CHARACTERIZATION OF THE DEGREE OF IMPREGNATION OF OUT-OF-AUTOCLAVE PREPREG

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ABSTRACT

The degree of impregnation (DOI) of out-of-autoclave (OOA) prepreg is the ratio between the volume of fibres that are surrounded by resin to the volume of dry fibre regions. There is a strong incentive to develop a quick and accurate method to measure prepreg DOI that can be incorporated into existing production environments.

This paper explores three methods of characterizing the DOI of a representative aerospace grade OOA prepreg. Firstly, X-ray computed tomography is used to set a baseline level of repeatability and accuracy. Secondly, the water pick-up test is a quick technique representing one commonly used in industry. Finally, infrared thermography is used to measure average thermal diffusivity through the thickness of a specimen which is correlated to the degree of impregnation. There is a strong correlation between the DOI measured using micro CT and the measured dry fibre volume fraction from the water pick-up test. Although the correlation is less strong between micro-CT and IR thermography, the latter method is fast and non-destructive, indicating a potential for in-line prepreg inspection.

1 INTRODUCTION

In recent years, advancements in out-of-autoclave (OOA) technology have allowed for production of similar quality parts to those made using traditional autoclave processes, but at a lower cost. However, OOA prepregs are particularly sensitive to initial material state due to reduced consolidation pressure. In order to mitigate this problem, OOA prepreg is manufactured with dry fibre regions which aid in the removal of gases entrapped during layup and released during resin cure. The size of these dry regions can influence the ability to manufacture large, void-free laminates under normal processing conditions.

Previous studies have examined various aspects of the processing cycle of OOA laminates, indicating that final part quality is highly dependent on the processing cycle as well as the initial material state [1, 2]. The initial degree of impregnation of out-of-autoclave prepreg has been shown to affect the ability to manufacture void free laminates [3]. The degree of impregnation (DOI) is the ratio between the volume of fibres that are surrounded by resin to the total volume of fibre regions (Fig. 1). As such, the size of the dry fibre pathways is inversely proportional to the DOI. While a high initial DOI would result in a reduced bulk factor, making it easier to avoid wrinkling and thickness variation, the restricted gas flow paths may close prematurely, resulting in inadequate gas removal. Conversely, if the initial DOI is too low, there may be insufficient time for the resin to fully impregnate the fibre tows before gelation. As such, there is a strong incentive to develop a quick and accurate method to measure prepreg DOI that can be incorporated into existing production environments.

Degree of impregnation has been studied since the 1980s when Thorfinnson and Bierman examined the impact on final part porosity of the ratio of fibres not infiltrated by resin to the total volume of fibres in a unidirectional prepreg. The authors suggest mercury porosimetry – a procedure which involves filling the prepreg with mercury under vacuum – as a method to measure degree of impregnation [4, 5]. In the 1990s, Peltonen and Järvelä used a coloring agent to identify dry regions in glass fibre reinforced thermoplastic prepreg [6]. Putnam et al. explained the link between prepreg tack and degree of impregnation. However, other factors such as resin plasticization (due to higher volatile content) play a role in tack level, making these test less reliable [7]. One commonality between the early methods to measure prepreg impregnation level, is the intrusive nature of the tests.

Since 2000, new methods to measure degree of impregnation have emerged. X-ray computed tomography was used to visualize variability in the mesostructure of fabric and prepregs [8] and Centea and Hubert used the technique to measure impregnation of fibre tows for woven fabric prepreg [9].
Finally, Fetfadsidis and Bhardwaj have introduced a method of non-contact ultrasound to measure dry regions within unidirectional prepreg [10].

![Diagram of degree of impregnation for a unidirectional tape (left) and fibre tow (right). Non impregnated regions are highlighted with dashed red lines.](image)

Figure 1: Degree of impregnation for a unidirectional tape (left) and fibre tow (right). Non impregnated regions are highlighted with dashed red lines.

In this paper, three methods will be compared to measure the level of impregnation of out of autoclave prepreg. First, X-ray computed tomography is used to obtain a baseline measurement. Secondly, the water pick-up test was used. This test requires less time than X-ray computed tomography and is thus more widely incorporated in industry. Finally, active infrared thermography is used as a non-destructive and non-intrusive method.

