



Rapidly cured epoxy/anhydride composites: Effect of residual stress on laminate shear strength



S.L. Agius^{a,*}, M. Joosten^{a,b}, B. Trippit^c, C.H. Wang^b, T. Hilditch^a

^a School of Engineering, Faculty of Science Engineering and Built Environment, Deakin University, Waurn Ponds, VIC 3217, Australia

^b Sir Lawrence Wackett Aerospace Research Centre, School of Aerospace, Mechanical, and Manufacturing Engineering, RMIT University, GPO Box 2476, Melbourne, VIC 3001, Australia

^c Simuserv Pty. Ltd, Melbourne, VIC 3123, Australia

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ABSTRACT

The drive towards rapid cure thermosetting composites requires a better understanding of the residual stresses that develop during curing. This study investigates the impact of residual stresses on the interlaminar shear strength of resin-infused epoxy/anhydride carbon-fibre laminates. The magnitude of the residual stress was varied by changing the initial injection cure temperature between 75 °C and 145 °C. The corresponding cycle times and the final glass transition temperature of the resin were also measured. The experimentally measured chemical shrinkage and thermal expansion properties of the resin after vitrification were used as inputs to a finite element analysis to calculate the peak residual stresses in the composite. An increase in the initial cure temperature from 85 to 135 °C resulted in an increase of 25% in the residual stress, which led to an experimentally measured reduction in the composite's short beam shear strength of approximately 16% (8 MPa), in good agreement with model prediction.

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1. Introduction

High production rates are critical to the widespread use of carbon-fibre reinforced thermosetting composites [1]. Resin injection coupled with high cure temperatures, typically greater than 130 °C, to facilitate faster cure rates have proven successful in achieving rapid cycle times [2]. However, high cure temperatures can negatively impact the final properties of resultant thermosetting composites as result of the high residual stresses and reductions to final glass transition temperature.

Residual stresses in cured epoxy composites are mostly unavoidable because fibre and resin have different thermal expansion properties [3]. Changes in resin volume can result from two mechanisms: chemical shrinkage and thermal expansion [4–6]. Higher cure temperatures have been shown to cause higher thermal residual stresses, which in turn can lead to excessive shape distortion and/or matrix cracking [4,6,7]. Composites are susceptible to the negative impacts of residual stresses when they are of the same sign as those resulting from external loads [8]. Material strengths in transverse tension, compression, flexure and interlaminar shear are affected by residual stresses [8,9]. Thus it is important to account for residual stresses when designing composite

structures. Except for the transverse tension case, the effects of residual stresses on other load cases have received less attention, particularly at the structural level [8]. The effect of residual stress on laminate shear strength will be the focus of this study.

This study investigates an epoxy/anhydride resin as they are suitable for land vehicle structures where high production rates and low costs are major requirements. Epoxy/anhydrides have been traditionally used in filament winding and pultrusion applications and their use in high temperature processes has received less attention despite offering several advantages over other matrix resin chemistries, such as epoxy/amines. With a low reactivity, epoxy/anhydrides are capable of longer pot lives and lower exotherm during cure. These characteristics can be particularly important for land vehicle structures, as they can be quite large and complex in nature, requiring large quantities of resin and longer working times. A shortcoming of their low reactivity is the need to cure at higher temperatures to achieve acceptable cure rates, and as discussed this poses problems regarding increases to residual stresses. Another problem lies in their sensitivity to the rate of reaction as a function of cure temperature. Zukas [10] reported that beyond a given initial cure temperature changes in chemical reaction can occur between the inherent ether and ester linkages, resulting in reductions to the final glass transition temperature by 27%. Understanding the point at which this change in reaction occurs is critical for the processing of epoxy/anhydride

* Corresponding author.

E-mail address: s.agius@deakin.edu.au (S.L. Agius).

composites as it will dictate the maximum achievable cure temperature and subsequently fastest cycle time.

The widespread use of epoxy/anhydrides for composite transport structures may therefore be dependent on an improved understanding of the trade-off between the desire to use high cure temperatures to reduce cycle times, and the potential negative effects on the glass transition temperature and mechanical properties such as laminate shear strength. This study utilises a two-stage curing method where an epoxy/anhydride composite is cured at an initial isothermal injection cure temperature which is then followed by a fixed final free standing post cure. The effect of the initial cure temperature is examined through changes to initial cure times, with measurements of the final glass transition temperature, residual stress and laminate shear strength reported. This is achieved by firstly understanding how changes in the initial cure temperature affects the overall kinetics of the resin, such as the vitrification, maximum conversion and gelation times. These data are required for predicting the processing condition at which residual stresses develop within the composite as a consequence of the chemically and thermally induced volume changes, measured using gravimetric techniques. The use of representative volume element (RVE) and finite element (FE) techniques within ABAQUS then employ these changes in the resin volume to predict residual stresses in multi-directional composite laminates. The predicted residual stresses are then compared to experimental short beam shear (SBS) strengths for specimens cured at 85 °C, 105 °C and 135 °C. A digital imaging correlation (DIC) technique is applied to the short beam shear test samples to study the effect residual stress on the onset of failure through distinct changes in the shear strain field.

