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OPTIMIZING THE FIBRE WETTING IN A NOVEL POLYURETHANE FOAM SANDWICH COMPOSITE

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ABSTRACT: This paper describes the process for manufacturing sandwich composites using polyurethane foam as core material, engineered fibreglass as reinforcement and thermoplastic skins. This new process is the result of a convergence of several processes: sheet thermoforming, preforming using ultraviolet cured adhesives, and polyurethane foam injection. It is greener than traditional sandwich composite processes, as no volatile organic compounds (VOC's) are emitted during manufacturing. The final products have a thermoplastic skin moulded in colour finish requiring no sanding or post painting. Depending on final use of the product, properties can be engineered for weather ability, chemical resistance, temperature resistance as well as colour and texture. The novelty of the process occurs when the liquid polyurethane resin is injected between two preformed thermoplastic sheets with fibreglass layers. The self skinning polyisocyanurate HFC blown foam[1] makes it possible to create the three-dimensional foam core, wet out the dry fibreglass and bond the thermoplastic surface skins during one processing step. This paper details the analysis of the polyurethane reaction that takes place during manufacturing. The foaming behaviour and wetting of the fibres were studied under different manufacturing conditions. Optimization of the composite laminate was finally carried out by improving the foaming conditions.

KEYWORDS: polyurethane, sandwich structures, UV cured preforms, RIM, LCM

INTRODUCTION

The use of composite materials in the industry has experienced significant growth during the 20th century. Composite materials are seen as a key alternative to common materials such as steel and other metals, because of the ability to tailor the mechanical properties and due to their low specific weights. Due to the rising cost of energy, the use of composites is expected to grow even faster, particularly in the automotive and aerospace industries. Energy efficiency is recognized as one of the greatest challenges facing these transport industries.

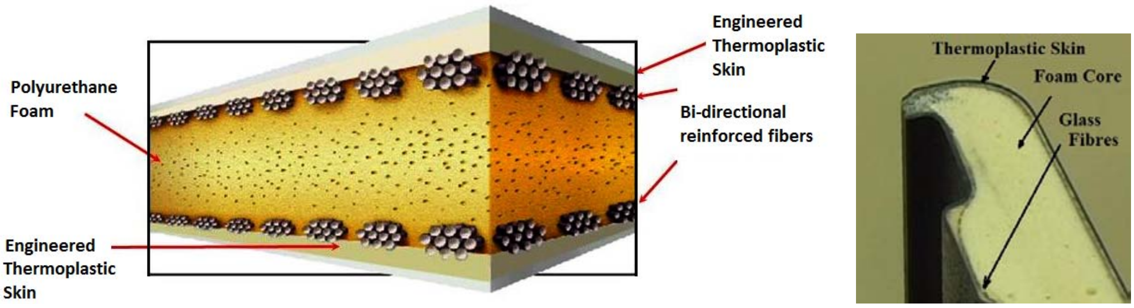
In the automotive industry, repeatability, robustness of the manufacturing processes and cycle time are key indicators of the performance capabilities needed for high volume manufacturing. Sandwich structures are in demand in these industries; however, processing techniques for such composite parts are often slow and expensive or rapid but with low mechanical properties.

A new foaming process recently developed for manufacturing composite parts has shown potential for making medium performance sandwich composites for high production volumes. This concept uses the versatility of polyurethane foam to create a low density foam core and a high density fibreglass laminate at the same time.

This research focuses on understanding the foaming process that takes place in a closed mould and the impregnation of the fibrous reinforcement. The polyurethane foam was characterized under atmospheric conditions and at elevated pressures up to 400 kPa (4 bars). The physical phenomena such as diffusion of the blowing agent, temperature and viscosity changes were studied to understand the foaming behaviour. The kinetic events such as the boiling of the blowing agent, followed by morphological changes in the foam, were also analysed in the present work. In order to characterize the foam at elevated pressures a test apparatus was designed and constructed. This test mould was also used to evaluate the impregnation of the fibres during foaming.

OVERVIEW OF THE MULTIPLE STEP PROCESS

The multiple step process studied in this work is the result of a convergence of various technologies; sheet thermoforming to form the inner and outer thermoplastic skins, vacuum pre-forming to shape the reinforcing fabrics, ultraviolet curing to rapidly bond the fabrics to the thermoplastic skin, reaction injection moulding (RIM) to create the core, wet out of the fibres and bond to the skins. The final product is a 3D sandwich structure, which is presented in Fig. 1a. A cross section of a moulded composite is also shown in Fig. 1b.



