, air was able to enter and leave the sample on the top and bottom surfaces (although this was found to be negligible in through-thickness testing), and in-plane on the sides of the laminate. As a result, the actual distance of 1-D gas flow is unknown. 1-D gas transport may in fact have been up to as long as 101.6mm (4in) the full length of the test sample. Using the longer 101.6mm length to calculate permeability would have resulted in the calculated permeability K being larger by a factor of 2. Because the test length of
50.8mm was the only region where 1-D gas flow was guaranteed, this length was used in the permeability calculations.

In addition to carrying out permeability testing under ambient conditions, heated in-plane permeability tests were performed by placing the in-plane permeability set-up on top of a custom made heating plate (Figure 3-6). This set-up provided tool side heating of the sample while simultaneously allowing permeability measurements to be made.

![Figure 3-6. Heated In-Plane Permeability Test Set-Up.](image)

The hot-plate consisted of a custom made flat heater and a stack of aluminum plates. The aluminum plates were added to provide additional thermal mass to the set-up to minimize temperature fluctuations and localized temperature gradients across the test tool surface. Heat was provided by a custom made 240V, 2800W heater controlled by solid state relay control with a temperature controller. Using the set-up, constant heating (ramp) rates and temperature holds
were imposed on a laminate while simultaneously measuring in-plane permeability. Laminate temperature was measured using vacuum sealed Type J thermocouples placed on both the top and bottom surfaces of the laminate. Other than the addition of heating the sample, heated permeability tests were carried out in the same manner as the ambient condition in-plane tests.

3.1.1.2 Through-Thickness

The experimental set-up used to measure through-thickness permeability was conceptually similar to the in-plane test set-up. By applying different air pressures on either side of the laminate sample (top and the bottom), gas flow though the sample in the thickness direction is induced. The through-thickness permeability tests were performed using the set-up shown in Figure 3-7.

![Figure 3-7. Through-Thickness Permeability Test Set-Up.](image)

Similar to the in-plane test, the pressure differential on either side of the sample was created by venting the bottom of the laminate to ambient air $p_a$, and drawing hard vacuum over the top surface $p_v$ (Figure 3-8).
Figure 3-8. Schematic of the Test Sample and the Pressure Differential Causing Air Flow in the Through-Thickness Direction Test Set-Up.

Test samples nominally 82.55mm x 82.55mm (3.25in x 3.25in) were placed directly onto a square piece of porous refractory brick of the same size. Ambient air was drawn up through a hole in the steel tool and into the refractory brick, where it was distributed across the entire bottom surface of the laminate sample. The permeability of the refractory brick was 4.16E-16 m², which was much greater than that of the prepreg. Therefore it was assumed the refractory brick did not have a substantial influence on the permeability measurement of the laminate. To ensure air flow was in the through-thickness direction, the sides of the laminate and the refractory brick were sealed with sealant tape. A traditional vacuum bag set-up was then employed to seal and enclose the laminate sample (with refractory brick support) to the standing tool rig set-up. Like the in-plane permeability test, air flow was measured at the air vent and vacuum ports using digital mass flow sensors. Appendix C details the test procedure to perform through-thickness permeability tests.
For heated through-thickness permeability testing, the through-thickness set-up was placed and heated inside a laboratory convection oven. The objective of this test was to observe the gas flow behaviour in the through-thickness direction during a cure cycle. Due to limitations of the oven used, precise temperature heating rates could not be achieved. Instead, a vacuum sealed Type J thermocouple was placed on the sample and the heating rate of the sample was recorded. Thermocouple data showed that heating rates were relatively constant between 2.2-3.4°C/min.

For the heated tests, laminate samples were placed directly onto the steel tool rig without the porous refractory brick. To prevent air from escaping in the in-plane direction, the sides of the laminate were sealed with sealant tape. Vacuum bagging of the test set-up was the same as for the ambient condition test. Due to the lack of support without the refractory brick under the laminate sample, a piece of compressed breather material was placed inside and supported within the hole in the tool. Because the tool hole opening was not the same size as the laminate test sample, through-thickness permeability could not be calculated as the exact area for gas flow was unknown. However, the test still allowed for evaluating the effect of temperature on gas flow in the through-thickness direction of the prepreg laminates.

### 3.1.1.3 Ply Terminations

When making tapered laminates, plies are typically terminated internally in staggered incremental steps while being protected by continuous skin plies (Figure 3-10). Laminate breathing is made more difficult at ply terminations (sometimes referred to as ply-drops), and may lead to pockets of entrapped air (Mallow & Campbell, 2000). To test the effect of ply terminations on in-plane gas transport, the in-plane permeability set-up was used (Figure 3-9). However, instead of a laminate of constant thickness, an 8-to-2 ply laminate sample was tested.
Like the in-plane test, the test area was nominally of length $L = 50.8\text{mm}$, and width $W = 101.6\text{mm}$.

Figure 3-9. In-Plane Ply Termination Permeability Test Set-Up.
Within the 50.8mm (2in) long effective test area, the laminate was 8-plies in thickness for the first 12.7mm (0.5in) followed by the dropping of internal plies at 6.35mm (0.25in) increments resulting in a 2-ply laminate for the last 6.35mm (0.25in). The laminate was debulked intermittently for 7 minutes every 4-plies during lay-up.

3.1.1.4 In-Plane Debulking Effect

An extension of the in-plane permeability test, the effect of debulking, i.e. applying full vacuum to a laminate to aid in compaction and tool conformation, on the in-plane permeability of MTM45-1/CF2426A laminates was also examined. The in-plane permeability test set-up as previously described was used to test an 8-ply laminate sample. To impose an effective debulk condition on the sample, full vacuum was pulled on all three compartments (see Figure 3-4). Permeability measurements were then made intermittently by venting one of the sample chambers after a defined time of debulking as previously described. Initially, the 8-ply sample was not debulked with the exception of the time required at full vacuum to perform the in-plane test set-up leak test. At this point a permeability measurement was taken representing time = 0. After this, debulking took place for specified time increments followed by permeability measurements. Debulking of the laminate sample was carried out over 24 hours.
3.1.2 Laminate Compaction Tests

When placed under vacuum, uncured composite laminates are observed to compress and reduce in thickness. During the heated cure process, the laminate is further compressed, with the final laminate essentially being void free. Figure 3-11 illustrates the evolution of laminate thickness with respect to stages of the cure cycle.

![Diagram of laminate thickness at various points during cure cycle](chart.png)

Figure 3-11. Laminate Thickness at Various Points During Cure Cycle (Temperature vs. Time).

It was proposed by Nam et al. (1995) that compaction of a laminate also results in the reduction in the void fraction of a laminate, resulting in a reduction in air permeation. As illustrated at the top of Figure 3-11, the support provided by the entrapped gas is lost as the gas is removed during vacuum evacuation.
3.1.2.1 Compaction Experiment #1

In Compaction Experiment #1, laminate compaction was measured over a 24 hour debulking period. By measuring laminate thickness after lay-up, and calculating the final cured laminate thickness from data provided by the prepreg manufacture (assumed to be a zero void content), the initial void content of the laminate was approximated. Comparing the compaction data (and the reduction in void content) to the in-plane permeability debulking effect test data, the relationship between laminate compaction and permeability reduction could be evaluated.

In this experiment, a laminate sample was debulked to a flat tool surface. Thickness measurements were taken at the centre of the of the sample surface throughout the experiment. A small High Density Polyethylene (HDPE) caul sheet was placed on top of the laminate to help negate the effect of surface texture inherent with the five-harness satin weave, to allow thickness measurements to be made. The vacuum bagging consumables were that recommended for processing by the prepreg manufacturer ACG (Figure 1-3) (ACG, 2007). A Teflon release layer followed by breather was placed over the sample. To ensure sufficient gas evacuation peel-ply strips were placed at the laminate edge. A cut-out in the bagging consumables (Teflon and breather) was made to allow for the caul sheet placement and thickness measurements to be made. The entire sample was vacuum bagged in standard manner with a vacuum port placed 101.6mm (4in) away from edge of the laminate. Figure 3-12 illustrates the experiment lay-out.
Initially, a partial vacuum of 10 inHg was initially pulled on the sample to ‘set’ the vacuum bag allowing an initial thickness measurement to be made. The sample was left under this state for 105 minutes allowing the pressure internally within the laminate sample to normalize and distribute prior to testing. No further debulking was performed on the laminate prior to the test in order to maximize the thickness change observed during the experiment. Thickness measurements were made using a dial gauge (Mitutoyo MODEL ID-C125EB) at the centre of the laminate.
3.1.2.2 Compaction Experiment #2

Compaction Experiment #2 was designed to evaluate the effect of inhibited edge breathing. This would give some insight whether or not the observed laminate compaction under vacuum evacuation was at least in part related to gas removal. In the test set-up (Figure 3-13), the breathing of long laminates was controlled to only occur at one edge. Thickness measurements were made to measure laminate compaction on two variations of this test. In one configuration, edge breathing was left uninhibited, while in the other configuration a portion of the breathing edge was purposely inhibited.

The laminate samples tested were 1 metre long, 8-ply laminates placed onto an aluminum tool and vacuumed bagged. No debulking was performed prior to testing in order to maximize the thickness change observed during the experiment. The laminate samples were sealed on all sides (using sealant tape) with the exception of one free edge, where a vacuum port was placed to provide vacuum evacuation. A HDPE caul sheet was placed on the top running the entire length of the laminate. The sealant tape and caul sheet restricted the internal gas transport to a 1-D flow situation. The HDPE caul sheet placed on top of the laminate had two roles: 1) To seal the top of the laminate from through-thickness gas transport. 2) To provide a flat surface so that thickness measurements could be made. The laminate thickness was measured using several dial gauges (Mitutoyo MODEL ID-C125EB, Mitutoyo MODEL ID-C150EB, and Starrett No. 656-617) placed at several locations along the length of the laminate. Several layers of stacked breather were butted against the open end of the laminate to provide edge breathing. The vacuum port was placed 76.2mm (3in) from the free edge of the laminate (where breathing occurred). The experimental set-up is illustrated in Figure 3-13.
To ‘set’ the vacuum bag prior to testing, an initial partial vacuum of 5 inHg was pulled on the bagged set up to conform the bag to the sample and caul sheet, and to allow an initial thickness measurement (time = 0) to be made. The partial vacuum was left for 135 minutes to allow for the pressure within the long laminate sample to normalize (time dependent) prior to the start of the test. The two edge breathing configurations are shown in Figure 3-14.
In Configuration #1, only one layer of breather was butted up against the free edge of the laminate. As vacuum was pulled, the vacuum bag was pulled down overlapping and effectively sealing the top half of the laminate. This left only the bottom half of the laminate edge open for edge breathing. In Configuration #2, three layers of breather were stacked and butted against the laminate providing sufficient gas evacuation across the entire breathing edge. By comparing the two scenarios, the importance of proper open uninhibited edge breathing was evaluated.