2 MATERIALS AND METHODS

2.1 Materials and Preparation

All specimens used in this study are made of a representative OOA aerospace prepreg, a modified version of CYCOM 5320 manufactured by Cytec Engineered Materials, with an 8-harness satin fabric architecture [11]. Squares samples with dimensions of approximately 200 mm by 200 mm are cut from the same roll and prepreg out-time is monitored to assure it did not surpass its useful life.

Interrupted vacuum holds at elevated temperatures are used to increase degree of impregnation in order to study specimens of various initial conditions. Four levels of impregnation were studied: initial, low (following a 16 h room temperature vacuum hold), medium (after 36 minutes at elevated temperature), and high (after 56 minutes at elevated temperature). The specimens heated to elevated temperature were heated at a rate of 1.21 °C/min in a convection oven from room temperature and immediately quenched in a freezer after the proper amount of time. The medium DOI specimen reached a maximum temperature of approximately 55 °C and the high DOI specimen reached a maximum temperature of approximately 80 °C. This procedure is shown schematically in Fig. 2.

![Diagram of processing cycle used to increase prepreg degree of impregnation. Times \(t_1\) and \(t_2\) represent 36 minutes and 56 minutes at elevated temperature, respectively.](image)

Figure 2: Schematic of processing cycle used to increase prepreg degree of impregnation. Times \(t_1\) and \(t_2\) represent 36 minutes and 56 minutes at elevated temperature, respectively.
2.2 X-ray Computed Tomography

A bench top X-ray microtomograph, Skyscan 1172, manufactured by Skyscan is used in this study. The X-ray source is operated with 64 kV and 157 μA and an exposure time of 1178 ms. The camera size is 4000 pixels wide by 2096 pixels high and the pixel size is 7 μm. Once scanned, the samples are reconstructed using Skyscan’s NRecon software and rotated for straightness using Skyscan’s DataViewer software. Finally, an in-house written MATLAB script is used to analyze the data.

Based on the resolution and pixel size, sections of approximately 13.5 mm by 13.5 mm are cut from the center of each specimen to remove edge effects. Sections are sandwiched between low density polystyrene foam and multiple specimens are scanned at once.

2.3 Water Pick-up Test

The water pick-up test is a much simpler test than micro-CT. It does not require expensive equipment and has very little specimen preparation and data analysis time. This test involves measurement of the change in mass of the prepreg after it is immersed in water. Similar to a paintbrush dipped in paint, when the prepreg is dipped in water, water fills the dry fibre regions due to capillary motion. Thus, the mass change is related to the prepreg DOI.

A simplified version of the test suggested by Hexcel Holding GmbH is used [12]. Sections of approximately 50 mm by 50 mm are cut from each specimen near the center. The specimens are weighed using a high precision scale, loosely gripped at one end, and the opposite end is immersed in room temperature distilled water. After 20 minutes the specimen is removed from the water and any excess water on the surface is removed. The samples are again weighed, and the quantity of water uptake can be measured from the change in specimen mass.

2.4 Active Infrared Thermography

Infrared thermography is a growing field in non-destructive testing of composites due not only to improved techniques but also increased availability and reduced cost of thermal cameras; it has been used to detect damage and predict porosity values of composite laminates [13].

There are different classification of thermographic approaches: passive, where the specimen is self-heated, and active, where an external excitation source is required. Additionally, there are two main configurations, reflection and transmission. In reflection mode, the heat source and camera are on the same side of the specimen which is better for defects near the surface. In transmission mode, the heat source and camera are on opposite sides of the specimen which is better for defects through the thickness. For these tests, an active, transmission method is used.