(a)

(b)

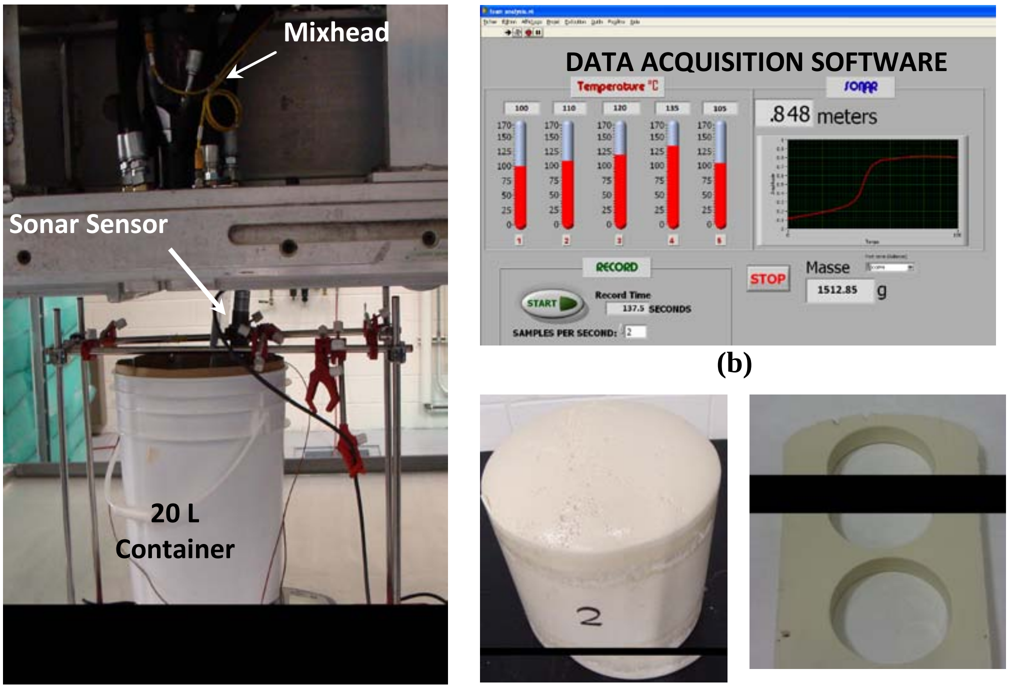
Fig. 1: (a) Schematic section of the PU sandwich composite[2]; (b) and cross-section of a moulded part.

Free Rise Analysis

The analysis carried out in this research focused on determining the free rise behaviour and density for a rigid polyisocyanurate foam formulation with different stoichiometric proportions. The stoichiometric proportion is evaluated through the isocyanate Index, which is defined as the ratio of isocyanate functional groups and available hydroxyl groups (NCO/OH). The isocyanate index can be calculated as follows[3]:

$$\text{Isocyanate Index} = \frac{\text{Actual amount of isocyanate used}}{\text{Theoretical amount of isocyanate required}} \quad \text{X} \quad 100 \qquad (1)$$

The experimental setup developed to measure the free rise behaviour is shown in Fig. 2a. The two component foam system was injected using a high pressure impingement metering machine (Cannon A200) with a Cannon FPL24 mix-head. The resin was injected at a constant flow rate of 1000 g/s, while the impingement pressures were set at 7000 kPa (70 bars) for each component. The ratio of the polymeric methylene diphenyl diisocyanate (pMDI) and polyol resin blend was varied to achieve the desired stoichiometric proportions. The mix was injected into a 20 litre container used to measure the free rise of the foam (see Fig. 2a). The rise profile was measured using Senix® ToughSonic®/PC distance sensor. The temperature readings were made using type J thermocouples, and the gas losses were measured with a precision scale.



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(b)

(a) (c) (d)

Fig. 2: (a) Foam free rise apparatus; (b) data acquisition software; (c) moulded part; and (d) location for the density samples.