### Microscopy

Using several methods, the internal gas transport (pathways) microstructure of the MTM45-1/CF2426A prepreg laminates was examined. Images were simply taken using a Nikon D100 SLR camera with a macro lens. Additional, micrographs were taken using Optical Microscopy (OM) and Scanning Electron Microscopy (SEM).
Obtaining micrographs of laminate samples in the ‘as-laid-up’ state proved to be difficult due to the soft nature of the prepreg material. Micrograph images of this process state have been reported in literature (Shim & Seferis, 1997) but no details were given of how exactly this was achieved. Any cutting in this soft state resulted in fibre pull-out, breakage, and resin smearing. Due to the need of a clean and flat prepared surface, samples were fully cured at low temperature without vacuum (80°C to reduce resin flow, followed by 180°C post cure). The hardened samples were then cut with a diamond saw. For SEM examination, due to the depth of field of the SEM, surface preparation was not necessary. Therefore, uncured samples were simply cut with a sharp knife, and viewed as-laid-up.

### 3.1.4 Water Visualization

To visualize the gas transport pathways a water visualization test was developed. In this test, air was pumped into laminate samples while they were submerged in water. The escaping air was then visualized in the form of bubbles. The test allowed for qualitatively comparing the competing in-plane and through-thickness gas evacuation paths in the laminate.

Samples tested were nominally 101.6mm (4in) in length. Air was forced into the sample by vacuum bagging one end of the sample. The vacuum bag chamber (Figure 3-15) was 50.8mm (2in) of the sample length. Air was constantly supplied into the sample through a tube connected to a standard aquarium fish tank pump.
The breather, illustrated in Figure 3-15, was used to promote even air distribution into the laminate. Air was supplied to the sample at a constant flow rate of 5.7 mL/min, controlled using an air flow control valve. The flow rate was verified using a volumetric flow rate meter (OMEGA 17190).
Figure 3-16. Water Visualization Test Set-Up.

Video was recorded using an underwater digital camera (Olympus Stylus 720 SW). Time elapsed storyboard images were then produced from the recorded video using open source video software KMPlayer.

3.2 Experimental Test Plan

Permeability testing of MTM45-1/CF2426A prepreg laminates represented the major component of the test plan for this thesis. Supplementary experiments such as laminate compaction, microscopy, and the water visualization test were conducted in order to gain additional understanding of gas transport.

3.2.1 Permeability Testing

The test matrix for in-plane testing examined several factors relevant in processing of composite structures. The effects of processing factors on in-plane permeability studied were: numbers of
plies, debulking, ply terminations, and temperature. The test matrix for in-plane permeability testing is shown in Table 3-2. For all in-plane permeability tests, a minimum of three repeat tests were performed. In some cases, more than three tests were carried out. In cases where no permeability was measured ($K = 0$), testing was stopped after two repeats.

**Table 3-2. In-Plane Permeability Test Matrix.**

<table>
<thead>
<tr>
<th>In-Plane Permeability</th>
<th>Plies</th>
<th>Ramp Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1°C/min</td>
</tr>
<tr>
<td></td>
<td>21°C (ambient conditions)</td>
<td></td>
</tr>
<tr>
<td><strong>Ambient</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>X</td>
<td>N/A</td>
</tr>
<tr>
<td>2</td>
<td>x</td>
<td>N/A</td>
</tr>
<tr>
<td>3</td>
<td>x</td>
<td>N/A</td>
</tr>
<tr>
<td>4</td>
<td>x</td>
<td>N/A</td>
</tr>
<tr>
<td>8</td>
<td>x</td>
<td>N/A</td>
</tr>
<tr>
<td>8-to-2</td>
<td>x</td>
<td>N/A</td>
</tr>
<tr>
<td><strong>Heated</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>4</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>8</td>
<td>x</td>
<td>x</td>
</tr>
</tbody>
</table>

Like the in-plane test matrix, the effect of the number of layers for through-thickness permeability was also investigated. Test samples were heated to 80°C, after which gas flow exceeded the measurement range of the mass flow sensor equipment. Due to the limit of control
capability of the test equipment available, setting a pre-described heating rate was not possible. However, thermocouple data showed that sample heating rates were consistently between 2.2-3.4°C/min. A minimum of two repeats were performed for all tests with the exception of 1-ply and 2-ply ambient condition test which showed larger sample to sample variability. Table 3-3 shows the through-thickness test matrix.

Table 3-3. Through-Thickness Permeability Test Matrix.

<table>
<thead>
<tr>
<th>Plies</th>
<th>Ramp Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>21°C (ambient conditions)</td>
</tr>
<tr>
<td></td>
<td><strong>Ambient</strong></td>
</tr>
<tr>
<td>1</td>
<td>x</td>
</tr>
<tr>
<td>2</td>
<td>x</td>
</tr>
<tr>
<td>4</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td><strong>Heated</strong></td>
</tr>
<tr>
<td>1</td>
<td>N/A</td>
</tr>
<tr>
<td>2</td>
<td>N/A</td>
</tr>
<tr>
<td>4</td>
<td>N/A</td>
</tr>
</tbody>
</table>

3.2.2 Laminate Compaction Testing

Prior to commencing the gas evacuation length test, partial vacuum was applied for a set period of time (bag set) to conform the vacuum bag to the sample, in addition to uniformly distributing the partial pressure within the laminate.
Table 3-4. Laminate Compaction Test - Experiment #1 Test Matrix.

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Bag Set</th>
<th>Test Duration</th>
<th>Measurement Location</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Vacuum</td>
<td>Time (min)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(inHg)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4&quot;x4&quot;, 8-Plies</td>
<td>5</td>
<td>105</td>
<td>24 hours</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Centre</td>
</tr>
<tr>
<td>6&quot;x6&quot;, 8-Plies</td>
<td>5</td>
<td>105</td>
<td>24 hours</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Centre</td>
</tr>
</tbody>
</table>

Table 3-5. Laminate Compaction Test - Experiment #2 Test Matrix.

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Bag Set</th>
<th>Test Duration</th>
<th>Measurement Locations</th>
<th>Breather Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Vacuum</td>
<td>Time (min)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>(inHg)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>W = 4&quot;, L = 39&quot;, 8-Plies</td>
<td>5</td>
<td>135</td>
<td>1&quot;, 5&quot;, 10&quot;, 20&quot;, 30&quot;, 38&quot;</td>
<td>Configuration #1</td>
</tr>
<tr>
<td>W = 4&quot;, L = 39&quot;, 8-Plies</td>
<td>5</td>
<td>135</td>
<td>5&quot;, 20&quot;, 30&quot;</td>
<td>Configuration #2</td>
</tr>
</tbody>
</table>

### 3.2.3 Microscopy

Microscopy images were taken from samples of different process history conditions: 1) the as laid-up microstructure of the prepreg, 2) the microstructure of prepreg cured without vacuum applied, and 3) the microstructure of the in-plane permeability tested prepreg. For the third
condition, the test samples were fully cured using two different curing conditions. The sealant tape surrounding the samples during the in-plane permeability testing was removed using an adhesive remover (DeBond Corp Marine Formula). This adhesive remover is specifically formulated to remove adhesive residue while not attacking the epoxy matrix.

### Table 3-6. Microscopy Test Matrix.

<table>
<thead>
<tr>
<th>Sample History</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SEM</td>
</tr>
<tr>
<td>As laid up</td>
<td>x</td>
</tr>
<tr>
<td>cure, no vacuum</td>
<td></td>
</tr>
<tr>
<td>In-plane permeability test, cured 120°C hold no vacuum</td>
<td>x</td>
</tr>
<tr>
<td>In-plane permeability test, cured 120°C hold full vacuum</td>
<td>x</td>
</tr>
</tbody>
</table>

### 3.2.4 Water Visualization

Water visualization testing was performed on select samples with varying process histories. Samples tested represented those after lay-up prior to a heated cure processing and several cured samples whose vacuum was interrupted at various points in the cure process cycle. Interrupting vacuum during the curing process was done to ‘freeze’ the microstructure of the samples at that
point in the process history. The samples were chosen to examine the evolution of change of gas extraction pathways out of the prepreg laminate during processing.

**Table 3-7. Water Visualization Test Matrix.**

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Hours of cure until vent</th>
<th>Notes:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lay-up #1</td>
<td>N/A</td>
<td>As-laid-up</td>
</tr>
<tr>
<td>Lay-up #2</td>
<td>N/A</td>
<td>As-laid-up</td>
</tr>
<tr>
<td>Vent #4</td>
<td>2 hr</td>
<td></td>
</tr>
<tr>
<td>Vent #7</td>
<td>16 hr</td>
<td></td>
</tr>
<tr>
<td>Vent #8</td>
<td>22 hr</td>
<td></td>
</tr>
</tbody>
</table>

Samples Lay-up #1 and #2 were freshly laid-up which represented the laminate gas extraction pathways condition prior to processing. However, the uncured resin softened with increasing time submerged in the water. This was evident after prolonged periods submerged in water by the formation of gas pathways which were not present initially. This included the formation of multiple through-thickness gas evacuation paths from the laminate sample. As a result, testing was only appropriate for a few minutes after which the resin softened to the point where the microstructure was considered to no longer be representative of the ‘as-laid up’ state. Cured samples Vent #4, 7, 8, were cured at 80°C for 20 hours. The low temperature cure was chosen to reduce resin flow and preserve the microstructure of the prepreg.
4. RESULTS

4.1 Permeability Testing

Permeability tests were performed to quantify the internal gas extraction pathways in MTM45-1/CF2426A prepreg laminates. The effect of the number of laminate plies on permeability in the in-plane, and through-thickness directions under ambient and heated conditions were examined. The effect of debulking and ply-terminations on the in-plane permeability was also investigated.