A Phoenix Indigo thermal camera is used to measure the spatial and temporal temperature distribution of the specimen. This camera has a 640 by 512 pixel spatial resolution and a temperature accuracy of 25 mK. The camera’s framerate is maximized based on the computer setup and is typically between 55 and 60 Hz. The specimen is excited with two Balcar FX 60 flash lamps with a pulse duration of 5 ms and which produce approximately 6.4 kJ of energy, respectively, pointed at the center of the specimen. The 200 mm by 200 mm specimens are placed in a wooden frame and only the center of each specimen is used in the analysis to avoid edge effects. The setup is shown in Fig. 3. Although this experiment is described last, chronologically, the tests were performed prior to the micro-CT and water pick-up experiments.
3 ANALYSIS AND DISCUSSION

3.1 X-ray Computed Tomography

Once the specimens sections are scanned and reconstructed, the in-house written MATLAB script is used to analyze the data and obtain degree of impregnation values. The script follows a similar procedure to the one outlined by Centea and Hubert in order to measure prepreg DOI based on dry tow area [9]. Carbon fibre and epoxy have similar densities; thus, it is difficult to distinguish fibres and resin at the given resolution. However, there is a good contrast between the carbon fibres / epoxy and the entrapped air (which appears much darker), making it possible to measure dry fibre area.

Starting from the cross sectional images (Fig. 4 a)), individual tows are selected (b) and an ellipse is drawn to roughly outline the tow (c). The area within the ellipse is isolated (d) and using greyscale thresholding the image is converted to black and white (e). Otsu’s method is used in order to determine a greyscale threshold value that minimizes intraclass variance [14]. Finally, the region of the tow is isolated by filtering, smoothing, and selection based on pixel area (f). This method has a good ability to select and measure dry tow areas as is shown in (g).

![Outline of procedure used to measure dry fibre regions.](image)

**Figure 4:** Outline of procedure used to measure dry fibre regions.

Fig. 5 shows the results of this analysis for a specimen of each of the four conditioned DOI as well as the average calculated DOI values found using Eq. 1 (described below). The method of increasing impregnation caused a decrease in prepreg thickness as well as a reduction of large inter tow macropores. Moreover, tow impregnation is not constant for all tows of a given prepreg condition. For example, the tow located at the cross over point of the 0° and 90° yarns generally has the largest dry area and is the last to fully impregnate. This variation in tow impregnation can be observed by the size of the error bars in the graph in Fig. 5, which represent one standard deviation. As the DOI is increased, the tows do not impregnate uniformly, thus the variability is increased.
Figure 5: Micro-CT analysis of dry tow area for specimens pre-conditioned to four levels of impregnation and the average degree of impregnation for the given condition.

The degree of impregnation is calculated from the dry fibre areas which are found from the procedure outlined above using Eq. 1,

\[ DOI = 1 - \frac{A_{\text{dry}}}{\sqrt{A_{\text{tow,av}}}} \]  

Where \( A_{\text{dry}} \) is the measured dry tow area and \( A_{\text{tow,av}} \) is the average measured dry tow area for the initial, unprocessed prepreg specimens. A degree of impregnation value is found for each tow and an average value is taken for the specimen. For cases when the measured dry area is less than the average initial dry area (this occurred for the unprocessed specimens), the DOI is set to 0. This procedure is done for 20 tows per specimen.

Micro-CT is used to determine a baseline level of accuracy. Nevertheless, there are still issues of operator induced error due to the manual post-processing procedure, such as tow selection and greyscale thresholding. While automatic methods of greyscale thresholding can mitigate this drop in reproducibility, it is difficult to fully automate the procedure when dealing with woven fabrics. The main drawback of the method is time and cost. Preparing the specimens, running the scans, and reconstructing and analyzing the results can take upwards of 6 hours for a single specimen, making it unsuitable to a time driven production schedule.

