Resin Film Infusion of Cyclic PBT Composites: Consolidation Analysis

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SUMMARY: The focus of this paper is to examine the mechanisms involved in the consolidation of stitched glass fiber reinforced cyclic polybutylene terephthalate (CBT™) during processing, with a particular emphasis on sandwich structures. A pre-preg form of the material was processed under vacuum using a resin film infusion technique on a flat plate heated mold with topside heating and temperature and displacement are recorded. The final application of these structures is aimed at the manufacture of wind turbine blades in the wind power industrial sector, where sandwich structures are typically used. CBT™ is a new low viscosity thermoplastic matrix, suitable for the manufacture of structural composites for automotive, electrical, sports equipment, powder coating, and wind power applications. Consolidation is examined as a function of the processing parameters - temperature, vacuum pressure and time, and mechanisms of intimate contact, autohesion and fiber impregnation are considered for a prediction model.

KEYWORDS: resin film infusion; cyclic polybutylene composites; consolidation; sandwich structures; thermoplastic matrix; intimate contact; autohesion; fiber impregnation.

INTRODUCTION

The CBT™ system holds a number of advantages over thermoset systems including improved toughness, damage tolerance and ease of recycling of composite parts. CBT™ has a low processing viscosity (below 20 mPa.s at 180°C) which allows rapid and thorough wet-out of fiber reinforcements[1,2]. The combination of low viscosity, rapid reaction or polymerization cycles, and the benefit of isothermal processing (hence the ability to demold at higher temperatures) leads to a shorter overall process time. The polymerization reaction emits no exotherm, which eliminates the problem of 'hot-spots' that may damage parts in the mold, or cause internal stresses or warping. This also greatly reduces cycle times in large parts where thermoset thick walled sections currently pose a threat of overheating unless processed slowly. This is particularly applicable to wind turbine blade 'root section' manufacture where slow cycle times increase cost and lower productivity. Most other sections of wind turbine blades are based on composite sandwich structures, which offer excellent stiffness and strength for low weight[3]. In this case, glass fiber reinforced CBT™ skins are separated by thick, lightweight, high

temperature, closed cell foam core – the thicker the core, the higher the flexural stiffness and strength of the panel – for minimum weight gain [4].
The resin film infusion process has increased in popularity for the manufacturing of structural composites for aerospace, automotive and military applications, and has been identified as an alternative cost-effective manufacturing technology to RTM [5]. The combination of an open mold and a vacuum bag reduces the tooling cost of the resin film infusion process, and facilitates the manufacture of large structures, such as wind turbine blades.

During the RFI processing of thermoplastic matrix composites, consolidation may be characterized by three major steps: intimate contact, autohesion, and fiber impregnation[6]. Consolidation of thermoplastic composites can be considered as an autohesion process in which molecular chains diffuse across the interface and entangle with neighboring chains. The interface is created by intimate contact achieved by an applied pressure [7].It is believed that the strength of the interply bonds depends mainly on two mechanisms: intimate contact and diffusion bonding. During fiber impregnation, the matrix is introduced into the space between the fibers. It is important to predict the thickness reduction of the material after processing in order to design for accurate tolerances, and allow for assembly of large structures such as wind turbine blades. Therefore, this paper focuses on the consolidation behavior of glass fiber reinforced cyclic PBT.

MATERIALS

The monomer used in these trials is cyclic butylene terephthalate oligomers (CBT™), and are supplied by Cyclics Corporation. The form of material used here is a one-part 'pre-preg'. The pre-preg supplied is unidirectional, stitched glass fabric (951 g/m^2) , dry on one side, and with a coating of pre-catalysed resin on the other, and is technically an intermediary between pre-preg and resin film material. Due to the design of the material a large degree of debulking is observed once vacuum is applied, represented by figure 1. A plot showing the debulking of ten layers of pre-preg material at room temperature is shown in figure 2, which illustrates a 42% decrease in thickness for a 12 ply laminate. The further trials discussed in this paper do experience the 'debulking' phenomenon, however consolidation is the main point of focus.

