Axial Impregnation of a Fiber Bundle.

Part 1: Capillary Experiments

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This is an experimental study to investigate the axial capillary impregnation of an E-glass fiber bundle by using two Newtonian infiltrating fluids: (a) silicone oil and (b) epoxy. First, the wetting properties of the systems were measured, i.e. the surface tension of each fluid and the contact angle between the fluid and a single fiber filament. Liquid height and weight data as a function of time were collected from several capillary impregnation experiments of single fiber bundles. A data analysis is presented following either the integral or the differential approach to determine capillary pressure, permeability and porosity. The so-determined values of capillary pressure were compared to corresponding theoretical values and were generally found much lower, unless the duration of the capillary experiment was around a month. This long-term experiment brought substantial improvement in the determined values of capillary pressure. The infiltration plots from the experimental data will be further used for validation of theoretical impregnation curves, which will result in the theoretical prediction of permeability and capillary pressure in Part 2 of this study (1).

1. INTRODUCTION

The axial impregnation of a fiber bundle is a fundamental phenomenon taking place in a variety of processes in the chemical, textiles and composites manufacturing industries. Given that many types of fabrics are made from fiber bundles, in-plane impregnation of a fabric is expected to contain a component of axial impregnation of fiber bundles. This flow component may be important depending on the processing conditions and the fabric architecture: for example it is dominant in the capillary in-plane impregnation of a woven fabric (2); on the other hand, in the in-plane infiltration of an assembly of woven fabrics, macro-flow between fiber bundles (3) is usually the important flow mode in resin transfer moulding (RTM) or structural reaction injection moulding (SRIM).

Whatever the dominant flow mode in the impregnation of fabric assemblies, it becomes increasingly popular to try to predict the infiltration characteristics of these assemblies by using known models and values of empirical constants for the basic components or units of such assemblies. In this case, it has been considered (2) that the fiber bundle is the basic unit the impregnation of which is modelled by following Darcy's law (4). The required parameters for such modeling (3) are the permeability and capillary pressure of the fiber bundle. An experimental study on the axial impregnation of a glass fiber bundle by two model Newtonian fluids, silicone oil and epoxy, is included in Part 1 of the present paper. Part 2 (1) contains the suggested models for the determination of the permeability of the fiber bundle and the capillary pressure, and their validation against the experimental data of Part 1. Permeability controls the infiltration of fibrous media in many manufacturing processes (5) in the composite materials and other industries. Capillary pressure plays an important role in the void formation (6–12) in the composites manufacturing and may not be negligible in the infiltration of high fiber volume fraction fiber assemblies at low injection pressures.

A number of past studies have considered the capillary axial impregnation of fiber bundles or aligned fiber beds. In many of these studies the fibers or fiber bundles were encased in a glass tube (13, 14) in order to be able to estimate the cross-sectional area of the fiber bed as that of the tube. The fiber-filled tube was suspended above a beaker containing the infiltrating liquid, with the lower end of the tube just below the liquid surface in the beaker. The rate of axial impregnation of the fiber bed under the effect of capillary pressure was determined by measuring the weight increase of the fiber-filled tube. Carleton and Nelson...
(13) reported that the fibers in the bundle were further compacted by the surface tension of the liquid being absorbed and the rate of impregnation could be increased by holding the fibers further apart mechanically. Generally, a pore distribution is expected for the fiber bed. Bayramli and Powell (15, 16) encased the aligned fibers between the lengths of two tapes. They concluded that higher rates of impregnation were observed for samples with larger pore heterogeneity than those with a narrower distribution of pore sizes.

According to the Young-Laplace equation (17), the capillary pressure depends on the wetting properties of single fibers and liquids, which incorporate the surface tension of liquid and the contact angle at the solid/liquid interface. Hsieh et al. (18) and Hsieh and Yu (19) carried out dynamic measurements of liquid wetting and retention characteristics in the interaction between a suspended fiber filament or woven fabric and a liquid. They concluded that the wetting characteristics of a fabric are the same as those of its constituent single fiber filaments.

The purpose of this study is to carry out axial, capillary impregnation experiments for a fiber bundle using either silicone oil or epoxy as model Newtonian fluids usually employed in permeability experiments. The fiber bed was not placed in any tube, in the current investigation. The data will be analyzed with the aim to be used in the validation of the theoretical modeling to predict permeability and capillary pressure in Part 2, as functions of the fiber characteristics and the fiber volume fraction (1). Measurements of the wetting properties, surface tension and contact angle, are also included in this study.