3.2 Water Pick-up Test

As the specimens’ degree of impregnation is increased, the mass of water that fills the prepreg decreases. Dry fibre volume fraction of the prepreg can be estimated using Eq. 2,

\[ \text{Dry fibre volume fraction} = \frac{V_{\text{uptake}}}{V_{\text{prepreg}}} = \frac{\Delta m}{\rho_{\text{water}} V_{\text{prepreg}}} \]  

Where \( V_{\text{uptake}} \) is the volume of water drawn into the dry regions within the prepreg, \( \Delta m \) is the change in mass of the prepreg before and after the test, \( \rho_{\text{water}} \) is the density of water, and \( V_{\text{prepreg}} \) is the volume of prepreg, found from the dimensions of the specimen and average thickness. In Fig. 6, the dry fibre volume fraction based on water uptake is plotted with the degree of impregnation found using micro-CT. Using a simple exponential curve, it is possible to obtain a very good fit through the data with a coefficient of determination of 0.99.
Intuitively, one would expect the relationship observed in Fig. 6 to be linear – the water pick up test measures the mass of water filling the dry tows and micro-CT measures the size of dry fibre tows. However, in the previous section it was explained that the method used to increase impregnation made alterations to the prepreg state; that is, a reduction in macro porosity was observed. These macro pores, which are not accounted for in the method of measuring tow impregnation could act as miniature water reservoirs. Moreover, small cavities bounded by a thin layer of resin film are located beside cross overs of perpendicular fibre tows. These cavities (shown in Fig. 7 a)) are a product of the hot melt prepreg manufacturing process. During this process carbon fabric is impregnated from both sides with resin film using heated compaction rollers and small films of resins can remain stuck to the backing film. It is clear from (Fig. 7 b)) that there is resin pooling in these regions as well.

Figure 6: Variation of dry fibre volume content with degree of impregnation of OOA prepreg measured by micro-CT.

Despite the drawbacks of this method, a good correlation is observed between water pick-up values and DOI measured using micro-CT analysis. Unfortunately, the method is destructive and although it may not be difficult to test scrap from a ply cutter, it only gives very local information and may not highlight problematic areas of the prepreg.

3.3 Active Infrared Thermography

Using MATLAB, the data recorded from the infrared camera is first converted to an integer matrix, then to temperature values based on calibration data of the infrared camera, and faulty pixels are
removed by replacing them with an average of the 8 surrounding pixels. Thus, the thermal history of the specimen is obtained. Next, following the method of linear diffusivity fitting (LDF) proposed by Hendorfer et al., the temperature history of each pixel is used to predict the average thermal diffusivity of the prepreg at that location [15]. Thus, the series of thermal response images is converted to a material quantity image. Thermal diffusivity is a ratio of a material’s ability to transfer energy to its ability to store energy,

\[
\alpha = \frac{k}{\rho c_p}
\]  

(3)

Where \( k \) is thermal conductivity, \( \rho \) is density, and \( c_p \) is the specific heat capacity. The presence of dry regions within the prepreg will change the way heat flows through the specimen and the overall thermal properties of the prepreg will be affected by the fibres, resin, and entrapped air. Fig. 8 a) shows the thermal response of a single pixel of the specimen. The average pixel temperature prior to the flash is calculated and subtracted from all values subsequent to the flash. The temperature of the back surface gradually increases as heat transfer through the specimen before it reaches a maximum and slowly decreases. Prior to reaching the maximum temperature change, the process is dominated by diffusion of heat through the specimen. After the maximum temperature, the convective cooling of the specimen dominates. As a result, the data prior to reaching maximum temperature is analyzed.

For the given flash lamp, the pulse duration is quite short, thus, it is assumed that the temperature on the back side of the laminate is not affected by the heat pulse during the duration of the pulse. As such, the prepreg is modelled as a semi-infinite solid. Starting from the one dimensional heat equation,

\[
\frac{1}{\alpha} \frac{dT}{dt} = \frac{d^2T}{dx^2}
\]

(4)

Where \( T \) is temperature, \( t \) is time, and \( x \) is the one spatial dimension. Assuming that radiation and convection heat losses are negligible, the one-dimensional temperature history can be found,

\[
T(x, t) = T_0 + \frac{Q}{\rho c_p \sqrt{\pi \alpha t}} \exp \left( -\frac{x^2}{4\alpha t} \right)
\]

(5)

Where \( Q \) is the quantity of heat absorbed by the prepreg. This equation can be linearized and solved for back surface temperature,

\[
\ln(T(L, t) - T_0) + \frac{1}{2} \ln(t) = -\frac{L^2}{4\alpha} + \ln \left( \frac{Q}{\rho c_p \sqrt{\pi \alpha}} \right)
\]

(6)

Finally, plotting \( \ln(T(L, t) - T_0) + 1/2\ln(t) \) against \( 1/t \), thermal diffusivity can be found from the slope (Fig. 8 b)). The average specimen thickness, \( L \), is found from the micro-CT images.