4.1.1 In-Plane

Although measured in 1-D, in-plane permeability has two directions; longitudinal and transverse, making it 2-D in most real vacuum gas evacuation situations. If edge breathing is provided on all sides of the laminate, this effectively doubles the measured breathability. The results obtained under ambient condition represent gas extraction prior to the heated curing process, while those under heated conditions represent the permeability of the laminate during the heated curing process.

4.1.1.1 Ambient Condition

Under ambient conditions, the effect of the number of layers (or plies) in a laminate (thickness) was examined. A minimum of three repeat tests for each condition were performed to get an average permeability. In some test groups as many as six repeat tests were performed on individual samples. Each error bar represents +/- one standard deviation. Samples 4-plies or greater were debulked according to the specifications sheet provided by the prepreg manufacturer (ACG, 2007). Due to the symmetry of the five-harness satin weave, gas flow in the
0° and 90° directions of the laminate should be similar; therefore, the effect of various lay-up angles was not investigated.

![Figure 4-1. In-Plane Laminate Permeability. Three Repeats Minimum. Error Bars Denote +/- One Standard Deviation.](image)

The in-plane permeability of all the samples was found to be on the order of 1E-14 m², similar to that reported for other prepreg systems (Arafath et al., 2009; Arafath et al., 2010). The measured permeability is approximately independent of the number of layers, indicating a good agreement with Darcy’s law, i.e. permeability can be considered a material property for in-plane gas flow under ambient conditions for laminates of this prepreg.
Slight differences in permeability between the intermittently debulked and non-debulked samples were observed, with the debulked samples showing somewhat lower permeability. Samples with 4-plies or more were debulked according to the manufacturer’s recommendation, whereas no debulking was performed on the 1 and 2-ply samples. Intermittent debulking, as shown later in the long debulking test results, has the effect of reducing the in-plane gas permeability. Consequently, the prepreg manufacturer cautions against excessive debulking due to the risk of closure of gas extraction pathways (ACG, 2007).

### 4.1.1.2 Heated Condition

The test matrix for in-plane gas permeability testing at ambient conditions was expanded to include the effect of temperature on permeability. The tests examined the effect of increasing the number of layers in the laminate as well as the effect of heating rate. Due to the dynamically changing internal structure of the laminate under heated conditions (results to follow), a steady-state gas flow condition through the sample is not reached.

By the steady-state definition, the heated in-plane test does not allow for the permeability to be calculated. Instead of permeability, presenting the collected data as gas flow is more accurate based on the testing conditions. However, presenting as air flow does not allow for a normalized comparison due to the different sample geometries tested, where the thicker the laminate stack has a greater volume of in-plane air flow. Therefore, the results are presented as ‘nominal permeability’, which is calculated assuming steady-stage gas flow, for the sake of comparison. Additionally, for comparative reasons, the temperature used in the analysis is that of the bottom laminate surface (heated surface). Temperature gradients between the bottom (heated) and top sample surfaces observed during testing are found in Appendix F.
4.1.1.2.1 Effect of Number of Layers

Examining the effect of the number of layers when heated, laminates of 2, 4, and 8-plies were tested with temperature ramp rates of 1, 2, and 3°C/min.

![Graph showing heated in-plane nominal permeability vs. temperature (1°C/min Heating Rate).](image)

**Figure 4-2.** Heated In-Plane Nominal Permeability vs. Temperature (1°C/min Heating Rate).

Due to sample to sample variability (prepreg material itself and also lay-up variability – operator dependent) all three samples had slightly differing initial permeabilities. As temperature increases, the in-plane permeability was found to decrease. In comparison to the starting permeability at ambient conditions, the in-plane permeability was found to drop as much as half.
Although the permeability decreased, gas flow was not found to completely close off and go to zero. At around 100°C, prior to all the internal gas transport pathways closing, gas permeability was found to suddenly increase. The flow increased to beyond the capacity of the mass flow sensors. This is represented as vertical dashed lines in the data plots. The measured permeability can only be considered valid up until this point in the tests. The ‘opening’ of the sample is likely due to low resin viscosity at elevated temperature and will be discussed further in the discussion section.

Ignoring the ‘opening’ of the laminate beyond a critical temperature, the in-plane permeability was found to decrease with increasing temperature. A hypothesis for the decrease in permeability is that as temperature increases, uncured resin begins to flow due to a lower viscosity. Additionally, under vacuum conditions, an effective 1 atm force is applied to the laminate. With the air within the gas pathways being removed the unsupported pathways partially collapse. Likely, a combination of both the gas pathways filling with resin and pathways collapsing is occurring.
Similar to the 1°C/min tests, from 70°C until the samples ‘open up’, the rate of decrease in in-plane permeability is more pronounced for thinner laminates. The ‘opening’ temperature for samples heated at 2°C/min is higher than for the 1°C/min samples. This is likely attributed to the temperature gradient through the thickness of the sample being greater as the temperature heating rate is increased. Because the prepreg has relatively poor thermal conductivity, the heat flow within the sample is slower than the heating provided to the bottom surface of the laminate, leading to a through-thickness temperature gradient (Appendix F).
Figure 4-4. Heated In-Plane Nominal Permeability vs. Temperature (3°C/min Heating Rate).

The same trend with regards to the decrease of in-plane permeability with increasing temperature, and the number of plies in the laminate as previously described was also found for the 3°C/min tests.

4.1.1.2.2 Effect of Heating Ramp Rate

To examine the effect of heating rate on the in-plane gas permeability for a given laminate thickness, the following plots were generated.
It was observed that increased heating rate resulted in a slower decrease in in-plane permeability with respect to temperature. Again, this is attributed to the thermal gradient that exists in the thickness direction of the laminate with the one sided heating of the sample. As previously described, the low thermal conductivity of the prepreg combined with increased heating rate leads to an increase in thermal gradient. This was verified by comparing the bottom laminate surface (being heated) temperature with the recorded top surface temperature (see Appendix F).
Figure 4-6. Heated In-Plane Nominal Permeability vs. Temperature (4-plies).

The trend observed for the 2-ply samples was also observed for the 4-ply and 8-ply samples.
4.1.2 Through-Thickness

Results for the through-thickness laminate permeability under ambient conditions and heated conditions are presented next. The modifications to the test set-up in Figure 3-7 were only implemented in time to repeat through-thickness testing under ambient conditions. The heated test data are from the unmodified set-up and can only confidently be reported as gas flow, rather than as permeability, as the true effective area was unknown.


4.1.2.1 Ambient Condition

Similar to the in-plane direction, the effect of the number of layers on laminate permeability in the through-thickness direction was investigated.

![Graph showing permeability results for different numbers of plies.](image)

**Figure 4-8. Through-Thickness Permeability Results.**

Increasing the number of plies in the laminate is found to decrease permeability. 1-ply is found to have permeability in the range of $1 \times 10^{-18}$ m$^2$ under ambient conditions. At 2-plies the permeability is found to decrease to lower than $1 \times 10^{-20}$ m$^2$. At 4-plies or greater, laminates were found to have no measurable air flow. This sudden decrease in permeability with the increasing numbers of plies is in disagreement with Darcy’s law. According to Darcy’s law, permeability should be independent of the number of plies in the laminate stack.
4.1.2.2 **Heated Condition**

To test the effect of heating on through-thickness permeability, the entire through-thickness test set-up was placed in a heated oven. The through-thickness test set-up used in the heated experiments lacked the refractory brick sample support with a defined effective test area. Although the heated set-up did not have a known effective test area in order to allow for the true permeability to be calculated, the set-up still allowed the relation between air flow and temperature to be observed (where gas flow is related to permeability). The following study looks at the effect of temperature and laminate thickness on air flow through the laminate. The temperature presented is from a thermocouple (Type J) placed on the laminate sample.

![Figure 4-9. Through-Thickness Air Flow vs. Temperature (1-ply).](image-url)
For a 1-ply laminate, the air flow in the through-thickness direction increased with increasing temperature. Just prior to 45°C, the measured air flow became beyond the measurable flow range of the mass flow sensors used to record data.

Unlike the 1-ply samples, for the 2-ply samples the air flow remained low during initial heating, until just past 45°C. At this point, the through-thickness direction gas transport pathways in the prepreg began to ‘open up’ and the measured air flow increased rapidly. By 55°C, the air flow went beyond the measurement capabilities of the mass flow sensors used.
When tested at ambient conditions, samples of 4-ply had no air flow in the through-thickness direction. This trend was also observed during the initial heating of the 4-ply laminates. Like the 2-ply heated samples, air flow in the through-thickness direction remained low; in this case essentially zero flow. Between around 55°C and 65°C, air flow begins to increase. In one test the air flow began to fluctuate with increasing temperature until flow remained open. This is likely indicative of a dynamically changing resin structure, where continuous gas pathways relative to each stacked layer are opening and closing. For both samples, after a certain temperature the mass flow sensors were beyond their flow range and measurements could no longer be taken.
4.1.3 Ply Terminations

Due to the changing laminate thickness in the 8-to-2 plies ply termination (ply-drop) sample, results are reported as volumetric gas flow instead of a normalized permeability value. All samples were of the same length and had the same effective test area as those used in the in-plane permeability tests. Compared are gas flow results for 2-ply, 4-ply, 8-ply, and 8-to-2 ply laminate samples.

![Graph showing gas flow results for different numbers of plies.](image)

**Figure 4-12. Ply Termination Air Flow Results.**

Considering batch-to-batch variability and measurement accuracy, it was observed that the 8-to-2 ply laminates had similar gas flow to that of the 2-ply laminate samples. This indicates that the lowest number of plies at any point in the laminate stack is the limiting governing resistance in
in-plane gas transport. It should also be mentioned that in Figure 4-12, air flow was not found to scale with the number of laminate plies. The measured air flow for 4-plies and 8-plies are lower than expected if scaling is based off the 2-ply air flow value. This can be attributed to the differing processing states between the sample groups. Laminates 4-plies and greater were intermittently debulked according to the prepreg manufacturer’s processing instructions (ACG, 2007). This reduction in air flow, normalized in terms of permeability, is seen in Figure 4-1.