2. MATERIALS

Capillary impregnation experiments were carried out using single fiber bundles removed from a plain-woven, E-glass-fiber fabric, Y0212, supplied by Fothergill Engineered Fabrics. The goal is to determine the infiltration properties of a single bundle of this fabric to use them as input parameters of the modeling of either the capillary impregnation of the fabric (2) or the impregnation of assemblies of this fabric in RTM (3). The fabric has an areal density of 0.546 kg/m² and a nominal thickness of 0.48 mm. Both warp and weft bundles are of 136 × 3 EC9 tex, with 2090 filaments in a bundle. A methacrylato chromic chloride (Volan) finish was applied to the fabric after weaving to enhance fiber compatibility/adhesion to polyester, epoxide and vinylester resin systems and aid handling. The diameter of the final coated filament was determined as 10.5 μm by optical microscopy and dynamic contact angle analysis (see section 3.1).

The fluids used in the capillary impregnation experiments were silicone oil and epoxy resin. The silicone oil was supplied by Dow Corning®. The hot-curing epoxy matrix system consisted of Araldite® LY 564, a bisphenol A epoxy resin, and hardener HY 2954, a cycloaliphatic amine hardener, supplied by CIBA polymers. Most impregnation experiments were carried out with the epoxy resin without the hardener so that no curing would occur during impregnation to prevent viscosity changes. When a composite sample was to be prepared for subsequent microstructural studies (see section 3.3) the curing epoxy system, including the hardener, was used. In the latter case, the mix ratio was 100:35 [Araldite®/hardener] parts per weight.

The viscosity of the infiltrating liquid was measured by using a Brookfield viscometer at the beginning of each impregnation experiment and at regular intervals during the experiment, for long term experiments. Both silicone oil and epoxy (non-curing) proved to be Newtonian fluids for frequencies of rotation in the range of 0.5 to 50 rpm. Table 1 presents the properties of the infiltrating liquids where the methodologies for the measurement of surface tension and contact angle are outlined in section 3.1. In general, a low contact angle implies good wetting between the fiber and the liquid and a low surface tension for a resin is expected to improve void elimination during impregnation in composites manufacturing (20).

3. EXPERIMENTAL PROCEDURES

3.1 Measurement of Wetting Properties

A DuNoiy ring, KRUS digital tensiometer—K10ST—was used in this study to measure the surface tension of liquids. This has a ring of known radius R, made from wire of radius r, (R ≫ r.). The plane of the ring is horizontally aligned to within ± 1° relative to the surface of the liquid, so that the ring is initially wetted by the liquid. The DuNoiy ring tensiometer (21) measures the maximum force that surface tension can exert on a wire ring, as it is withdrawn upwards from the surface of a liquid.

The contact angle between the fiber and the infiltrating liquid was determined by the single fiber pull-out test based on the Wilhelmy principle (14, 22–24) using a dynamic contact angle analyzer, DCA-322 CAHN. In this, a single fiber filament is suspended from a balance and a beaker containing the wetting

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Density (kg/m³)</th>
<th>Viscosity (mPa s)</th>
<th>Surface tension (N/m)</th>
<th>Contact angle with E-glass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicone oil</td>
<td>850–855</td>
<td>105–125</td>
<td>23 × 10⁻³</td>
<td>21°</td>
</tr>
<tr>
<td>Epoxy resin (non-curing)</td>
<td>1130–1133</td>
<td>1600–2200</td>
<td>44 × 10⁻³</td>
<td>57°</td>
</tr>
</tbody>
</table>
fluid is slowly (50 μm/s) moved upwards via a movable platform, which will eventually make the fiber contact the fluid surface; this experimental procedure produces a measurement of the advancing contact angle. Although the receding contact angle could also be obtained, only the advancing contact angle data were used in this study since the capillary impregnation experiments involved primarily advancing flow.

When contact between the test fluid and the fiber occurs, the wetting force exerted on the fiber, \( F_w \), is recorded. The contact angle, \( \theta \), is then given by

\[
\cos \theta = \frac{F_w}{\sigma \phi_f}
\]

where \( \sigma \) is the surface tension at the liquid/vapor interface for the infiltrating fluid and \( \phi_f \) is the perimeter of the fiber filament. The latter was determined by using a standard fluid (hexadecane), with known surface tension and low contact angle (\( \theta = 0^\circ \)). From this an average filament diameter of 10.5 μm was determined for the employed fiber bundles.