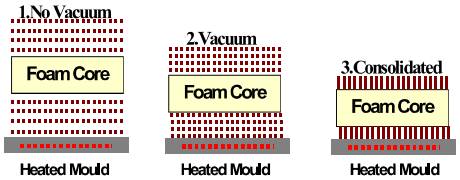

Fig.1: Debulking Phenomenon

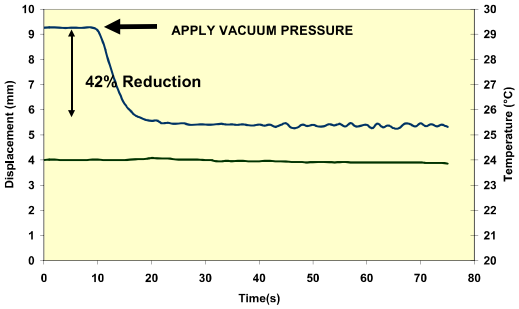

Fig.2: Debulking at room temperature

The pre-preg was dried under vacuum at 120°C for 12 hours, and purged periodically with nitrogen to remove any residual moisture, as moisture acts as an inhibitor to the polymerization reaction. Thick parts are processed with thermocouples placed at intervals between layers to monitor the temperature throughout the part. For sandwich panels, the CBT™ resin is processed

'in-situ' with a high temperature closed cell foam, thus ensuring an excellent bond between the two surfaces. The foams used in these trials include a high temperature Polyisocyanurate (PIC) foam and a developmental grade of PET foam.

EXPERIMENTAL

The tooling used in these experiments is a stainless steel flat plate mold, capable of rapid heating rates, precise temperature control, uniform temperature distribution, and accurate temperature monitoring capabilities. A portable heated top plate is used to ensure uniform heating. An ideal process temperature-time plot for processing of CBT™ is outlined below (fig.3), and is described in four stages; stage 1: initial temperature ramp, stage 2: drying stage (120°C), stage 3: secondary temperature ramp, and stage 4: secondary dwell (190°C). The critical stage in this process is the secondary temperature ramp, or stage 3, at which point the melt viscosity starts to decrease[8]. Temperature was measured using a temperature data logging system, reading temperatures from thermocouples located at various points in the lay-up. This data logging system also recorded displacement by using data from a linear variable differential transducer (LVDT). Vacuum pressure was monitored using a digital vacuum gauge.