### 4.1.4 In-Plane Debulking Effect

The permeability presented in Figure 4-13 is normalized with respect to the initial permeability measured immediately after lay-up without any debulking. The gap in the data represents the over night period during which no measurements were taken.
It was found that almost half of the observed decrease in permeability occurred within the first hour. At the end of 24 hours, the in-plane permeability was still approximately 60% of the initial permeability of the sample, indicating the presence of significant open gas pathways. The hypothesis for this observation is that the uncured tacky resin cold-flows under vacuum and slowly begins to fill the interconnected pathways inside the laminate. After long periods of debulking, the remaining gas transport likely occurs within the dry fibre bundles not wetted out dby resin cold-flow. In order for these intra-tow pathways to close, sufficient resin flow must occur to impregnate the dry fibres, which likely implies the resin has to be heated to reduce the viscosity while a low air pressure is maintained within the dry fibre tows. This plateau shows
that internal gas transport in laminates of the MTM45-1/CF2426A prepreg will not close off under extended periods of debulking. This suggests that it is satisfactory to draw a hard vacuum for extended periods of time for maximum laminate gas evacuation prior to the cure process.

The observed change in in-plane gas permeability, with debulking, may be the result or combination of two factors: 1) As gas is evacuated from inside the laminate where the internal pressure is close to 0 atm at full vacuum, the internal gas transport pathways partially collapse. 2) Under vacuum pressure the resin flows into the dry fibre tows by cold-flow. This observed plateau in permeability may be the result of the first factor having reached its limit. Approaching 24 hours, it is hypothesized that the larger gas transport pathways have been evacuated and collapsed. Remaining gas transport is then limited to dry fibre regions where resin flow has not yet filled the void space within the tows. Any further changes in the in-plane permeability at this point would be the result of fibre wetting, which is temperature, vacuum pressure, and time dependent.

4.2 Laminate Compaction Testing

In the first experiment, a set of square laminates were debulked for 24 hours with thickness measurements being taken as a function of time. Thickness trends were compared to in-plane permeability debulking trends to evaluate if there was a correlation. In the second experiment, a controlled 1-D laminate gas evacuation was performed to look at the importance of free edge breathing. The effects of uninhibited edge evacuation and a purposely partial restricted gas evacuation scenario were compared.
4.2.1 Compaction Experiment #1

Compaction Experiment #1 simulated a typical debulking scenario. Here, edge breathing occurs on all four free edges of the laminate. Due to the low through-thickness permeability measured under ambient conditions, it was assumed that gas evacuation primarily occurs in-plane and therefore no sealing (full caul sheet or film adhesive) was applied to the top of the laminate sample. A small caul sheet was placed in the centre region so that thickness measurements could be taken. Two laminates of different sizes were tested, nominally 101.6mm x 101.6mm (4in x 4in) and 152.4mm x 152.4mm (6in x 6in). However, due to their similar and relative small sizes, there was little difference in gas evacuation times.

![Figure 4-14. Compaction vs. Debulk Time over 24 Hours (Compaction Experiment #1).](image-url)
The compaction vs. time curves for both samples showed similar behaviour. The slight offset in the compaction trends between the two samples is likely due to sample lay-up variability and not sample size differences. However, the final measured thickness of the two laminates after debulking for 24 hours were found to be the same. At this point, a compaction limit may have been reached. Comparing Figure 4-14 to the in-plane permeability trend during similar debulking in Figure 4-13, the two curves have a similar shape. The relationship between laminate compaction and the reduction in in-plane permeability during debulking is plotted in Figure 4-15.

![Figure 4-15. Normalized Laminate Thickness (Averaged Results from Samples in Compaction Experiment #1) vs. Normalized In-Plane Permeability (24 Hour In-Plane Debulking Test).](image-url)
Normalized laminate thickness (with respect to initial laminate thickness), averaged from the two laminate samples tested in Compaction Experiment #1, is plotted against normalized in-plane permeability (with respect to initial laminate permeability) over a 24 hour debulking period. This relationship supports the hypothesis put forth by Nam et al. (1995) where a reduction in interconnected laminate voids is reflected in a reduction in gas permeability.

### 4.2.2 Compaction Experiment #2

Figure 4-16 shows results from Compaction Experiment #2 where two different edge breathing configurations were evaluated. By comparing the compaction differences between the different edge breathing configurations, inferences could be made as to whether laminate compaction was in part influenced by entrapped gas removal, and the effect of inhibited edge breathing.

![Figure 4-16. Average Laminate Compaction vs. Time (Compaction Experiment #2).](image)
Chapter 4 Results

Both breather configurations exhibited a similar compaction vs. time trend. However, the observable compaction difference between the two configurations indicates that gas extraction is heavily influenced by obstructions or lack there of, at the free edge where gas evacuation the laminate occurs. In both the scenarios, the bulk of the laminate compaction occurs during the initial period of vacuum evacuation. In Configuration #2, where edge breathing is uninhibited, the laminate compacted nearly twice as much as the laminate in Configuration #1 over the full 24 hours. This suggests the inability for entrapped gas located at the upper plies in Configuration #1 to be transported (through-thickness) to the lower laminate plies where in-plane edge breathing is uninhibited. This hypothesis is supported by the through-thickness permeability measurements at ambient conditions, which show that gas transport in the laminate through-thickness direction is minimal.

4.3 Microscopy

Figure 4-17 shows a micrograph representing an ‘as-laid-up’ laminate cured using the low temperature 80°C method mentioned previously. The image was taken using a Nikon D100 SLR camera fitted with a macro lens. Illustrated at the bottom (Figure 4-17) is a cross-sectional schematic of a five-harness satin weave. Clearly visible is the ‘nesting’ that occurs between the laminate plies which form large inter-laminar macro-voids. Nesting due to prepreg surface topography is described by Ahn et al. (1991) as one possible source for inter-laminar gas permeation pathways for laminate gas transport.
Due to the nature of the five-harness satin weave, the uneven surface topography creates large inter-laminar macro-voids during lay-up. The formation of these voids is variable and dependent on how the raised tows from each ply nest relative to one another. In addition to macro-voids, another void type is smaller micro-voids. These are formed within individual fibre bundles where resin has not impregnated. Examples of these in laminates in the ‘as-laid-up’ state are shown in Figure 4-18.
The micrographs in Figure 4-18A) and B) were taken with a SEM with the prepreg sample being in an uncured ‘as-laid-up’ state. Curing in this case was not necessary due to the depth of field of the SEM. These micrographs show smaller micro-voids in the resin around individual fibre bundles in the laminate after lay-up. Although small in physical size, these micro-voids appear to be numerous and together may act as gas transport pathways if interconnected. During initial debulking, it is expected that the larger inter-laminar micro-voids may collapse, which is reflected in initial laminate compaction; the smaller intra-laminar micro-voids remain until filled in with resin in the heated curing process.

In the heated in-plane permeability test results, the ‘opening’ of the laminate test samples was described. To verify this, select samples were held at 120°C for 4 hours (full cure according to the MRCC) after ‘opening’ to cure the samples. In one case, the sample was left in the ‘open’ state during the cure. The other sample was allowed to first ‘open’, and then full vacuum was applied for the 4-hour dwell at 120°C (vacuum pulled on all in-plane permeability test set-up
compartments – as was done in the in-plane debulking test). Figure 4-19 shows a cross-sectional comparison of the two samples.

![Figure 4-19. Micrograph of Heated In-Plane Permeability Test Samples Fully Cured (4-layers). Top - Full Vacuum During 4 Hour 120°C Temperature Hold. Bottom - Vent Air Pulled Through Sample During 4 Hour 120°C Temperature Hold. Samples Separated by (Orange) Silicone Sheet.](image)

The bottom sample (left in the ‘open’ state) shows that the gas pathway ‘opening’ was in fact a physical phenomenon and not a test anomaly due to test equipment. The physical size of these paths is close to that of pin-holes (smaller than 0.5mm in diameter). The top sample (vacuum drawn after the ‘opening’ of the sample) indicates that under vacuum conditions, a combination

![Figure 4-20. Zoomed-In Portion of Figure 4-19 Micrograph (Area Indicated by Dashed Box). Pin Hole Sized Air Paths Indicated on Micrograph.](image)
of low resin viscosity and vacuum is able to fill in these pin-hole sized paths. Of practical significance, the ability for these paths to open and then close with low resin viscosity and vacuum can be beneficial with respect to off-gassing. Volatiles that may be given off during the heated cure cycle would have the ability to bubble and bleed their way out of the laminate. This process is similar to a resin bleed curing prepreg systems where entrapped gases bubble their way out of low viscosity resins. The phenomenon of off-gassing is described later in section 5.3.3.

4.4 Water Visualization

8-ply laminates with various process histories were observed in the water visualization set-up. Video was recorded and are presented here as time-elapsed storyboard images. The images show qualitatively the changes in the internal gas evacuation pathways as air bubbles leave the laminate (both free edge and surface). In these tests, an important consideration when inferring the results is the distance the air must travel to exit the laminate. Ideally, a sample of cubic geometry would provide the most unbiased test. In the laminate geometry tested, the shortest distance to travel is through-thickness, promoting through-thickness gas evacuation. Keeping this bias in mind, definitive conclusions on the preferential gas transport direction in the laminates can be made.

Selected images (A, B, C, and D) from the storyboard images for samples Lay-up #1, Lay-up #2 and Vent #4 are shown in more detail. Specific features in these images are highlighted for discussion.
Figure 4-21. Time Elapse Images of Water Visualization Experiment. Sample: Lay-up #1.
Figure 4-21 A). Sample Lay-up #1 (10 Seconds Elapsed). A Few Small Bubbles are Present on Outer Parameter.
Figure 4-21 B). Sample Lay-up #1 (1 Minute Elapsed). Fine Bubbles not Present in Previous Image Appear at the Top Right Edge of Laminate (Circled).
Figure 4-21 C). Sample Lay-up #1 (1 Minute, 40 Seconds Elapsed). Bubbles in Circled Region Continue to Grow in Size as Air Exits the Laminate.
Figure 4-21 D). Sample Lay-up #1 (2 Minutes, 40 Seconds Elapsed). More Bubbles Appear in Circled Region. Previously Present Bubbles Continue to Slowly Grow in Size.