3.2 Axial Capillary Impregnation Experiments for a Single Fiber Bundle

Figure 1 shows the experimental apparatus used to follow the axial capillary impregnation of a fiber bundle. A fiber bundle is suspended from an electrobalance with its lower end immersed in the infiltrating liquid, which is placed in a beaker. The liquid uptake during the impregnation of the bundle may be recorded in two ways: (a) rise of fluid height in the bundle, which was monitored using a traveling microscope and also with the help of a ruler displayed in Fig. 1; (b) weight increase of the fiber bundle being impregnated with the liquid. Weight changes were followed with an accuracy of 0.1 mg. The experiments were conducted at room temperature.

The fiber bundle was carefully removed from the original fabric and cut to the desired length; one extreme was attached with adhesive tape to the fiber support, whereas the other end was attached to a small weight, in an attempt to keep the fiber bundle...
The determination of the bundle cross-sectional area were avoided here in order to prevent edge effects. The bundle was formed and, at that time, the current values for height and weight were considered the initial values and the stopwatch was started. This experimental procedure has an additional advantage of reducing, or eliminating, the influence of the wetting forces, present in the early stages of the impregnation process.

Although glass capillaries to encase the fibers (13, 14, 22) and taping of the fiber bundles (15, 16) have been used in past studies, these techniques were avoided here in order to prevent edge effects. On the other hand, extra care had to be paid to avoid fraying of the fiber bundle, which would change the flow characteristics, and the cross-sectional area of the bundle became an extra variable to be measured. The determination of the bundle cross-sectional area and the pore fraction from the available experimental data of liquid height and weight increase is described in section 4.

The long-term epoxy resin capillary experiments produced only height readings. Weight readings were not obtained since such a length of bundle (> 0.2 m) could not be fitted inside the balance chamber and, also, the balance could not be allocated for the experiment for such a long period of time. In order to reduce the swelling of the fiber bundle being infiltrated by the epoxy for a long time, the lower end of the bundle was taped to the bottom of the beaker.

### 3.3 Microstructural Analysis of Fiber Bundles and Burn-off Tests

Since only height data of the liquid uptake were available for the long-term impregnation experiment using the epoxy resin (see section 3.2), it was not possible to estimate the bundle cross-section and pore fraction according to the method described in section 4 for this experiment. Hence, it was decided to carry out microscopy analysis on independent bundles infiltrated by a mixture of epoxy resin and hardener. The same setup as that described in section 3.2 was used, again with the lower end of the bundle taped to the bottom of the beaker as in the long-run experiment. The resin managed to rise to a height of ~20 mm above the surface of the liquid in the beaker before hardening.

The thus-obtained epoxy/fiber bundle composites were cross-sectioned, polished, examined under an Axioscop optical microscope and image-analyzed using UTHSCSA ImageTool software v. 1.27. The procedure involves a color to gray scale transformation, finding objects in the image via thresholding (used to create a binary image), spatial calibration and object analysis. The attributes of objects determined in this work were: (a) Area: measured as the number of pixels in the polygon; (b) perimeter: the length of the outside boundary of the object; (c) Feret diameter: the diameter of a circle having the same area as the object; (d) major axis length: the length of the longest line that can be drawn through the object; and (e) minor axis length: the length of the shortest line that can be drawn though the object perpendicular to the major axis. From the total area of the bundle, $A_B$, the pore area of the bundle cross-section, $A_{br}$, and the porosity of the fiber, $\varepsilon$, are evaluated as follows:

$$A_{br} = A_B - \pi N R_f^2$$  \hspace{1cm} (2)

$$\varepsilon = A_{br}/A_B$$  \hspace{1cm} (3)

where $N$ is the number of filaments in the fiber bundle and $R_f$ is the filament radius.

Two examples of micrographs used for the image analysis are shown in Fig. 2, where the cross-sections of two individual fiber bundles can be seen, and the results of the image analysis are summarized in Table 2. In this table, it can be seen that both samples give very similar results for the total area of the bundle. The measurements of the major and minor axes are just for illustrative purposes and suggest a difference in the shape of the bundle, which, however, has not altered the porosity ($\approx 0.47$). Considering the excellent agreement between these values, a porosity of 0.47 for the bundle was attributed to experiments using the procedure of attaching the lower end of the bundle to the bottom of the beaker, which restricts the swelling of the bundle by the infiltrating liquid.