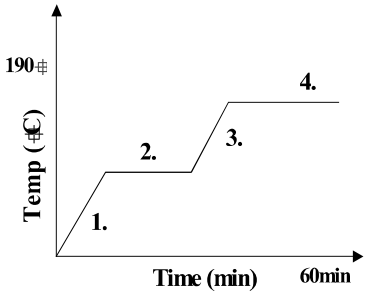

Fig.3: Ideal Temperature-Time Profile for Profile for Processing of CBT™

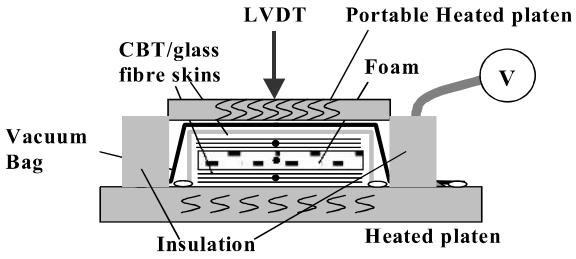

Fig.4: Tool Schematic

The apparatus described was used to study the behavior of CBT glass fiber reinforced parts, and sandwich structured parts using the same materials. Figures 5&6 below are plots taken from the process cycle of a 12 ply, unidirectional glass fiber reinforced CBT™ part, and a sandwich structured part consisting of six layers of unidirectional glass fiber with a PIC foam core (thickness=6mm), respectively. In figure 5, a slight decrease in thickness may be observed as the temperature is increased from room temperature to 120°C. This is due to the nature of the pre-preg material which is quite 'boardy' and becomes slightly more pliable as the temperature is increased. During the drying stage, the part thickness reaches an stable value, as the temperature is stabilised at 120°C. The matrix remains in solid form at this stage. A slight decrease in thickness may be observed between 130°C and 140°C as the resin starts to soften, and this is followed by a more rapid decrease as the temperature is increased to the processing temperature of 190°C. The part thickness decreases rapidly, within a few minutes. The rapid increase in temperature causes the viscosity of the resin to decrease dramatically (from 150mPa.s to <20mPa.s),and flow readily, leading to the impregnation of the glass fiber as the resin fills the spaces between the fibers thus resulting in a 65% decrease in thickness.

Figure 6 shows a similar pattern for the sandwich panel. The initial increase in the thickness of the part, as highlighted in section 'A', is due to the foam core, which expands slightly due to off-gassing once temperature is applied, however the degree of expansion is negligible, (<0.5mm). This effect was studied in trials measuring the effect of temperature on the foam alone. Again, the thickness of the part decreases rapidly as the temperature rises. Section 'B' exhibits a thickness 'recovery'. It is hypothesized that this phenomenon is a viscoelastic recovery of the matrix due to an increase in the partial pressure within the vacuum bag as the resin starts to flow, and vacuum channels are limited. The low viscosity of the resin at high temperatures means that it is imperative that the resin is not encouraged to flow outside of the fiber reinforcement as this will increase the volume fraction of the composite part, which has implications for mechanical properties. The effects of squeeze flow have been examined in this study and it has been shown that displacement is much greater for parts where squeeze flow has occurred.

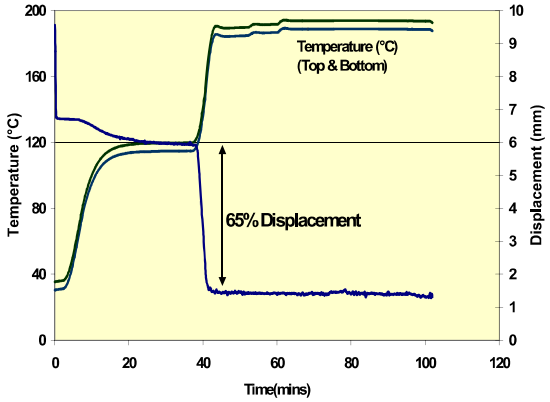

Fig.3: Displacement:12 ply UD laminate

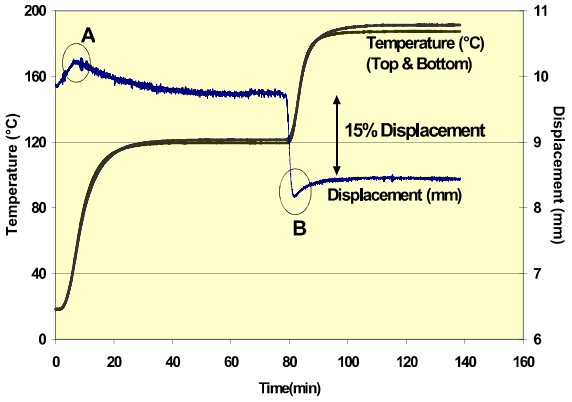

Fig.4: Displacement:6 ply UD; foam core

MODELING

In consolidation models to date, based on pressure evolution analysis[9,10], it is assumed that the applied pressure is transmitted to the matrix as well as to the fiber reinforcement. The forces in the resin and the fibers balance the applied load thus;