Sample Lay-up #1 was an uncured sample in the ‘as-laid-up’ state. Very fine air bubbles (images B, C, and D) were seen exiting the laminate and forming primarily on the edge of the sample. Despite the bias for the shorter distance of through-thickness evacuation, this indicates a preference for in-plane gas transport.
Figure 4-22. Time Elapse Images of Water Visualization Experiment. Sample: Lay-up #2.
Figure 4-22 A). Sample Lay-up #2 (5 Seconds Elapsed). Numerous Fine Bubbles Appear on Outer Surface Similar to Sample Lay-up #1 (In-Plane Laminate Gas Evacuation). In Addition, an Air Bubble is Found on Front Surface of Laminate (Circled), Indicating Through-Thickness Gas Evacuation.
Figure 4-22 B). Sample Lay-up #2 (30 Seconds Elapsed). More Fine Bubbles Appear on the Outer Parameter (In-Plane Laminate Gas Evacuation). The Bubble on the Front Sample Surface from Previous Image (Through-Thickness Laminate Gas Evacuation) Migrates Further up the Sample as a New Bubble Exits the Same Location.
Figure 4-22 C). Sample Lay-up #2 (56 Seconds Elapsed). A Third Bubble is Observed Exiting from the Same Location on the Front Sample Surface (Circled). This Appears to be the Only Region Where Through-Thickness Laminate Gas Evacuation is Occurring.
Figure 4-22 D). Sample Lay-up #2 (1 Minute, 11 Seconds Elapsed). A Forth Bubble Exits from the Same Sample Surface Location (Circled).

A second ‘as-laid-up’ sample (Lay-up #2) showed similar bubble formation as sample Lay-up #1. Perhaps more distinct in this test, the bubbles form primarily on the outer edges and are very fine in size. Also visible is one individual location where through-thickness evacuation is occurring (circled in images A, B, C, and D). From this location, a few bubbles are seen leaving the sample and slowly migrating along the sample surface. These bubbles are larger in size than those forming on the laminate edge, indicating a larger gas evacuation pathway at this location than those exiting in in-plane directions.
Figure 4-23. Time Elapse Images of Water Visualization Experiment. Sample: Vent #4.
Figure 4-23 A). Sample Vent #4 (23 Seconds Elapsed). Unlike Previous Samples, no Fine Bubbles are Observed Forming on the Outer Parameter of the Sample. Air Bubbles are Seen Exiting from Only Two Locations (Circled). Bubbles are Exiting Rapidly from the Circled Area on the Left Side of the Sample.
Figure 4-23 B). Sample Vent #4 (2 Minutes, 45 Seconds Elapsed). The Bubble at the Top Right Edge of Sample Continues to Slowly Grow in Size. Meanwhile, Bubbles Continue to Exit at a Rapid Rate from the Location on Left Side.
Figure 4-23 C). Sample Vent #4 (3 Minutes, 56 Seconds Elapsed). The Bubble at the Top Right Edge Continues to Slowly Grow in Size.
Figure 4-23 D). Sample Vent #4 (6 Minutes, 18 Seconds Elapsed). The Same Bubble Located at the Top Right of Sample Continues to Further Grow in Size.

In the vacuum interrupted test sample Vent #4, vacuum was released at hour 2 of the 20 hour isothermal temperature dwell (full cure is 20 hours at 80°C). The images in Figure 4-23 show the bubbles here were much larger than the bubbles observed in the ‘as-laid-up’ samples. Gas evacuation occurring primarily at two locations (circled in images A, B, C, and D). Of these two locations, most of the air is observed to exit from the location on the left edge of the sample. The fast rate of air release makes it difficult to distinguish this location in the still single video frame images. At the top right edge of the sample, a gas bubble is seen forming and growing in size throughout the duration of the video. The nature of the bubbles observed and the limited number of locations where they form, indicate that the physical nature of the gas extraction pathways
have changed in comparison to the ‘as-laid-up’ state; where they have become larger in size, but fewer in numbers.

Figure 4-24. Time Elapse Images of Water Visualization Experiment. Sample: Vent #7.

Sample Vent #7 was a laminate where the vacuum was released during the cure at hour 16 of the 20 hour isothermal hold. However, at this stage in the cure cycle, there appears to be no gas evacuation pathways leading out of the laminate as no air bubbles were observed. At this stage in the cure cycle, resin flow has occurred to fill in these voids/paths.
Figure 4-25. Time Elapse Images of Water Visualization Experiment. Sample: Vent #8.

Sample Vent #8 represented a sample where full vacuum was maintained during the entire cure cycle process. As already seen in sample Vent #7, no gas pathways appear to be present leaving the sample. Like Vent #7, the gas extraction pathways in laminates were already filled in and removed prior to the end of the cure cycle.
5. DISCUSSION

From the testing carried out in this study, several conclusions of the processing of MTM45-1/CF2426A prepreg laminates in ambient and heated conditions can be made regarding gas flow directionality, the effect of ply-drops in gas extraction, rough evacuation times required for gas extraction, and the importance on uninhibited edge breathing.

5.1 Application and Applicability of Darcy’s Law

Permeability testing measured steady-state gas flow quantifying the breathability of MTM45-1/CF2426A prepreg laminates. Darcy’s law was used to convert measured gas flow into a laminate permeability removing any influence of sample geometry. Darcy’s law has been used previously in the literature to characterize prepreg and composite preform permeability.

Despite its use in the literature, the applicability of Darcy’s law for this study should be discussed. Darcy’s law was originally developed to characterize water flow through a fluidized bed in pipes with two distinct assumptions: 1) the fluid flow is laminar, and 2) the fluid is incompressible. However, for gas transport in porous media where the gas flow is viscous (slow), Darcy’s law is considered appropriate (Gas Transport in Porous Media, 2006). By calculating the Reynolds number (Appendix D), gas transport in this study was verified to be in the laminar flow regime. The fluid in this study was air and modified version of Darcy’s law was used that accounted for the compressibility of the air by invoking the ideal gas law.

Darcy’s law takes into account sample geometry, essentially normalizing the flow with respect to laminate cross-sectional area and length. Results from the in-plane permeability tests show good agreement with Darcy’s law. All results were relatively consistent to one another irrespective of
the number of laminate plies. The through-thickness permeability results however did not show
good agreement with Darcy’s law. Increasing the number of laminate plies dramatically reduced
the permeability of the laminate. Samples with the same number of plies showed large sample to
sample variability. It is hypothesized that gas transport in the through-thickness direction is
governed by air pathways through the resin films. The path in which gas travels is largely
influenced by how ‘holes in the resin film’ line up with respect to one another as the number of
plies in the laminate is increased. Therefore, the drop in permeability with increasing layers may
be explained by an increase in tortuosity of the gas transport paths in Equations [2] and [3].

5.2 Gas Transport Directionality

Based on the measured in-plane and through-thickness permeability, it can be concluded that for
MTM45-1/CF2426A prepreg laminates the in-plane direction is the preferred direction of gas
transport. With 100 000 times higher permeability it is still easier to transport in-plane, despite
the shorter physical distance for through-thickness gas evacuation. Physically, for gas to travel 1
mm in the through-thickness direction would be the equivalent of traveling 100m in the in-plane
direction. Practically, this means that nearly all gas extraction is achieved through laminate edge-
breathing. Therefore, great care should be taken to ensure that edges breathe freely using
specialized edge breathing dams, cork, breather, dry tow or by other similar means in the vacuum
bagging lay-out. If edge breathing pathways are inhibited, gas evacuation is greatly reduced as
through-thickness transport is effectively zero for thick real-world structural laminates (which
are more than a few plies thick).

Although quantified independently in this study, laminate gas transport occurs over multiple
directions. In some evacuation scenarios, gas transport may require a combination of both in-
plane and through-thickness directions. Examples may include: transporting gas around an embedded insert, the evacuation of gas from a sandwich panel core, or if edge breathing is partially inhibited. In these scenarios, multi-direction gas transport may occur (illustrated in Figure 5-1).

![Figure 5-1. Multi-direction gas transport for entrapped gas evacuation.](image)

As illustrated, at one point in its travel, entrapped gas traveling in the in-plane direction is required to move in the through-thickness direction. In this example, entrapped gas must move into an adjacent ply to be evacuated from the laminate. The through-thickness transport then becomes the limiting factor of entrapped gas removal.

### 5.2.1 Effect of Ply Terminations

Results from the ply termination tests showed that in-plane gas extraction is governed by the permeability of the thinnest region of the laminate. Because tapering of composite structures (through ply-drops) is inevitable in many realistic structures, this result should be considered when examining the time required for sufficient vacuum evacuation. In terms of gas extraction, a predominantly 8-ply laminate that tapers down to 2-plies should be treated as a 2-ply laminate
with extra evacuation time given to account for the reduced gas flow. Edge-breathing is discussed in more detail in section 5.6.1.

5.3 Effect of Temperature

The permeability of MTM45-1/CF2426A prepreg laminates, both in the in-plane and through-thickness directions, were found to be affected by temperature. Effects on permeability are likely attributed to the MTM45-1 matrix resin behaviour with respect to temperature as shown in Figure 5-2.

![Figure 5-2. Viscosity vs. Temperature for MTM45-1 Resin at 2°C/min. Reproduced from Advanced Composites Group MTM45-1 Data Sheet PDS1205/11.07/3](image-url)
As temperature increases, resin viscosity drops until viscosity begins to increase (chemical curing reaction intensifies). The drop in viscosity is important to allow resin to flow and fill in the laminate gas transport pathways prior to curing. The wetting out of dry fibres (originally un-impregnated) and filling in of any voids must occur prior to gelation of the resin. Otherwise, the final laminate will contain dry fibre spots and voids. The resin chemistry for MTM45-1 is tailored to allow for the gas evacuation pathways to remain open for as long as possible to maximize the capacity for gas transport (Ridgard, 2009).

5.3.1 In-Plane Permeability

As temperature increases, the in-plane permeability was found to decrease. As mentioned, this is thought to occur due to lowering resin viscosity. This trend continued to occur until the laminate sample began to ‘open up’ in gas flow at just past 100°C. As shown in Figure 5-2, this temperature approaches the lowest viscosity point of the MTM45-1 neat resin. However, it should be noted that at no point in the heating process did the in-plane gas permeability close off. Although gas flow was reduced, gas transport pathways remained available until this ‘opening’ occurrence.