Burn-off tests were carried out for 36 independent cured epoxy/fiber bundles to confirm the results obtained via image analysis. An empty ceramic crucible was preheated to 600°C and weighed after cooling to room temperature. The sample was then placed into the crucible followed by a new weighing of the system (crucible + sample) prior to its placement in a furnace, preheated to 600°C, for one and a half hour. After that, the resin is expected to be totally burned off, while the glass-fibers are expected not to have been degraded. Then the crucible was removed from the furnace and allowed to cool to room temperature prior to a new weighing of the system. An average porosity value for the fiber bundles can be calculated from the following relation (25)

$$\varepsilon_y = 1 - \left( \frac{W_C - W_B}{\rho_{glass}} \right) \left( \frac{W_A - W_C}{\rho_m} + \left( \frac{W_C - W_B}{\rho_{glass}} \right) \right)$$  \hspace{1cm} (4)

where $W_A$ is the mass of the crucible and the sample (pre-burn-off), $W_B$ is the mass of the crucible, $W_C$ is the mass of the crucible and the glass-fibers (post-burn-off), $\rho_{glass}$ is the density of E-glass and $\rho_m$ is the density of the epoxy matrix. The average porosity within the bundle from the 36 experiments was found to be $\varepsilon_y = 0.45$, close to the results from the microscopy analysis.
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Fig. 2. Micrographs of two single cured epoxy/fiber bundles cross-sections (size bars represent 100 μm).

Table 2. Results of the Image Analysis of the Photographs of the Cross-Sections of Cured Epoxy/Single Fiber Bundles (Figure 2) Obtained From the Optical Microscope.

<table>
<thead>
<tr>
<th>Photograph</th>
<th>Total major axis length</th>
<th>Minor axis length</th>
<th>Total bundle area ( \times 10^3 ) (m²)</th>
<th>Perimeter ( \times 10^3 ) (m)</th>
<th>( A_{TV} \times 10^3 ) (m²)</th>
<th>( \varepsilon )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>3.450</td>
<td>1.500</td>
<td>3.235</td>
<td>1.150</td>
<td>0.424</td>
<td>1.636</td>
</tr>
<tr>
<td>(b)</td>
<td>3.388</td>
<td>0.899</td>
<td>2.692</td>
<td>0.494</td>
<td>0.46</td>
<td>1.574</td>
</tr>
<tr>
<td>Average</td>
<td>3.42</td>
<td>1.02</td>
<td>2.96</td>
<td>0.46</td>
<td>1.60</td>
<td>0.47</td>
</tr>
</tbody>
</table>

4. ANALYSIS OF THE EXPERIMENTAL DATA OF THE AXIAL CAPILLARY IMPREGNATION OF A FIBER BUNDLE

When a capillary channel is brought into contact with a liquid surface in a container, a driving pressure difference, \( \Delta P \), is generated because of capillary action and is responsible for the liquid rising above the surface of the liquid in the container while acting against the weight of the column of liquid.

\[
\Delta P = P_c - \rho gh
\]  

where \( P_c \) is the capillary pressure, \( \rho \) is the liquid density, \( g \) is the gravitational acceleration and \( h \) is the height of liquid rise.

When an equilibrium height, \( h_e \), is reached, the liquid stops rising (\( \Delta P = 0 \)), and

\[
P_c = \rho gh_e
\]  

A similar approach described by Batch et al. (14) for analyzing data for a rising liquid in the capillary impregnation of a porous medium has been followed in this study. The fiber bundle is considered to contain an assembly of tortuous capillary channels, where by applying Darcy's law the velocity of the rising front of the liquid, \( dh/dt \), can be expressed as a function of the driving pressure difference, \( \Delta P \), given by Eq 5, as follows:

\[
\frac{dh}{dt} = \frac{\kappa}{\mu e} \left( \frac{P_c - \rho gh}{h} \right) \quad (7)
\]

Rearranging Eq 7 yields

\[
\frac{dh}{dt} = \frac{\kappa P_c}{\mu e h} - \frac{\kappa \rho g}{\mu e} = \frac{\alpha}{h} - \frac{b}{h} \quad (8)
\]

By using Eq 8, a differential approach of analyzing the fluid height increase data is presented, where by plotting \( dh/dt \) against \( 1/h \) a straight line should fit the data with:
The capillary pressure, \( P_c \), and the permeability, \( \kappa \), can then be estimated by the relations

\[
P_c = \rho g \left( \frac{a_h}{b_h} \right) \quad \text{(10)}
\]

\[
\kappa = \frac{\mu e}{P_c a_n} \quad \text{(11)}
\]

For short times, the second term on the right-hand side of Eq 8 can be neglected and the new equation can be integrated to produce

\[
h^2 = \frac{2\kappa P_c}{\mu e} t \quad \text{(12)}
\]

Hence, the initial points of the experimental data plotted as \( h^2 \) against \( t \) should fit a straight line passing through the axes origin.