A total of 12 injections were carried out using 6 different conditions, varying the isocyanate index from 110 to 160. For each injection, the exothermic temperature was recorded at five positions vertically along the wall of the container at 50 mm increments and one in the bottom centre of the container 25 mm deep into the foam. The rise profile and weight loss were also recorded. Fig. 3 shows the experimental results for an injection at an isocyanate index of 160. It can be seen that the start of polymerization occurred at 12 seconds after injection (see cream time in Fig. 3, position (a)). The gelation occurred at 66 seconds (see string gel in Fig. 3, position (b)), and the foam surface became tack free at 88 seconds (see tack free time in Fig. 3, position (c)). The foam rising behaviour shows a constant foaming rate until 60 seconds followed by a second acceleration. This acceleration occurs slightly ahead of the exothermic gradient. The foam movement prior to gelation will aid in fibre wetting; therefore it is desirable that the foam penetrates the fibres as early as possible before gelation.

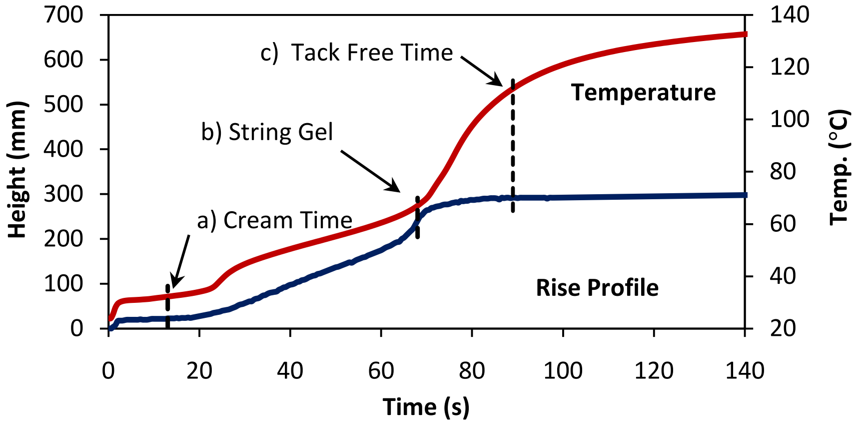


Fig. 3: Experimental results of an injection at isocyanate index of 160.

Fig. 4 illustrates the experimental results for an injection carried out at an isocyanate index of 140. Comparing with Fig. 3 it can be seen that while the beginning of polymerization occurs at the same time, decreasing the stoichiometric ratio speeds up the gel time to 62 seconds, and the tack free time to 82 seconds. The exothermic peak is also higher for the lower stoichiometric ratio (147°C versus 140°C not shown in the graphs). At the lower index the quantity of amine catalyst and HFC blowing agent increases while the quantity of isocyanate decreases. The increase in available catalyst accelerates the rates of reaction while the larger amount of HFC blowing agent increases the mechanical blowing of the foam.

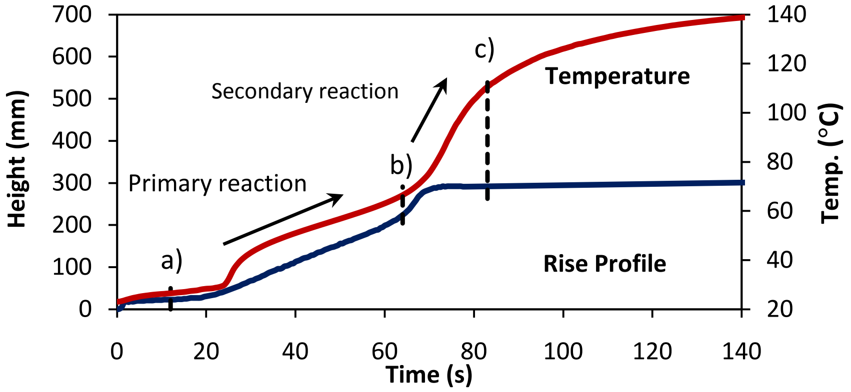


Fig. 4 : Experimental results of an injection at isocyanate index 140.

The 12 sample parts injected under different conditions were cut into smaller cylinders to measure the foam density according to ASTM D1622. The samples measured approximately 150 mm in diameter x 40 mm in thickness; they were cut through the central axis of the part as shown in Fig. 2d. This cutting plan was chosen to observe potential density gradients. As shown in Fig. 5, the lowest foam density was achieved at an isocyanate index of 110. While increasing the stoichiometric proportions the caloric value in the foam increases; however, the percentage of blowing agent and percentage of primary and secondary catalysts decrease. This results in a decrease in blowing efficiency and a smaller developing pressure in the foam. As well, there is a delay in the gelation of the foam. In this particular process, where a fibrous reinforcement is impregnated by the collapsing foam, resin pressure and viscosity turn out to be important parameters so as to enable the wetting phenomena.

