Composites Science and Technology 71 (2011) 593–599

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: www.elsevier.com/locate/compscitech

Measuring the impregnation of an out-of-autoclave prepreg by micro-CT

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ARTICLE INFO

Article history:
Received 29 June 2010
Received in revised form 22 November 2010
Accepted 2 December 2010
Available online 9 December 2010

Keywords:
Out-of-autoclave
A. Polymer-matrix composites
B. Porosity/voids

ABSTRACT

Resin flow into dry reinforcement regions is the main microstructural change during the processing of out-of-autoclave prepregs and influences air evacuation and void suppression. Such impregnation flow was investigated experimentally during the processing of a second-generation out-of-autoclave prepreg. First, laminates were partially processed to different stages of a simple cure cycle. Then, samples from each laminate were scanned using X-ray microtomography (micro-CT) to obtain 3D microstructural data. This data was used to investigate the initial microstructure of the material and measure the extent of impregnation at each processing stage, the rate of impregnation, and the evolution of macro-porosity within the material.

1. Introduction

A new generation of out-of-autoclave prepregs aims to offer performance and quality comparable to autoclaved materials through vacuum-bag-only cure in an oven [1,2]. Since the maximum applied compaction pressure during such processing is 1 atm. and some load is carried by the fibre bed, the resultant resin pressure may not be sufficient to suppress voids [3]. Thus, the evacuation of entrapped air, vaporized moisture or other volatiles prior to resin gelation becomes essential for low porosity parts. Initially, out-of-autoclave prepregs feature dry, relatively permeable areas that allow gas evacuation when vacuum is applied at the beginning of the cure cycle. These areas may consist of macro-porosity between plies and around the reinforcement architecture, or micro-porosity inside the tows, between individual fibres. During processing, these spaces are progressively infiltrated by resin to produce a uniform, void-free structure [1,4]. The dynamics of this impregnation influence both gas evacuation and final part porosity, and require thorough understanding.

1.1. Background

The behaviour of fibre/thermoset-matrix systems has been the subject of considerable research and a large amount of experimental and modelling knowledge has been compiled [5,6]. Prepregs are processed by consolidation: the application of pressure and heat compresses the fibre bed, induces resin flow and collapses voids. Generally speaking, when a consolidation pressure is applied and a prepreg laminate compacts, two coupled flows may be present: flow between plies into local macro-voids and resin infiltration into micro-porous dry fibre tows, or tow impregnation [7]. For net resin content, no-bleed out-of-autoclave prepregs, neither flow is likely to involve large-scale resin transport of the type found in liquid moulding processes.

Specific literature on quantifying flows in out-of-autoclave processing is limited. Lucas et al. suggested that the impregnation level of a vacuum-bag-only prepreg may be qualitatively determined by the prepreg's appearance when deformed [4]. Thomas et al. provided an overview of resin flow measurement techniques used in liquid moulding, including visual flow front tracking, fibre-optic sensing, microstructural analysis, and pressure field monitoring [8]. Due to small scales and internal, localized flows, many of these techniques are difficult to apply to prepregs. In the same work, Thomas et al. successfully measured the transverse impregnation rate of partially infused carbon fibre layers using ultrasound. Meanwhile, Tavares et al. [9] considered the impregnation dynamics of a semi-preg material in the context of the evolution of its permeability, while Wysocki et al. [10] and Louis et al. [11] observed the microstructures of partially impregnated prepregs using optical and sweep electron microscopy. Overall, there is a continuing need for experimental methods and data that clarify out-of-autoclave prepregs' initial microstructure and the key physical phenomena involved in its evolution during processing.

1.1.1. X-ray microtomography

X-ray microtomography (or micro-CT) is an experimental technique used to obtain three-dimensional microstructural information. A sample is placed on a rotating stage between an X-ray generator and a detector. For a specific exposure time, X-rays radiate from the generator, penetrate the sample, attenuate proportionally to the material's density, and reach the detector to form

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doi:10.1016/j.compscitech.2010.12.009

a shadow projection containing information about the microstructure. The sample is then rotated a fractional amount, and a second shadow projection is obtained from a slightly offset perspective. An 180° rotation generates a sufficient set of such projections, which are reconstructed into parallel micro-slices using a mathematical algorithm [12]. Micro-CT has recently been used in composites research to characterize woven fibre architectures and to inspect damage [13–17].

1.2. Objectives

This study aimed to use micro-CT to investigate the impregnation behaviour of an out-of-autoclave prepreg. The proposed steps consisted of processing laminates to different stages of a simple cure cycle, scanning samples from these laminates using micro-CT, and using the resulting data to quantify the evolution of impregnation and porosity due to resin flow.

2. Materials

The out-of-autoclave prepreg chosen for this study was manufactured by the Advanced Composites Group (ACG). The reinforcement consisted of a 5-harness satin weave of 6 K carbon fibre tows. The matrix was ACG's MTM45-1, a high-performance toughened epoxy resin optimized for low temperature vacuum-bag-only cure, with cycles ranging from 20 h at 80 °C to 2 h at 130 °C. The prepreg, partially impregnated by resin film on both sides, had a 36% by weight net resin content and an areal density of 375 g/m^2.