Taken into account that

\[
w = h \rho A_{\omega} \quad \text{(13)}
\]

where \( w \) is the weight increase of the fiber bundle due to the liquid uptake during the impregnation and \( A_{\omega} \) is the total pore area in a bundle available to flow, Eq 8 can be rewritten on a weight basis as follows:

\[
dw \over dt = \frac{\kappa \rho a_{\omega}}{\mu e} \frac{2P_{\omega}^2}{A_{\omega}} \frac{1}{W} \frac{-\kappa \rho a_{\omega}}{\mu e} \frac{2P_{\omega}^2}{A_{\omega}} \frac{g}{W} = a_{\omega} - b_w \quad \text{(14)}
\]

By using Eq 14 a differential approach of analyzing the fluid weight increase data is presented, where by plotting \( dw/dt \) against \( 1/w \) a straight line should fit the data with:

\[
slope = a_w = \frac{\kappa \rho a_{\omega}}{\mu e} \quad \text{and}
\]

\[
intercept = b_w = \frac{\kappa \rho a_{\omega} g}{\mu e} \quad \text{(15)}
\]

Again, for short times Eq 14 can be simplified to produce:

\[
w^2 = \frac{2\kappa P_{\omega}}{\mu e} t \quad \text{(16)}
\]

The disadvantage of using the weight data in Eq 14 and 16 is that, in order to estimate \( P_{\omega} \) and \( \kappa \), \( A_{\omega} \) is required to be known. Nevertheless, once the slopes \( a_h \) and \( a_w \) are known from the height and weight data respectively, \( A_{\omega} \) can be estimated according to the following relation, obtained by mathematical manipulation of Eq 9 and 15

\[
A_{\omega} = \frac{1}{\rho} \sqrt{\frac{a_w}{a_h}} \quad \text{(17)}
\]

The porosity of the fiber bundle can then be evaluated as

\[
\varepsilon = \frac{A_{no}/(N \pi R^2_f + A_{no})}{(18)}
\]

where \( N \) is the total number of fiber filaments in a fiber bundle and \( R_f \) is the radius of the fiber filament.

Upon integration of Eq 8, the time necessary for the liquid to reach a particular height can be determined as follows:

\[
t = a_h \frac{b_h}{b_h} \ln \left( 1 - \frac{b_h}{h} \right) + \frac{h}{b_h} \quad \text{(19)}
\]

or

\[
t = \left( \frac{-h}{b_h} \right) \left[ \ln \left( 1 - \frac{h}{h} \right) + \frac{h}{b_h} \right] \quad \text{(20)}
\]

where: \( b = \kappa \rho g / \mu e \) and \( h = a_h / b_h \) relax. Equation 20 is the basis of the integral approach in the analysis of the fluid height data. By replacing \( h = 0.99 h \), in Eq 20, the equilibrium time, \( t_{eq} \), arbitrarily defined as the required time for the fluid to reach 99% of the theoretical equilibrium height, can be estimated as

\[
t_{eq} = \frac{3.62 h}{b_h} \quad \text{(21)}
\]

A software package with a non-linear parameter fitting procedure can be used to fit the time against height data to Eq 20, producing values for \( h \), the equilibrium height, and \( b \), which can then be used to estimate \( a_h \), \( P_c \) and \( \kappa \). A similar integral procedure can be followed to analyze the weight data.

5. CAPILLARY FLOW EXPERIMENTS: RESULTS AND DISCUSSION

5.1 Silicone Oil

Every experiment produced data of fluid height and weight increase due to the liquid infiltration of the fiber bundle as a function of time, similar to the pattern of data shown in Fig. 3. The equilibrium state was not reached since both height and weight were still varying with time at the end of the experiment.

The integral fitting approach produced a good representation of the data, as can be seen in Fig. 3. By following the differential approach, the data of \( dh/dt \) against \( 1/h \) were fitted linearly according to Eq 8, although data scatter was more often observed, as can be seen in Fig. 4a. The \( 1/h \)-intercept of this curve corresponds to the equilibrium height, i.e. the time when the flow of liquid stops. Overall, for both height and weight data, the integral approach proved to be more reliable than the differential one, since the former smoothes the flow front advancement by considering all the history at each point while in the latter small variations in singular data point readings can lead to a much greater scatter. In the case where the integral fitting procedure is used to derive the \( dh/dt \) versus \( 1/h \) data points, a much better fitting is obtained, as can be observed in Fig. 4b. A similar trend was found...
in the analysis of the corresponding weight data (Figs. 5a and b). Hence, only the results of the integral fitting approach will be presented in the following paragraphs.