This method allows calculation of an average thermal diffusivity through the thickness of the specimen and does not require measurement of the quantity of heat absorbed by the prepreg.
Thermal diffusivity of the prepreg that contains higher thermal diffusivity. The average values are indicated.

The average thermal diffusivity is plotted against the measured degree of impregnation in Fig. 9. It shows the thermal diffusivity maps for specimens of each level of impregnation. The images have been filtered using a 3 × 3 median filter and the average values are indicated.

Figure 9: Thermal diffusivity maps for specimens of various levels of impregnation.

There are a few trends to notice from this data. First, as the degree of impregnation increases, the thermal diffusivity increases as well. It is known that the thermal properties of porous carbon fibre reinforced laminates follow an adjusted rule of mixtures based on fibre volume content and porosity. With the presence of air decreasing the thermal conductivity as well as the density of the laminate [16]. Moreover, Mayr et al. explain that the thermal diffusivity of porous carbon fibre reinforced laminates is also affected by pore size and morphology [17]. It seems intuitive that this explanation can be used for uncured prepreg as well. The thermal diffusivity of the prepreg that contains more entrapped air (lower impregnation) will be less than the prepreg with less entrapped air (higher impregnation). Secondly, a pattern characteristic of the carbon fibre weave is visible in the images. This pattern is likely caused not only by the fabric architecture, but by the air within the dry tows, causing local variations in diffusivity.

The average thermal diffusivity is plotted against the measured degree of impregnation in Fig. 10. As with the water pick-up test, an exponential curve is fit through the data with a coefficient of determination of 0.93. The correlation between this method and micro-CT is less than the water pick-up test. However, there is less spread in the data at lower degree of impregnation values, which are more likely to be seen in practice.
Figure 10: Variation of prepreg thermal diffusivity with degree of impregnation of OOA prepreg.

3.4 Comparison of Methods

Table 1 compares the three aforementioned methods based on different considerations. Micro-CT is the longest test to perform; it requires significant amount of time scanning, reconstructing, and processing data. Water pick-up and IR thermography are both very quick tests which require less than an hour to capture and process data. Water pick-up has, by far, the lowest cost as it requires little to no equipment. Contrarily, micro-CT and IR thermography both require expensive equipment and post-processing tools. The correlation between water pick-up and micro-CT is better than IR thermography. The only method that is both non-destructive and has a potential for in-line application is IR thermography. This method essentially requires finding the slope of a temperature vs. time plot and it is conceivable that a single static camera could be replaced by two cameras and the technique could be applied to a moving production line. Thus, despite the drawbacks and uncertainties involved with this method, there is clear potential for use as a quick, in-situ, non-intrusive prepreg inspection technique.

![Graph showing variation of prepreg thermal diffusivity with degree of impregnation measured by micro-CT.](image)

\[
y = 2.06E-07e^{1.85x} \\
R^2 = 0.93
\]

Table 1: Comparison of three methods.

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<th>Micro-CT</th>
<th>Water pick-up</th>
<th>IR thermography</th>
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4 CONCLUSION

Three techniques were used to measure the degree of impregnation of a carbon fibre reinforced epoxy prepreg with an 8HS fibre architecture: X-ray computed tomography, water pick-up, and active infrared thermography.

X-ray computed tomography serves as a baseline level of accuracy and repeatability. The test is time consuming and includes uncertainties due to manual post-processing techniques and greyscale thresholding which can be partly mitigated by using automatic procedures. The water pick-up test is much quicker and the results correlate well with micro-CT. However, both of these methods are destructive and only give local values of the area tested. Active infrared thermography is used to calculate an average thermal diffusivity through the thickness of the specimen. While the correlation of this method is less than for the water pick-up test, this test is very quick and non-destructive and has the potential for an in-line prepreg inspection application.
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