2. Materials and methods

2.1. Material and manufacturing procedure

The resin used in this study was a cyclohexylmethyl 3,4 epoxy cyclohexane carboxylate, bi-functional cycloaliphatic epoxy infusion resin with an anhydride hardener and imidazole catalyst. The carbon fibre was Toray's T620.

Multi-directional and unidirectional laminates were prepared using non-crimped fabric (NCF). This was achieved by stacking uniaxial plies according to the following two lay-ups $[-45/0/+45]_3 + [+45/90/-45]_3 + [-45/0/+45]_3$ and $[0]_{22}$, respectively. The aerial weight for the 0° and 90° layers within the multi-directional laminates was 500 gsm while the 45° layers was 200 gsm. The aerial weight for the 0° layers within the unidirectional layers was 365 gsm.

The initial curing for composite laminates was carried out using the Resin Transfer Moulding (RTM) process. Prior to resin injection, the multi-directional stack was placed within a closed mould with an internal volume of $492 \times 552 \times 7.5$ mm (W, L, t). The aerial weight of the fabric and nominal density of the fibre (1.79 g/cc) were used to calculate the number of plies required to achieve a fibre volume fraction of approximately 60%. The RTM tool was then inserted within a hydraulic press, compressed and heated to the desired initial cure temperature while the mould cavity was placed under vacuum. Once an adequate vacuum of less than 5 mbar had been reached, a leak down was conducted. A leak down less than 15 mbar in 15 min was considered acceptable for injection. The epoxy and hardener were thoroughly mixed and degassed for 15 min. The resin was then transferred to a pot that was pressurised to 5 bar absolute pressure prior to resin injection. After injection and complete infusion of the fibre, the outlet was clamped off and the hydrostatic pressure of the resin maintained throughout the remainder of the cure. The recommended cure

cycle temperature and time was 105 °C at 80 min. Additional isothermal cure schedules either side of the recommendation were also trialled (75–145 °C). Pertinent initial cure temperatures and cycle times were calculated using Differential Scanning Calorimetry (DSC) further discussed in Section 2.2. After initial cure, specimens were cooled to 60 °C at a rate of 1 °C/min. To achieve full degree of cure and the recommended final glass transition temperature of 200 °C, laminates were subjected to a free standing post cure within a convection oven. The post cure was achieved through a temperature ramp from 60 °C to 180 °C, an isothermal hold at 180 °C for 60 min and finally a cool down to 60 °C. The heat and cool down rate was 1 °C/min.

2.2. Determination of isothermal degree of cure, vitrification and gelation times

The isothermal degree of cure (DOC) of the resin was determined to calculate the pertinent cycle times required for degree of cure equilibrium. It should be noted that all DSC measurements were conducted using a TA Instruments Q200 Analyser with TA Instruments Tzero aluminium pans and lids for solid specimens and hermetic lids for liquid specimens. The degree of cure equilibrium is defined as the point in time when a maximum degree of cure is reached for a given isothermal cure temperature. The isothermal degree of cure was calculated using the following equation,

$$DOC = \frac{\Delta H_T - \Delta H_R}{\Delta H_T} \quad (1)$$

The total heat flow ΔH_T of the resin was measured by subjecting unreacted resin specimens to a dynamic scan from 30 °C to 300 °C at a rate of 10 °C/min. A linear integration of the area underneath the exothermic peak gives the total heat flow. An average of three scans resulted in a total heat flow of 385 ± 2.2 J/g.

Isothermal degree of cure was determined by subjecting unreacted resin specimens to isothermal scans within a DSC at 75 °C, 85 °C, 95 °C and 105 °C. Isothermal scans were held at temperature until degree of cure equilibrium had been reached as indicated by constant residual heat flow. The same specimen was then subjected to a repeat scan in order to produce a baseline correction. Once corrected the residual heat flow ΔH_R was integrated for each isothermal cure temperature and the isothermal degree of cure calculated using Eq. (1).

The point of vitrification has been defined as the point in the cure where residual stress begins to build within a composite [5]. Vitrification was determined by understanding the relationship between degree of cure and T_g . This was achieved by subjecting pre-cured resin specimens with varying degrees of cure to a modulated temperature ramp within a DSC. Details of their manufacture process are given in Section 2.3. The DSC modulated method involved equilibrating the specimens to 60 °C for 1 min then ramping 10 °C/min with a modulating amplitude of ± 1.6 °C every 60 s to a final temperature of 300 °C. The T_g for two specimens at each pre-cure temperature, 85 °C, 115 °C and 180 °C was measured, where the T_g was determined at the midpoint inflection of the heat flow curve. The calculated T_g was then plotted as a function of degree of cure equilibrium. The results suggest that the degree of cure at vitrification DOC_{vit} relates to the initial isothermal cure temperature $T_{g, initial\ cure}$, $DOC_{vit} = T_{g, initial\ cure} + 0.49$. This relationship can be used to predict the degree of cure for a given vitrification temperature or the isothermal cure temperature.

Finally gelation times were determined using a TA Instruments Discovery HR-3 rheometer using the parallel plate geometry. The 25 mm parallel plates were preheated to a given initial isothermal cure temperature. Unreacted premixed resin was then syringed between the plates and closed to achieve a gap of 500 μ m. The

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