The reasoning for the ‘opening’ of gas transport pathways is not known. However, this phenomenon was observed for every heated in-plane permeability test and is likely an artifact of the test set-up. It is possible that as temperature increases, the resin viscosity becomes low enough that the force of the air drawn through the sample is greater than the resin pressure, thus opening up pathways within the sample. This opening of in-plane gas pathways would likely not be observed in real processing conditions as a constant supply of 1 atm pressure air being pulled through the laminate is not typical. The only processing situation where 1 atm conditioned air
may be pulled through the laminate during the cure process is in the case of a major vacuum bag leak if hard vacuum is continually drawn. This opening of gas pathways could also have been aided by potential off-gassing of moisture entrapped in the prepreg prior or during the lay-up process. Under partial vacuum conditions, water boils at a temperature lower than 100°C. Any boiling moisture would then force its way out of the sample through the low viscosity resin. This situation is similar to air removal in a bleed cure resin systems. Where entrapped volatiles are removed as resin bleeds out from the laminate (Boyd, 2003). Due to this uncertainty, the test should only be considered valid prior to this ‘opening-up’ point.

There appears to be no correlation between the number of layers in the laminate and its effect on the temperature at which suddenly in-plane permeability pathways are created. However, there does appear to be a trend related to the number of layers in the laminate and temperature prior to this point. It is observed that as temperature increases, the rate of decreasing in-plane permeability is more pronounced with less plies in the laminate. This is particularly noticed in the temperature range beyond 70°C. Until gas pathways ‘open up’, the 2-ply sample is found to steeply decrease in in-plane permeability, whereas the 8-ply sample is found to have a more linear permeability decrease with increasing temperature. Meanwhile, the 4-ply sample lies somewhere between these two with regards to the rate of decreasing permeability with respect to temperature. Rationale for these observations can likely be attributed to the temperature gradient produced in the thickness of the sample due to the one-sided heating at the bottom of the laminate. As the number of plies in the laminate increases, so does the thermal gradient within the laminate. This was verified with the temperature recorded by the thermocouple placed on the top surface of the laminate. The temperature gradients observed are found in Appendix F.
5.3.2 Through-Thickness Permeability

As temperature increased, the through-thickness permeability increased. In both 2-ply and 4-ply samples, air flow appears steady until suddenly increasing at a critical temperature. This sudden increase in permeability is likely due to viscous flow of the resin combined with the driving force of air being pulled through the laminate in the test. Off-gassing from moisture boiling under vacuum may also aid in these pathways opening up. The critical temperature for the increased gas transport for the 1, 2, and 4-ply tested samples was averaged and plotted in Figure 5-3.

![Figure 5-3. Critical Pathway Opening Temperature for Gas Flow Through-Thickness.](image)

The 1-ply samples ‘open up’ at a lower temperature than the 2-ply samples, which open before the 4-ply samples. Through-thickness air flow begins to increase suddenly at 35°C, 49°C, and
62°C respectively. The same argument for through-thickness flow at ambient conditions, likely also applies for heated conditions. For gas flow to occur in the through-thickness direction, ‘holes’ on the resin film must line up relative to those in the adjacent layers. This becomes more difficult with thicker laminates. Creating continuous through-thickness gas paths between plies is aided by larger openings in the resin film. The formation of larger resin openings is aided by low resin viscosity, requiring higher temperature and increased time. This hypothesis is supported by the observed greater spread in critical temperature for gas flow to begin in the samples of 4-plies. Therefore, for through-thickness breathing to occur in laminates thicker than 4-plies, higher temperature or longer time is needed.

### 5.3.3 Off-Gassing

Off-gassing is considered as the release of any boiling moisture or volatile by-product from within a laminate during processing. Moisture may be absorbed by the laminate prepreg or core material (in the case of sandwich panel structures) during lay-up. At elevated temperatures during the cure process or even under vacuum, the moisture boils off as steam. Vacuum aids boiling where lower pressure lowers the boiling point of water, and in the case of full vacuum (30 inHg) this can occur at room temperature. Additionally, other chemical by-products from within the resin, including that from the condensation reaction, which is common in many epoxies, may be released during the cure process. If not evacuated from the laminate, off-gases result in voids in the cured laminate structure.

### 5.4 Permeability Compared to Other Prepreg Systems

To gain a sense of the quantified permeability measured in this study, permeability of other porous materials are listed in
Table 5-1. Here, the order of magnitude of obtained permeability for MTM45-1/CF2426A prepreg under ambient conditions is compared to the permeability of rock, and of a dry composite fibre bundle bed (non-impregnated).

Table 5-1. Permeability of Some Porous Materials.

*Resin Permeability (Kliquid < Kgas by up to 10 Times (Tanikawa & Shimamoto, 2006)

<table>
<thead>
<tr>
<th>Material</th>
<th>Permeability ( (m^2) )</th>
<th>Comments</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rock</td>
<td>( 10^{-12} ) to ( 10^{-23} )</td>
<td></td>
<td>(Tanikawa &amp; Shimamoto, 2006)</td>
</tr>
<tr>
<td>Dry Fibre bundle [parallel flow]</td>
<td>( 10^{-12} ) *</td>
<td>60 vol. %</td>
<td>(Choi et al., 1998)</td>
</tr>
<tr>
<td>MTM45-1 (5HS) [in-plane]</td>
<td>( 10^{-14} )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>MTM45-1 (5HS) [through-thickness]</td>
<td>( 10^{-19} )</td>
<td>single-ply</td>
<td></td>
</tr>
</tbody>
</table>

Depending on its type, the permeability of rock has a wide range; from granite at the low end to porous lava stone at the high end. In general, carbon fibre reinforcement (un-impregnated) has a relatively low permeability, close to that of porous rock. In the prepreg form (fibres now impregnated with resin) MTM45-1/CF2426A permeability was found to be similar to lime-stone or sandstone. In general, the permeability of composite materials is very low.

Intuitively, it is expected that the permeability of an OOA prepreg would be higher than a conventional autoclave prepreg due to the importance of removal of entrapped gases by vacuum mechanisms only. However, this was not found to be the case. In Table 5-2 and
Table 5-3, the permeability of MTM45-1/CF2426A prepreg is compared to that of carbon/epoxy prepreg Toray T800H-6K PW/3900-2 (plain weave), an autoclave prepreg system. The permeability of T800H-6K PW/3900-2 is from a manuscript in preparation by Arafath et al. (Arafath et al., 2010). The plain weave fabric laminates had a 0°/90° lay-up.

Table 5-2. In-Plane Permeability of Different Carbon/Epoxy Prepreg Material Systems.

<table>
<thead>
<tr>
<th>Material</th>
<th>Lay-up, layers</th>
<th>Permeability (m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]₂</td>
<td>2.52E-13</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]₄</td>
<td>5.05E-13</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]₆</td>
<td>3.82E-13</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]₈</td>
<td>5.49E-13</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>1</td>
<td>5.33E-14</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>2</td>
<td>5.04E-14</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>4</td>
<td>3.51E-14</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>8</td>
<td>3.18E-14</td>
</tr>
</tbody>
</table>

In the in-plane direction, T800H-6K PW/3900-2 was found to be more permeable than MTM45-1/CF2426A by nearly an order of magnitude.
Table 5-3. Through-Thickness Permeability of Different Carbon/Epoxy Prepreg Material Systems. Toray T800H-6K PW/3900-2 Data Source: (Arafath et al., 2010)

<table>
<thead>
<tr>
<th>Material</th>
<th>Lay-up, layers</th>
<th>Permeability (m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]1</td>
<td>6.69E-16</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]2</td>
<td>1.15E-15</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]4</td>
<td>2.25E-15</td>
</tr>
<tr>
<td>3900-2 (Fabric)</td>
<td>[0]8</td>
<td>3.52E-15</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>1</td>
<td>6.19E-19</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>2</td>
<td>1.65E-20</td>
</tr>
<tr>
<td>MTM45-1 (5HS Fabric)</td>
<td>4</td>
<td>0.00E+00</td>
</tr>
</tbody>
</table>

T800H-6K PW/3900-2 through-thickness permeability was found to be several orders of magnitude higher, close to the in-plane permeability of the MTM45-1/CF2426A prepreg. Unlike T800H-6K PW/3900-2, MTM45-1/CF2426A was found to decrease dramatically in through-thickness permeability as the number of plies in the laminate was increased. At 4-plies and beyond, no measurable gas flow in the through-thickness was observed.

5.5 Void Reduction Permeability Change at Ambient Conditions

Using the compaction measured in laminate Compaction Experiment #1, and using areal density, volume densities of fibre and resin and per-ply thickness from the prepreg manufacturer, the initial amount of voids after lay-up was calculated to be 21%. Subjecting the laminate to 24 hours of vacuum debulking, the laminate was observed to reduce in thickness by 7%. Assuming
this all occurred in the void region (Nam et al., 1995), 33% of the voids have been removed. The 24 hour in-plane permeability debulking test showed a reduction in permeability by 38% (Figure 4-13). Comparing the in-plane debulking test to the numerical analysis based on results from Compaction Experiment #1, it can be seen the two are similar. This suggests a relationship between laminate compaction and a reduction in laminate void volume. As suggested by Nam et al. (1995), this supports the idea of a corresponding decrease in gas permeation pathways, which the interconnect voids make up. Table 5-4 summarizes the void content calculations based on compaction data from Compaction Experiment #1. Full calculations along with the assumptions made are found in Appendix E.

<table>
<thead>
<tr>
<th>Table 5-4. Percent Void and Permeability Reduction in 8-ply MTM45-1/CF2426A Laminates Debulked for 24 hours (Compaction Test #1 and In-Plane Permeability Debulking Test).</th>
</tr>
</thead>
<tbody>
<tr>
<td>Percentage of Laminate Total</td>
</tr>
<tr>
<td>Initial void content of 8-ply laminate</td>
</tr>
<tr>
<td>Void reduction (measured and calculated in Compaction Experiment #1 – 24 hours debulk)</td>
</tr>
<tr>
<td>Permeability decrease measured (24 hours debulk)</td>
</tr>
</tbody>
</table>

Where in the laminate the reduction in voids is occurring during debulking is unknown. Due to the relatively low permeability measured, the large inter-laminar voids seen in Figure 4-17 are
thought to either be isolated (not continuous flow paths that contribute to permeability) or collapsed shortly after vacuum is drawn prior to steady-state gas flow being reached. The low permeability measured suggests that the gas flow pathways consist mostly of interconnected fibre and resin voids.