If resin gelation occurs too early in the process, the fibrous reinforcement will not be properly impregnated. Reducing the stoichiometric proportions increases the foam pressure, but speeds up the cure reaction. Mechanical properties of the polyisocyanurate foam also decrease with the stoichiometric proportions, partly due to a plasticizing effect caused by the HFC blowing agent. Optimizing the foam formulation becomes then a key part of the process, since maximizing the stoichiometric ratio will lead to better foam properties, but will increase the complexity of the fibre impregnation stage. As shown in Fig. 5, the lowest free rise density is achieved at an isocyanate index of 110. The free rise foam density is much lower than the density needed to fill the mould. This is due to frictional resistance and back pressure created inside the closed mould. In order to assess foam filling capability, the amount of "overpacking" the foam should be determined. The pressure in the mould is dependent on the degree of overpacking *E*. This is defined by[4]:

$$E = \frac{\text{Required foam density}}{\text{Free rise foam density}} \tag{2}$$

The degree of overpacking increases pressure, thus making it easier to fill the mould. For these experiments the moulding densities equate to *E* values from 3 to 4. A second set of experiments was run from at moulding densities from 192-256 kg/m^3.

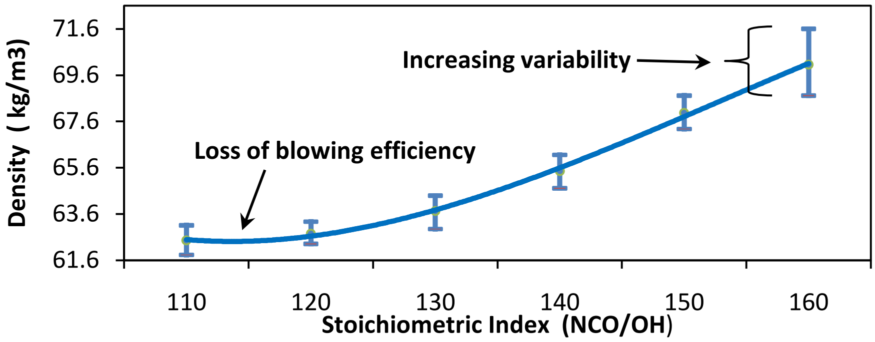
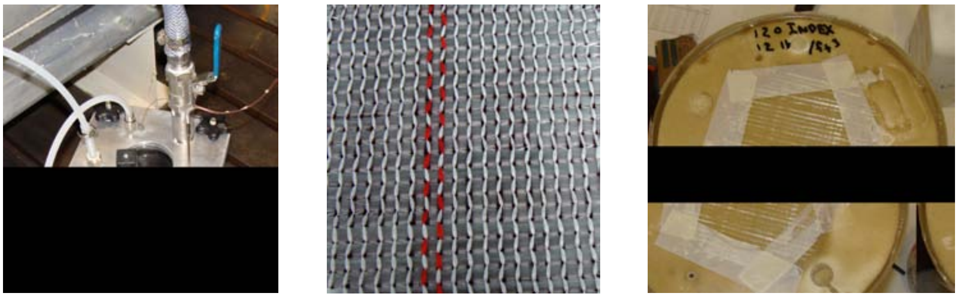


Fig. 5 : Results of the free rise density characterization.

Analysis of resin foaming under pressure

A total of 9 injections were made over a range of isocyanate indices from 120 to 140 and a range of densities from 192 to 256 kg/m^3. The objective of this analysis was to understand the dynamic viscosity, pressure development and exothermic reaction that take place during foaming in a closed mould. This study was carried out using a closed mould conceived to measure internal pressure of the foam, its dynamic viscosity and to observe the impregnation of the fibrous reinforcement. The cover of the mould was designed to visualize the impregnation of the fibres through a glass window. The lid was fitted with a Micromet impedance sensor, a thin film pressure transducer model DTMFB and a J-type thermocouple positioned 25 mm into the cavity of the mould. The polyurethane foam was injected into the mould using a hose connection to the mix-head. A piece of reinforcing fabric was bonded to the internal side of the lid using two sided adhesive. An engineered glass fabric manufactured by Fiber Glass Industries with an areal weight of 486 g/m^2 was used for this set of experiments (see Fig. 6b). Before resin injection, the atmosphere inside the mould was reduced to near full vacuum. The injection parameters were kept constant with the first set of experiments. A moulded foam part with the glass fibres is shown in Fig. 6c.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(a) **(b)** **(c)**

Fig. 6 : (a) Closed mould, (b) dry glass fabric and (c) moulded part.