The scatter mainly observed in the low height readings in Fig. 4a may be due to difficulties in reading low height values, close to the liquid surface in the beaker, and also to the fiber wetting effect. The scatter in Fig. 5a is also mainly observed at the initial weight readings and could be attributed to the fiber wetting effect, the dominance of which is diminished as the experiment proceeds; a similar trend was reported by Hsieh (22).

The graphs presented in Figs. 6 and 7 refer to Eqs 12 and 16, respectively. According to these equations, the initial experimental points should not be dependent on the gravitational force acting on the column of liquid above the level of the liquid surface of the beaker and, therefore, the data in Figs. 6 and 7 should lie on a straight line. A straight line can indeed be fitted through the data up to about 500 s. The influence of gravity becomes increasingly important thereafter, slowing down the rate of height and weight increase with time and causing deviation of the data from the straight line.

Height and weight readings from a number of experiments conducted by using silicone oil as the infiltrating liquid are displayed in Figs. 8 and 9, respectively. The Figures show two main features that are applicable to all capillary infiltration experiments. First, the fitting procedure might fit more accurately a limited range of experimental points (see data from experiment 11, for example). This can be expected to influence the output values of the fitting procedure.

Second, as suggested by Batch et al. (14), the duration of the experiment influences the estimation of the equilibrium height \( h_e \). Examination of Figs. 8 and 9 and Table 3, especially experiments 8, 13 and 14, illustrates that although the fitting curves are practically coincident, the final values for the equilibrium height are 3.1 \times 10^{-2} m, 3.5 \times 10^{-2} m and 5.2 \times 10^{-2} m, respectively. The further the height is from the equilibrium position, the more underestimated the \( h_e \) value will be. Hence, the evaluated value for \( b_h \) will also be influenced by the duration of the impregnation experiments. An equivalent equilibrium height \( h_{eq} = w_e/A_\nu \) can also be predicted from the weight measurements and is presented in Table 4.

Tables 3 and 4 display the calculated variables from the analysis of the liquid height and weight readings, respectively, from a number of axial impregnation experiments of fiber bundles by silicone oil. The integral approach has been followed in the data analysis. First of all, it can be noted that the pore area of the bundle cross-section, \( A_\nu \), and the porosity, \( \varepsilon \), vary between the experiments. In a microstructural analysis of an epoxy composite specimen containing an uncompressed layer of fabric Y0212, it was found that \( A_\nu = 13.1 \times 10^{-6} \) m² on average for a fiber bundle in the fabric. In most experiments in Table 3, \( A_\nu \) is larger than that. One of the reasons for this might be linked with the handling of the fiber bundle during the experimental procedure. Another reason might be swelling of the fiber bundle as it is being impregnated by the liquid although the experimental data in Figs. 8 and 9 do not suggest acceleration of the impregnation as
time increases. Considering that different experiments were associated with different porosity values for the fiber bundle, the curves of height and weight changes as functions of time corresponding to the different experiments have no longer to be coincident.

Fig. 4. Analysis of height data of a typical experiment of axial capillary impregnation of a fiber bundle. (a) data fitting line according to the differential approach; (b) data points derived following the integral approach.

In fact, the capillary channels of the bundle and their properties are expected to be affected by many different factors, such as pore connectivity, distribution of pore sizes, total pore volume and surface properties of the individual fibers. Besides, the liquid is responsible for
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Fig. 5. Analysis of weight data of a typical experiment of axial capillary impregnation of a fiber bundle. (a) data fitting line according to the differential approach; (b) data points derived following the integral approach.
shifting the pore arrangement throughout the infiltration process (13, 22). The geometric properties of the fibrous medium and the flow behavior are, therefore, of a complex nature and difficult to quantify. As far as this work is concerned, the capillary data presented here correspond to an averaged behavior of the flow through the fiber bundle.

Given that the presented capillary experiments are far from equilibrium, the following relation has been used to calculate a theoretical value for the capillary pressure

\[ P_c = \frac{F (1 - \varepsilon)}{2 R_f \varepsilon} \sigma \cos \theta \]  

(22)
Fig. 8. Height data versus time from four experiments of axial capillary impregnation of a fiber bundle by silicone oil. Fitting curves have been constructed following the integral approach.