5.6 Evacuation Time at Ambient Conditions

The venting of air and volatiles from a laminate is both time and size dependent. For a larger laminate, the time required for evacuation is greater than that of a smaller laminate. This is due to the greater physical distance a bubble entrapped at the middle of the laminate has to travel for evacuation. To determine the time required, permeability can be applied as a parameter in a simple gas transport model. From this, a rough guideline determining how long a structure of a given geometry must be gas evacuated for to remove a sufficient amount of entrapped gases can be generated. This evacuation time represents the debulking stage in the composite process flow given in Figure 1-2. As it was shown that gas flow in MTM45-1/CF2426A prepreg laminates at ambient conditions was predominantly in the in-plane direction, it can be assumed gas evacuation occurs by edge breathing (in-plane only). To generate a rough guide for gas evacuation time, in-plane permeability results were applied to a previously developed 1-D gas transport model by Arafath et al. (2009). The 1-D transport model evacuation time is represented in Equation [15].

\[
time = \frac{\mu L^2}{P_o K} \left[ -\frac{1}{0.92} \ln \left( \frac{m}{m_o} \right) \right]^{0.54} [15]
\]
where the variables are defined:

\[ \mu = \text{dynamic viscosity [Pa} \cdot \text{s]} \]

\[ P_o = \text{ambient condition pressure [101,000 Pa]} \]

\[ L = \text{length of 1-D gas transport [m]} \]

\[ K = \text{in-plane gas permeability of laminate [m}^2]\]

\[ \frac{m}{m_o} = \text{mass fraction of gas remaining in laminate with respect to initial mass of gas entrapped} \]

No geometry term for thickness is found in Equation [15] as in-plane permeability (edge breathing) was found to be independent on the number of laminate plies. The only size dependent geometry variable of consideration is length L, the physical distance in which evacuating gas must travel to evacuate at the laminate edge (evacuation length).

As a case study, in-plane permeability K was applied into Equation [15] for a laminate where 1m of 1-D gas transport was required for gas evacuation. This scenario represents a 2m x 2m MTM45-1/CF2426A laminate, whereby a gas bubble entrapped at the centre of the laminate must travel 1m for in-plane edge evacuation (Figure 5-4). A rough guide for evacuation times based on 1-D gas transport for 1m was generated in Table 5-5.

\[ L = 1\text{m} \]

Figure 5-4. Edge Breathing 1-D Gas Evacuation Illustration.
Table 5-5. Time Required for Various Percentage of Gas Removal 1-D Gas Transport (1m Distance).

<table>
<thead>
<tr>
<th>% gas removed</th>
<th>(m/m₀)</th>
<th>Time Required</th>
</tr>
</thead>
<tbody>
<tr>
<td>10%</td>
<td>0.9</td>
<td>~ 1.7 min</td>
</tr>
<tr>
<td>50%</td>
<td>0.5</td>
<td>~ 55 min</td>
</tr>
<tr>
<td>80%</td>
<td>0.2</td>
<td>~ 4.4 hours</td>
</tr>
<tr>
<td>90%</td>
<td>0.1</td>
<td>~ 8.6 hours</td>
</tr>
</tbody>
</table>

In Table 5-5, (m/m₀) represents the fraction of the mass of gas remaining in the laminate with respect to the initial mass of gas entrapped within the laminate. The results show that initially 10% of entrapped gas is removed within the first few minutes of vacuum evacuation. It takes just over 55 minutes for half of the initial entrapped gas to be removed. Yet to remove 90% of the initial entrapped gas would take over 8.5 hours. Whereas 80% of the initial entrapped gas can be removed in half the time at just over 4 hours. Considerations for how long to evacuate also depend on overall processing time, out-time of the material, and the recommended debulk time by the prepreg manufacturer. For MTM45-1/CF2426A, the prepreg’s manufacturer ACG cautions against excessive debulking (ACG, 2007). As shown earlier, in-plane gas permeability was found to decrease with debulking. In addition to just MTM45-1/CF2426A, changes in permeability with evacuation time has been observed in some prepreg systems (Shim & Seferis, 1997). This model does not take into account the reduction of air evacuation pathways due to resin cold flow or pathway collapse due to the applied vacuum pressure. The reduction in gas paths would inherently increase the required evacuation times compared to those shown in Table 5-5. A more accurate model would integrate the in-plane gas permeability change with debulking.
5.6.1 Importance of Edge Breathing

In OOA prepregs, proper laminate edge breathing is important to make full use of the gas evacuation pathways engineered into the prepreg. For the prepreg in this study, manufacturer ACG recommends the use of peel-ply, or dry glass fibre tows, at regular intervals along the laminate edge to ensure open laminate edge breathing (ACG, 2007). Failing to do so may result in other vacuum bagging consumables, some of which are non-permeable, blocking the laminate free edge and effectively sealing it. Additionally, localized pressure from vacuum bagging at the laminate free edge can ‘pinch’ a laminate’s edge, reducing edge breathing. Some prepreg manufactures, like Cytec, recommend the use of porous cork, or other edge dam materials to be placed at the edge of the laminate to aid edge breathing. Additionally, Cytec stresses that the height of the edge breathing dam with respect to laminate thickness is important for optimum edge breathing (Boyd, 2003). However, this was not prescribed for the processing of MTM45-1/CF2426A, where the edge breathing and vacuum bag set-up was shown earlier in Figure 1-3.

The gas evacuation times predicted by the applied 1-D flow model (Equation [15]) assume laminate edge breathing is uninhibited. However, poor vacuum bagging practices may result in restricted gas evacuation. If edge breathing is inhibited, gas evacuation times would be much longer than those predicted in Table 5-5. The same applies to ply terminations. These two cases are similar to the illustration in Figure 5-1, where entrapped gases in laminate are required to transport through-thickness to plies with open breathing edges for evacuation from the laminate. In the extreme case of blocked edge breathing, the desired amount mass fraction of initial entrapped gas removed may never be achieved due to low through-thickness gas transport. Under ambient conditions, this was shown to be very difficult for MTM45-1/CF2426A laminates.
Chapter 5 Discussion

The importance of edge breathing was highlighted with the variations of Compaction Experiment #2. In Configuration #1, once hard vacuum was pulled, the vacuum bag was let to fold over and conform to the top half of the laminate edge effectively sealing it from gas extraction. This is similar to bagging consumable such as non-porous Teflon or FEP folding over if improper bagging is done, where no peel-ply or other porous materials is between the laminate and release layer. In Configuration #2, enough breather was stacked and butted against the laminate free edge so that the entire free end was uninhibited for gas evacuation. Both configurations followed similar compaction vs. time curves where just their magnitudes were offset. In the restricted case, the laminate was found to compact only half the amount as the unrestricted test (Figure 4-16). This suggests a substantial amount of gas remained trapped within the laminate. For a larger laminate, that the entrapped gas was unable to sufficiently migrate into the lower adjacent plies with open evacuation paths and remained in the laminate after 24 hours. This result is similar to the previously described ply terminations which reduced in-plane gas flow. Gas that cannot be evacuated in-plane is then forced to travel in the through-thickness direction to adjacent layers, which was shown to be difficult. For real composite structures this becomes a greater concern for large composite structures gas transport is time dependent. Any gas transport that must travel in the through-thickness direction which is 100 000 times greater resistance than in-plane, adds to evacuation time.

An additional consideration for the importance of edge breathing is intermittent debulking. As highlighted in Compaction Experiment #2, laminate compaction was greatly reduced from a partially blocking breathing edge. This may be of concern if intermittent debulking is done to conform and consolidate laminate plies to curves of a complex tool geometry. Insufficient edge breathing at intermittent debulk steps during lay-up may lead to reduced compaction of
individual prepreg plies to one another. This may lead to dimensional issues in the final cured structure. Therefore, ensuring all laminate edges are free for edge breathing must be done to maximize laminate compaction.
6. CONCLUSIONS

The main objective of this work was to characterize and quantify the gas permeability of laminates made from an out-of-autoclave (OOA) carbon/epoxy composite prepreg. The prepreg used in this study was MTM45-1/CF2426A, an out-of-autoclave (OOA) prepreg produced by the Advanced Composites Group (ACG). Characterization was done using qualitative visual techniques such as microscopy and water visualization and quantified by permeability. The following conclusions can be made:

1) **Gas transport is very anisotropic**

Under ambient conditions, in-plane gas permeability was greater than through-thickness permeability. Gas transport was 100 000 times lower in the through-thickness direction. In-plane permeability was found to be independent on the number of plies in the laminate. The exception to this was ply-terminations, which reduced gas flow. Gas transport in this case was governed by the fewest number of plies at any point in the laminate stack. For through-thickness direction gas transport, gas flow decreased dramatically with increasing the number of plies. By four plies, no through-thickness gas transport was detectable.

2) **The permeability of out-of-autoclave prepregs is not higher than autoclave prepregs**

The permeability of MTM45-1/CF2426A was not found to be greater than autoclave prepreg T800H-6K PW/3900-2 (plain weave). The two prepregs had similar in-plane gas permeability, while through-thickness permeability was greater for 3900-2.
3) The physical nature of the gas transport pathways changes with processing state

Debulking decreased in-plane gas permeability. Intermittent debulking (7 minutes every 4 plies) reduced in-plane permeability by about $1/3$. Extended debulking (24 hours) decreased in-plane permeability by approximately the same amount. It was observed that the majority of the reduction occurred in the initial minutes of debulk.

Temperature was found to affect gas permeability. As temperature increased, in-plane permeability decreased. However, in-plane permeability never went to zero prior to low viscosity resin flow. Although air flow was found to increase again during the heated tests, this is considered a test artifact of the 1 atm conditioned air being pulled through the laminate sample continually throughout the test. Caution should be used when interpreting this phenomenon, as this may not occur under normal processing conditions. Through-thickness permeability had the opposite behaviour. As temperature increased, through-thickness permeability increased. Through-thickness laminate gas transport, which was not present under ambient conditions, occurs at elevated temperatures.