The cure kinetics and viscosity of the MTM45–1 resin were previously characterized by Kratz, using experiments and models outlined and described by Khoun et al. for the characterization of epoxy resins [18,19]. The evolution of the degree of cure (α) and viscosity (μ) were measured experimentally using differential scanning calorimetry and parallel plate rheometry, respectively, and fit to semi-empirical equations that allow their prediction for any cure cycle. The cure kinetics model is given in Eq. (1) and the viscosity model in Eq. (2). The numerical values of each model's parameters are listed in Table 1. Both models show good agreement with the experimental data.

$$\frac{d\alpha}{dt} = A\ \exp\left(\frac{-E_A}{RT}\right)\frac{\alpha^m(1-\alpha)^n}{1+\exp\left(C(\alpha-(\alpha_{C0}+\alpha_{CT}T))\right)} \tag{1}$$

$$\mu = A_\mu\ \exp\left(\frac{E_\mu}{RT}\right)\left[\frac{\alpha_g}{\alpha_g-\alpha}\right]^{(A+B\alpha)} \tag{2}$$

Table 1
Parameters used in the MTM45-1 cure kinetics and viscosity models.

Cure kinetics model	Viscosity model
E_A = 60,628 J/mol	R = 8.314 J/K mol
$A = 2.28 \times 10^4\ s^{-1}$	E_μ = 79,500 J/mol, 20 °C < T < 100 °C
	E_μ = 42,000 J/mol, T > 100 °C
m = 0.526	$A_\mu = 1.8 \times 10^{-10}$ Pa s, 20 °C < T < 100 °C
	$A_\mu = 3.0 \times 10^{-5}$ Pa s, T > 100 °C
n = 0.946	A = 5
C = 54.26	B = −5
α_{C0} = −1.076	α_g = 0.40
$\alpha_{CT} = 4.44 \times 10^{-3}\ K^{-1}$	

3. Procedures

3.1. Laminate preparation and partial processing

Ten centimeter by 10 cm laminates were prepared. Most laminates consisted of four plies $[(0°/90°)_2]_s$; laminates 8 and 10 had 8 plies ($[(0°/90°)_4]_s$) due to early test trials. No differences were expected or seen in the results due to this variation. Each laminate was laid up by placing the prepreg plies on non-perforated release film on a flat aluminum tool plate. Another layer of non-perforated release film was laid on top of the stack to prevent through-thickness resin bleed. The laminate perimeter was surrounded by edge breathing dams consisting of sealant tape wrapped in fibreglass cloth, to provide an in-plane gas evacuation path, avoid edge pinching and prevent resin bleed. Two layers of breather covered the entire arrangement, and were carefully placed in contact with the edge breathing dams. A vacuum valve and the bag completed the lay-up.

The process cycle for the partially cured laminates consisted of an hour-long vacuum hold at 25 °C, a 2 °C/min (±0.5 °C) ramp to 85 °C and a 90 min isothermal dwell at that temperature for a total process time of 180 min. The room-temperature hold was included to remove entrapped air as well as to investigate the presence of room-temperature resin flow under applied compaction. The low 85 °C temperature of the heated dwell was chosen to maintain a relatively high resin viscosity and, consequently, a relatively low rate of flow.

The laminates were partially processed to different points within the cycle, as shown in Table 2. Once each laminate reached its desired processing stage, it was removed from the oven and the vacuum source and quickly cooled in a freezer to stop any resin flow. The bag temperature and the corresponding skin temperature (measured from an additional laminate processed in identical conditions), were tracked using thermocouples.

3.2. Micro-CT

3.2.1. Scanning

Two samples (A and B) nominally 15 mm × 15 mm were cut from the center of each of the 10 laminates. In addition, one sample (C) was cut from laminates 5 and 10 each for additional high-resolution scans. Prior to scanning, each sample was mounted in a custom grip consisting of a Styrofoam block with a cut notch on one side and a machined hole on the other. The notch held the sample in place with minimal contact and pressure while the hole allowed easy but tight placement onto a copper mounting rod. The Styrofoam's low density and X-ray attenuation allowed the entire sample, including the section being gripped, to be scanned.

Table 3
Micro-CT scan parameters.

Parameter	Value
Filter	None
Resolution (samples A and B)	7 μm/pixel
Resolution (sample C)	2 μm/pixel
Image size	4000 × 2096 pixels
X-ray voltage	64 kV
X-ray intensity	154 μA

Table 2
List of partially processed laminates.

Laminate	1	2	3	4	5	6	7	8	9	10
Layers	4	4	4	4	4	4	4	8	4	8
Process time (min)	0	15	30	45	60	80	93	104	110	180
Process conditions	None	Room temperature + vacuum				Elevated temperature + vacuum				