Equation 22 has been derived (26) for the axial impregnation of a unidirectional fiber assembly on the basis of the Young-Laplace equation with a theoretical value of the empirical parameter $F = 4$. By using Eq. 22 for the porosity values of the fiber bundles in the experiments in Table 3, theoretical values of different variables are calculated at equilibrium and are presented in Table 5. A comparison between Tables 3 and 5 indicates that both the $P_c$ and $h_e$ values derived from the experimental data are much lower than the corresponding values predicted by Eq. 22. At this point it must be mentioned that the values for $h_e$ determined

Fig. 9. Weight data versus time from five experiments of axial capillary impregnation of a fiber bundle by silicone oil. Fitting curves have been constructed following the integral approach.
from the experimental data are of the same order of magnitude as the ones determined experimentally by Batch et al. (14) for dioctyl phthalate (DOP) oil impregnating continuous roving glass fiber beds (with a similar \( \mu \)). The investigators in that study also expressed the opinion that equilibrium was not reached.

The values of \( P_e \) determined from the experimental data of this section are not consistent with the corresponding values of \( A_p \), and \( \epsilon \) of the fiber bundle, as it would be expected according to Eq 22. This might be a direct consequence of the influence of insufficient experimental time, which is further supported by the estimation of \( t_{eq} \), suggesting that the experiments were still far from equilibrium.

Table 3 also presents permeability values determined from the experimental data in each experiment. At this stage, considering the uncertainties about the determination of \( P_e \) from experimental data of impregnation curves still away from equilibrium, it is best not to try to correlate the variation of \( \kappa \) values with the porosity of the fiber bundle. However, this issue will be pursued rigorously in Part 2 of this study (1).

Following the progress of silicone oil through a glass-fiber bundle experiment for longer periods of time proved to be troublesome because of the arduous task of visually observing the flow front. Owing to the inherent characteristics of the oil and the low amounts of oil involved, the flow front tended to faint with time and not even the inclusion of blue or red dyes proved satisfactory.

\[ 5.2 \text{ Epoxy Resin} \]

The liquid height and weight data from three axial capillary impregnation experiments of fiber bundles conducted by using the epoxy resin (without a curing agent) are presented in Figs. 10 and 11. The height measurements showed a particularly good agreement between the three experiments and comparable results can be expected.

Table 4 shows the final results from the integral method of data analysis of the height readings and a very good agreement can be observed between the three experiments. Although the experiments were carried out for up to 2.5 hours, the estimated equilibrium times were much longer (\( \approx 18.4 \) h). The epoxy high viscosity was responsible not only for the long equilibrium times, but also for large \( A_p \) values (5.5 \( \times 10^{-2} \) m\(^2\)). It seems that due to insufficient load imposed to the bundle, as the fluid flowed through the pores, it opened them at an even larger extent than that obtained for the silicone oil, increasing the area available for the flow. Consequently, the bundle porosity in this case was higher (\( \epsilon \approx 0.75 \)). Table 7 presents the final values from the analysis of the weight data, with \( h_{eq} \) values lower than \( h_e \). This might be reflecting further difficulties in setting the balance reading to zero as soon as the bundle was put in contact with the high viscosity resin.

Table 8 presents the theoretical values of variables for the epoxy resin as estimated according to Eq 22 for two distinct values of porosity of fiber bundle. The

<table>
<thead>
<tr>
<th>Experiment</th>
<th>( P_e ) (Pa)</th>
<th>( h_{eq} ) theoretical (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>1919</td>
<td>0.23</td>
</tr>
<tr>
<td>11</td>
<td>5684</td>
<td>0.68</td>
</tr>
<tr>
<td>13</td>
<td>11771</td>
<td>1.40</td>
</tr>
<tr>
<td>14</td>
<td>8862</td>
<td>1.06</td>
</tr>
</tbody>
</table>
Axial Impregnation of a Fiber Bundle. Part 1

Fig. 10. Height data versus time from three experiments of axial capillary impregnation of a fiber bundle by epoxy. Fitting curves have been constructed following the integral approach.

The same problems encountered for the silicone oil are repeated here, where the values for $h_e$ and $h_{eq}$ estimated from the experimental data are much lower than the theoretical values because of insufficient duration of the capillary impregnation experiments.

In this case, it was decided to carry out another experiment for a single fiber bundle using the epoxy resin, where the experiment was allowed to run for a much longer period ($\approx 23$ days). The same setup was used as in the case of silicone oil, but this time the lower end of the fiber bundle was attached with tape at the bottom of the beaker as an attempt to restrict the opening of the pores within the bundle due to the flow of resin.