A two factor three level experiment was conducted. The factors were: isocyanate index (120–130–140) and density (192–224–256 kg/m^3). To simulate the viscosity behaviour the logarithmic changes in the ionic viscosity were monitored. There has been considerable work in-situ monitoring of matrix properties during cure using electrical methods such as dielectric sensing.[5] Additional measurements included real time pressure, and temperature recording. Physical measurements on the parts included density, skin thickness and laminate thickness.

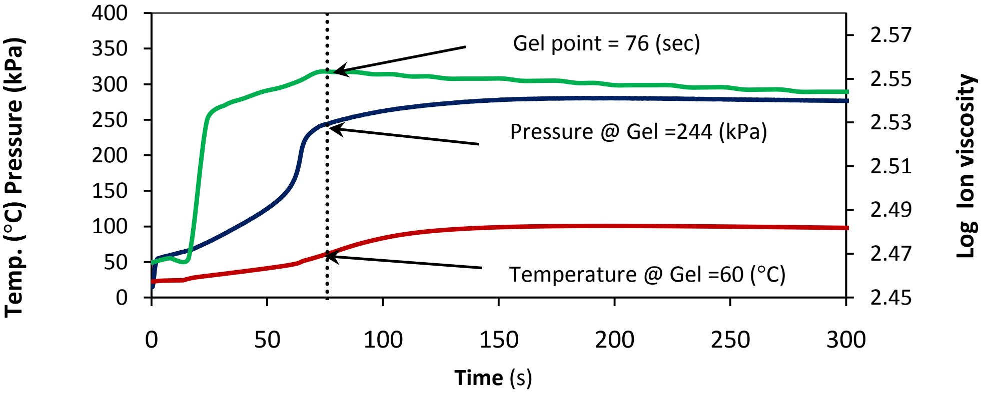


Fig. 7 : Observations at an Index 120 and density of 192kg/m3.

As shown in Fig. 7 & 8, significant changes in the foam behaviour were observed when varying the manufacturing conditions. At higher index and higher density (see Fig. 8) the gel point is delayed 8 seconds, while pressure is increased by 71 kPa and temperature of reacting foam is 19°C higher at gel. These conditions result in changes in the foam morphology and skin thickness as shown in Fig. 9.

The experiments demonstrated that the index has less influence than the density. The results for the fibre impregnation are shown in Fig. 10. Improved wetting resulted when the index was 140, and the density 256 kg/m^3. For this experiment the pressure at gel was measured at 315kPa. The two images also show that by increasing the density and the isocyanate index simultaneously the saturation of the fibreglass improves. In the experiments where the density was constant at 192 kg/m^3 and the index values range from 120 to 140, the fibre saturation was reduced and continued to decrease as the index increased. At this lower density the pressure prior to gelation is insufficient to wet out the fibres measured at 244 kPa for the 120 index part and 225 kPa for the 140 index part. This drop in pressure occurs because the percentage of blowing agent and catalyst decrease as the index increases. In order to develop the best sandwich composite, the properties of the laminate and the foam must be optimized together. Increasing the index improves the mechanical properties of the foam, but makes it more difficult to wet out the fibres and increasing the density add weight and cost to the final part.