$$P_{app} = P_f + P_r \qquad (1)$$

Where P_{app} is the applied pressure, P_f is the fiber pressure, and P_r is the resin pressure.
Consolidation at process temperature without vacuum pressure results in the plies bonding together by resin bridges, however large voids visible to the eye are present between the plies suggesting that no intimate contact is developed under these conditions. Therefore, to complete intimate contact, pressure must be applied to complete the resin bridges. The Lee & Springer model of the degree of intimate contact in unidirectional fiber-reinforced thermoplastics represents the initial ply surface as a series of equisized rectangles, with the present voids also represented as equisized triangles [11]. Based on this geometry, the deformation of the rectangular resin elements can be defined as;

$$b(t) = b_0\left[1+\frac{5\phi P_{app}t}{\eta}\left(1+\frac{w_0}{b_0}\right)\left(\frac{a_0}{b_0}\right)^2\right]^{\frac{1}{5}} \quad (2)$$

where b(t) is the width of the rectangular elements at time t, η is the viscosity, w_0, a_0 and b_0 are the initial dimensions of the rectangular resin elements, φ is a constant for the pressure distribution and P_{app} is the applied pressure. Intimate contact is achieved once b(t) is equal to b_0+w_0. Based on this equation, the time needed to complete intimate contact t_{ic} is;

$$t_{ic} = \left[\left(\frac{b_0+w_0}{b_0}\right)^5 - 1\right]\left(1+\frac{w_0}{b_0}\right)^{-1}\left(\frac{a_0}{b_0}\right)^{-2}\left(\frac{\eta_0}{5\phi P_{app}}\right)$$

(3)

where η_0 is the shear viscosity (10 Pa/s) and is expressed as:

$$\eta_0 = 6x10^{13}\exp\left(-0.779/T\right) \quad (4)$$

where T is the temperature in degrees Kelvin. The constants were determined based on data supplied by the manufacturer. The viscosity of the resin is greatly dependent on the temperature to which it is exposed, and this relationship is important when considering the final step in the consolidation process, fiber impregnation. Impregnation is achieved by heating the lay-up to a temperature at which the matrix becomes liquid enough to penetrate the remaining space between the fibers, The depth of the fibers penetrated by the matrix in a given time can be described by Darcy's Law[12]:

$$v_{flow} = \frac{K}{\eta}\frac{dP}{dx}$$

(5)

where v_{flow} is the flow rate of the matrix through the fiber bed, K is the permeability, η is the viscosity, and dP is defined as $P_{atm} - P_{par}$ where P_{atm} is perfect vacuum, or atmospheric pressure, and P_{par} is the partial pressure within the vacuum bag, which for perfect vacuum, is assumed as negligible. Therefore equation (5) becomes:

$$\frac{d\zeta}{dt} = \frac{4K_p}{\eta}\frac{P_{atm}}{\zeta-\zeta_0} \quad (6)$$

where $K_p=K/(1-V_f)$ where V_f is volume fraction, and ζ_0 is the thickness of the fiber tow, and ζ is the thickness of the non impregnated zone. Integrating and solving, gives:

$$\zeta(t) = \zeta_0 - \sqrt{\zeta_0^2 - 2\left[\zeta_0\zeta_i - \frac{4K_p}{\eta}\left(P_{atm}-P_{par}\right)t\right]} \quad (7)$$

This model gives an approximation for the thickness of the impregnated zone as a function of time, and temperature, and at constant pressure.

CONCLUSIONS & FURTHER WORK

An experimental study has been carried out on the consolidation behavior of cyclic polybutylene terephthalate resin (CBT™) and a prediction model has been put forward. Experimental results show that at constant pressure (vacuum pressure) the change of viscosity with respect to temperature is the main governing factor behind the consolidation of the composite parts. Once polymerization of CBT™ commences the viscosity of the resin starts to increase until crystallization is complete. The model gives an approximation of thickness. Work is ongoing to complete comparisons of experimental and numerical results. Areas to be investigated include the effect of fiber orientation on consolidation – to date, unidirectional fibers only have been used. Further work will also investigate the consolidation behavior of very thick parts, in excess of 40 ply's.

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