The height of the flow front as a function of time and the fitting curve according to the integral method are shown in Fig. 12 along with the experimental data (experiments 19, 20 and 22) previously shown. The

Fig. 11. Weight data versus time from three experiments of axial capillary impregnation of a fiber bundle by epoxy. Fitting curves have been constructed following the integral approach.
fitting curve of the long-run experiment is within reasonable agreement with the bulk of the previous experiments and differences should be mainly attributed to individual characteristics of porosity and, consequently, capillary pressure and permeability. Equilibrium was still not reached after 23 days, with a resin rise of 5 mm/day at the end of the experiment. The values derived from the two fitting procedures of data analysis for the long-run experiment are shown in Table 9, and it can be seen that the \( P_c \) values are now much greater for both fitting methods. One peculiarity is that \( t_{eq} \) times are now in the range of years. The permeability value estimated from the analysis of the epoxy height readings of the long-run experiment was \( \kappa = 1.2 \, \mu m^2 \).

Equation 22 was applied again to calculate the theoretical values of variables at equilibrium for a porosity of the fiber bundle \( \varepsilon = 0.47 \), corresponding to the porosity of the fiber bundle in the long term experiment as determined by the microstructural studies described in section 4.3. The estimated theoretical values for \( P_c \) and \( h_r \) are shown in Table 8. It can be seen that now the theoretical values for \( h_r \) and \( P_c \) are in a much better agreement with the experimental data of the long-run capillary experiment (Table 9), within a 7% difference.

### 6. CONCLUSIONS

This study comprises experimental investigations into the axial capillary impregnation of an E-glass fiber bundle by two alternative Newtonian fluids: (a) silicone oil and (b) epoxy (without the hardener). First of all, the surface tension of the fluids and their contact angle with a single E-glass fiber filament were measured. The values were fed into Eq 22 to yield theoretical predictions of \( P_c \) for axial capillary impregnation of an aligned fiber bed. In general the wetting properties of the liquid/fiber systems yielded higher capillary pressure for the epoxy than the silicone oil for the same value of porosity of the fiber bed.

Axial capillary impregnation experiments of a single E-glass fiber bundle using either silicone oil or epoxy as the infiltrating liquid yielded liquid height and weight data as a function of time, which could be further analyzed to determine the capillary pressure and the permeability. The integral approach proved better in the data analysis and was generally applied in this study. Capillary experiments of a few hours duration resulted in underpredicting the capillary pressure of the system, which is consistent with past studies from the literature. A long-term capillary experiment of 23 days using epoxy yielded a value of \( P_c = 9600 \, Pa \), which was comparable to the theoretically predicted value of \( P_c = 10,290 \, Pa \). This is a substantial improvement compared to values of \( P_c \) previously reported in the literature and leads to the conclusion that data plots obtained from typical capillary impregnation experiments of short to medium duration may not be suitable to determine accurate values for capillary pressure and permeability. However, they are still useful to validate theoretical infiltration curves which will really supply more accurate values for \( P_c \) and \( \kappa \). Such theoretical modeling and validation is going to be conducted in Part 2 of this study (1).
Axial Impregnation of a Fiber Bundle. Part 1

![Graph showing height data versus time for four experiments of axial capillary impregnation of a fiber bundle by epoxy. The fitting curve has been constructed following the integral approach.]

Fig. 12. Height data versus time from four experiments of axial capillary impregnation of a fiber bundle by epoxy. The fitting curve has been constructed following the integral approach.

Table 9. Long-Term Axial Capillary Impregnation Experiment for a Fiber Bundle Using Epoxy: Estimated Results From Height Readings.

<table>
<thead>
<tr>
<th>Method</th>
<th>$b_h \times 10^6$ (m/s)</th>
<th>$A_h \times 10^7$ (m$^2$)</th>
<th>$\varepsilon$</th>
<th>$P_s$ (Pa)</th>
<th>$h_s \times 10^2$ (m)</th>
<th>$\tau_s$ (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Integral approach</td>
<td>1.29</td>
<td>1.61</td>
<td>0.47</td>
<td>9600</td>
<td>0.86</td>
<td>2816</td>
</tr>
<tr>
<td>Differential approach</td>
<td>1.32</td>
<td>1.61</td>
<td>0.47</td>
<td>9860</td>
<td>0.89</td>
<td>2819</td>
</tr>
</tbody>
</table>

ACKNOWLEDGMENT

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REFERENCES

1. S. Amico and C. Lekakou, Polymer Composites, this issue.