4) Laminate compaction is related to gas evacuation

Laminate compaction was found to in part be related to gas evacuation. During debulking, laminate compaction was found to correlate to the change in in-plane permeability. Laminate compaction was also found to be sensitive to edge breathing obstructions, with gas transport being limited when edge breathing was partially blocked.
6.1 Future Work

Based on the conclusions obtained from this study, the following recommendations are made for future work:

**Streamline test matrix for future characterization of new prepreg systems**

Permeability testing as-performed was very time consuming. Based on that under ambient conditions in-plane permeability was found to be independent of the number of plies in the laminate, and that through-thickness permeability was dramatically affected by the number of plies (no gas flow in laminates 4-plies and greater – which will always be the case for realistic structures), the test matrix can be greatly reduced if these conditions apply.

**Heated cure cycle study**

A more thorough evaluation of laminate gas transport during the heated cure process should be performed. Permeability should be linked to a resin characterization rheology study. Neat resin viscosity data should be further linked to observed gas transport (permeability) behaviour. This would give better judgment whether or not changes to the cure cycle should be made. For example, determining whether using intermediate temperature dwells are needed, at what temperature, and for how long? Or whether gas extraction should primarily occur at ambient conditions (debulk)? Balancing of the two could further optimize laminate quality and reduce overall processing time.

**Effect of vacuum pressure**

The evaluation of vacuum pressure on the internal gas evacuation pathways should be examined, which was not done in this study. This should include the rate at which vacuum is applied and
whether this has consequences on laminate pinching (which reduces edge breathing). Additionally, this effect should also be studied during the heated cure process where vacuum levels may influence the ability for gas paths to remain open for as long as possible.

**Evaluation of breathing techniques and materials**

With the importance of edge breathing for OOA prepregs, an examination of various edge breathing techniques (peel-ply, dry tows, staggered edge ply-drops, etc.) and specialized breathing dams (cork, breathing dams) should be performed. Although through-thickness breathing was minimal at ambient conditions, gas transport in the thickness direction was found to occur at moderately elevated temperatures. Therefore, evaluation of through-thickness breathing consumables may also be performed. These include: tool-side breathers, perforated release layers (FEP), etc.
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*Viewpoint set no. 42 "Nanoscale materials for hydrogen storage", 56(10), 859-862.*


APPENDIX A. IN-PLANE PERMEABILITY TEST SET-UP LEAK TEST

1. Draw hard vacuum on each individual chamber and remove vacuum pump

2. Place vacuum gauges on each vacuum port. Monitor rate of vacuum drop.

3. All three compartment chambers should be able to hold full vacuum

4. Remove vacuum gauge on vent chamber (left side) and release vacuum. Monitor pressure drop in middle and right compartments

   The middle chamber should remain at full vacuum. This indicates laminate sample is sufficiently sealed, otherwise a vacuum pressure drop will be observed.

   The vacuum chamber (right side) should initially hold vacuum pressure. Rate of vacuum pressure drop should then occur slowly. If the vacuum drop was fast, this indicted that air was not only flowing through the laminate, but also by-passing the sample.

5. Reseal the in-plane permeability test set-up as necessary

   *Note – The rate of vacuum drop depends on the number of laminate plies and the permeability of the prepreg system.
APPENDIX B. IN-PLANE PERMEABILITY TEST PROCEDURE

1. Draw hard vacuum on each individual chamber. Leave vacuum connected to vacuum compartment (right side)

2. Vent the vacuum chamber (left side) and keep vented throughout test.

3. Record flow data when steady-state air flow is observed. Typically achieved in less than 5 minutes.

The time to reach steady-state air flow is done not only to achieve steady-state airflow through the laminate but also to remove any entrapped air from within the experimental set-up.
APPENDIX C. THROUGH-THICKNESS PERMEABILITY TEST

PROCEDURE

1. Vent bottom vacuum port. And keep vented throughout full duration of test. Failure to vent prior to test may result in deformation of test sample leading to erroneous test data.

2. Draw hard vacuum to top vacuum port. Leave vacuum connected to vacuum during duration of test.

3. Record flow data when steady-state air flow is observed. Typically achieved in around 1 minute once all air in system (vacuum bag set-up is removed).
APPENDIX D. CALCULATION OF REYNOLDS NUMBER

Darcy Velocity Equation [A1]

\[
\bar{u}_g = -\frac{k_g}{\mu_g} \left( \nabla P_g - \rho_g \bar{g} \right)
\]  

where the variables are defined:

\( k_g \) = gas permeability (m\(^2\))

\( \mu_g \) = dynamic viscosity of gas, air = 1.82E-5 (Pa·s)

\( \rho_g \) = density of gas, in this case air (kg/m\(^3\))

\( \nabla P_g \) = pressure gradient, \( \frac{\Delta P_g}{L} \) (Pa/m)

\( \bar{g} \) = gravitational constant, 9.8(N/kg)

Calculation of pressure gradient:

\[
\nabla P_g = \frac{1 \text{ atm}}{L} = \frac{101325 \text{ Pa}}{0.0508 \text{ m}} = 1.9946 \times 10^6 \frac{N}{m^3}
\]

Where 1 atm = 101325 Pa, \( L = 2 \text{ in} = 0.0508 \text{ m} \)

Calculation of Darcy velocity:

Substituting \( \nabla P_g \) into Equation [A1]:
Appendix D Calculation of Reynolds Number

\[ \bar{u}_g = -\frac{3 \times 10^{-14} m^2}{1.82 \times 10^{-5} Pa \cdot s} \left( 1.9946 \times 10^6 \frac{N}{m^3} - \left( 1.205 \frac{kg}{m^3} \times 9.8 \frac{N}{kg} \right) \right) = 3.288 \times 10^{-3} \frac{m}{s} \]

Where \( k_g \sim 3E-14 \ m^2, \rho_g = 1.205 \ kg/m^3 \) (@ 20°C)

The Reynolds number (Equation [A2]) can be calculated from the above calculated Darcy fluid velocity (\( \bar{u} \))

\[ \text{Re}_k = \frac{\rho_g \mu_g k_g^{1/2}}{\mu_g} \]  \hspace{1cm} \text{[A2]} \]

Where Darcy fluid velocity (\( \bar{u} \)) is related to material permeability (k), fluid dynamic viscosity \( \rho_g \) = density of gas, in this case air (kg/m³)

\( k_g \) = gas permeability (m²)

\( \mu_g \) = dynamic viscosity of gas, air = 1.82E-5 (Pa·s)

Reynolds number calculation:

Substituting in Darcy velocity \( \bar{u}_g \) into Equation [A2]:

\[ \text{Re}_k = \left( 1.205 \frac{kg}{m^3} \right) \left( 3.2878 \times 10^{-3} \frac{m}{s} \right) \left( 3 \times 10^{-14} m^2 \right)^{1/2} \left( 1.82 \times 10^{-5} Pa \cdot s \right) = 3.77 \times 10^{-5} \]

Where 1 Pa = 1 N/m² = 1 kg/m s²

3.77E-5 <<<< 1 = LAMINAR FLOW CONDITIONS
APPENDIX E. CALCULATION OF INITIAL VOID CONTENT IN LAMINATE

Calculating initial void content:

It is assumed that mass is from the fibres and resin only. Air ~ no mass

Volume of composite = fibres + resin + voids

Sample basis: 4 in x 4 in – 8ply

Thickness \( t_0 \) = 3.92mm
Appendix E Calculation of Initial Void Content in Laminate

Thickness \( t_{24} = 3.65 \text{mm} \)

Using the material properties (from manufacturer data sheet):

FAW = 375g/m\(^2\)

RW\%= 36%

density fibre = 17190 kg/m\(^3\)

density resin = 1180 kg/m\(^3\)

mass fibres \( m_f \) = FAW x Area x plies

\[
m_f = 375 \times \frac{g}{m^2} \times ([0.1016 \times 0.1016m] \times 8) = 30.96768 \text{g}
\]

mass resin \( m_r \) = \( \frac{\text{mass fibre}}{0.64} \times 0.36 \)

\[
m_r = \frac{30.96768 \times 0.36}{0.64} = 17.41932 \text{g}
\]

\( V_c = V_f + V_r + V_v \)

Rearranging for volume initial voids \( V_v \):

\( V_v = V_c - V_f - V_r \)

\[
V_v = (3.92 \times 10^{-3} \times 0.1016 \times 0.1016) - \frac{0.03096768 \text{kg}}{1790 \text{kg/m}^3} - \frac{0.01741932 \text{kg}}{1180 \text{kg/m}^3} = 8.40192 \times 10^{-6} \text{m}^3
\]
% Voids = \frac{V_v}{V_c}

% Voids = \frac{8.40192 \times 10^{-6} \text{ m}^3}{4.04644 \times 10^{-5} \text{ m}^3} \times 100 = 20.76\%

Void reduction during 24 hour debulk:

Reduction = \Delta t_{24} - t_0 = 3.65\text{mm} - 3.92\text{mm} = -0.27\text{mm}

% Reduction = \frac{-0.27\text{mm}}{3.92\text{mm}} \times 100 \cong -7\%

% Void reduction = \frac{-7\%}{21\%} \cong -33\%
ΔT between bottom and top of the laminate sample surfaces was taken at 67°C (bottom surface). This was same temperature point reference where the heating rate of the sample was verified.

<table>
<thead>
<tr>
<th>Plies</th>
<th>Heating Rate</th>
<th>Max ΔT&lt;sub&gt;(bottom-top)&lt;/sub&gt;</th>
<th>Average</th>
<th>Notes:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(°C/min)</td>
<td>(°C)</td>
<td>(°C)</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>1.98</td>
<td>3.04</td>
<td></td>
</tr>
<tr>
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<td>2</td>
<td>3.89</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>3.26</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1</td>
<td>3.53</td>
<td>3.45</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>2.62</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3A</td>
<td>4.19</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3B</td>
<td>N/A</td>
<td>3.45</td>
<td>Top TC failed</td>
</tr>
<tr>
<td>8</td>
<td>1</td>
<td>8.13</td>
<td>8.04</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
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