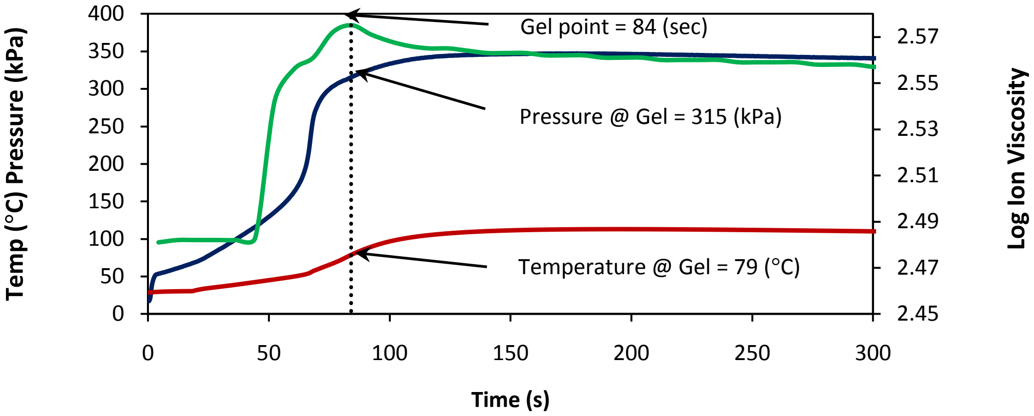
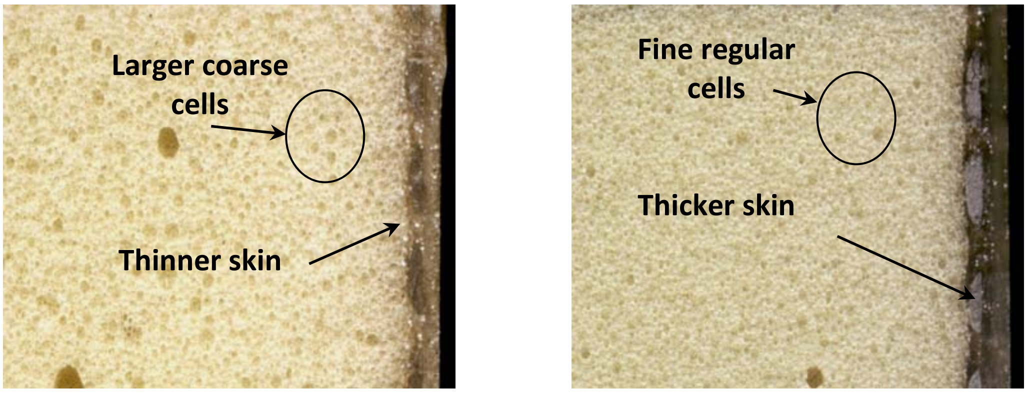
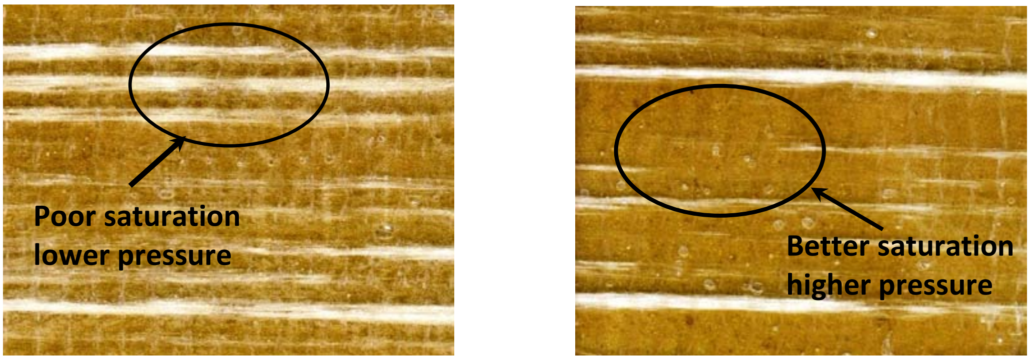


Fig. 8 : Observations at Index 140 and 256 kg/m^3.



(a) (b)

Fig. 9 : (a) Foam observations at 120I and 192kg/m^3 and (b) at 140I and 256kg/m^3.



(a) (b)

Fig. 10 : (a) Laminate observations at 120I and 192kg/m^3 and (b) 140I and 256kg/m^3.

CONCLUSIONS

This novel technique for developing advanced composite sandwich structures using the self skinning behaviour of polyurethane foam offers exciting new opportunities to rapidly manufacture complex structures with a reduced number of manufacturing stages. This research focussed on understanding the wetting of the fibrous reinforcement during foaming. Two moulds were conceived and instrumented to analyse the foam free rise and wetting of an engineered glass fabric. It was shown that there is an optimum index and density which result in a delayed gel time and increased pressure at gel. This combination results in improved fibre wetting behaviour. Increasing the foam density also improves wetting of the fibres due to an increase in internal pressure, but can negatively impact the properties and costs.

Future work will focus on the understanding the chemical changes in the foam as well as their impact on mechanical properties of the sandwich composite.

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