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Effect of Healing Agent Concentration on Viscosity of Epoxythermoplastic Blend and Void Distribution of Composite Laminates

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Abstract: Fabrication of self-healing Carbon Fibre Fabric Reinforced Polymer (CFFRP) composite laminates were carried out by hand lay-up and density was measured via water buoyancy. The results obtained from two methods were consistent showing laminates with a higher concentration of healing agent had greater void contents. Void content was also found to be higher near the edges of laminates compared to the central areas. High viscosity of the polymer blends due to the addition of thermoplastic healing agent is suspected to be the main reason for the void distribution pattern; air bubbles entrapped in laminates during fabrication are difficult to move in viscous resin during vacuum-degassing such that eventually the movement could be halted and voids accumulate near the laminate edges. The composite laminates fabricated in this study support the potential suitability for industrial applications.

Key words: Composite, smart materials, voids

INTRODUCTION

The development of smart materials particularly composites with the capability of self-healing has been a major research focus in recent years. This study can provide a potential alternative to facilitate repair of internal crack and delamination in composites which are currently difficult to detect and repair (Talreja, 1989). To confer healing potential in composites, particularly in fibre-reinforced polymers, a healing material (agent) with or without an additional containing mechanism can be integrated into the composite system (Kessler, 2007; Zhang and Rong, 2011; Van der Zwaag and Schmets, 2007). Healing can either be activated as a response to damage (Brown *et al*., 2004; Pang and Bond, 2005) or by stimuli such as heat (Hayes *et al*., 2007a, b). Healing is expected to restore composite capability in order to extend its service life.

This study presents a research on hand lay-up fabrication of self-healing composite laminates with embedded thermoplastic as a healing material. The produced laminates were assessed with respect to void content using the Archimedes' principle.

MATERIALS AND METHODS

Composite materials and fabrication: The main materials used to fabricate the composites included diglycidyl ether of bisphenol A epoxy resin Epikote-828 as the composite matrix, thermoplastic poly (bisphenol A-co-epichlorohydrin) (PBE) as a healing agent and unidirectional carbon fibre fabric Gurit UT-C200 as the composite reinforcement. Epikote-828 and PBE were purchased from Fraser Brown and Stratmore Limited and Sigma-Aldrich, respectively, in New Zealand. Gurit UT-C200 with a nominal area weight of 200 g m^{-2} was purchased from Gurit Ltd. (Australia), whilst Porcher Style-3085 was from Porcher Industries (China), having a nominal area weight of 196 g m^{-2}. All materials were used as received.

The fabrication of thermoset composite laminates embedded with thermoplastic poly (bisphenol A-co-epichlorohydrin) (PBE) as a healing agent began with blending a desired amount of PBE in the form of 1-3 mm droplets in liquid epoxy diglycidyl ether of bisphenol A (DGEBA or Epikote-828) just above the PBE melting temperature (76°C) at 80°C on top of a hot plate until no sign of undissolved droplets was observed. The entire

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process could take up to over 24 h, depending on the amount of PBE being dissolved. During lamination, each polymer blend was heated to a temperature sufficiently high to lower the viscosity for impregnation of fibre. All blends were mixed with curing agent and were heated and stirred throughout the lamination. The lay-ups were placed on top of a hot plate to ensure heat was continuously applied to ensure they remained in workable viscosity. For each laminate, six layers of pre-cut fabric sheets were wetted with mixture and stacked on top of each other. Precaution was taken to ensure the fabrics stayed intact by applying consistent pressure with a squeegee. Stacked laminates were sandwiched with Teflon cloth and flat glass panels before placing in a vacuum oven for degassing and curing. Three groups of laminates with different healing agent concentrations (0, 10, 20 wt%) were fabricated. All laminates were stored and conditioned in a humidity chamber ready for testing according to standard test procedures (ASTM D618-05, 2000).

Density and void content measurement: Once PBE was dissolved in epoxy, the Epikote-828/PBE polymer blend became increasingly viscous. Brookfield DV-II Digital Viscometer was used to measure the viscosity of Epikote-828/PBE blend at different heating levels.

Void content of composite laminates was determined according to ASTM D2734: Standard test methods for void content of reinforced plastics (ASTM D2734-94, 2003). The void content (V) was calculated according to:

$$V = 100\left(1 - \rho_m / \rho_t\right) \tag{1}$$

where, ρ_m is the measured density of composite laminates and ρ_t is the theoretical density calculated as follows:

$$\rho_t = 100 / \left(M_r / \rho_r + M_f / \rho_f\right) \tag{2}$$

where, M_r and M_f are the weight percentages of resin and fibre content, respectively in the composite, whilst ρ_r and ρ_f are the densities of the resin matrix and reinforcing fibre, respectively. Resin content and fibre content were determined by matrix digestion using nitric acid (ASTM D3171: Standard test methods for constituent content of composite materials (ASTM D3171-11, 2011)). The density of laminates, their respective polymer matrices and reinforcing fibres were measured according to ASTM D792: Standard test methods for density and specific gravity (relative density) of plastics by displacement (ASTM D792-13, 2008) and ASTM D3800: Standard test method for density of high-modulus fibers (ASTM D3800-99, 2010). Figure 1 shows apparatus for measuring the density of test samples. Each sample (ASTM D792 and ASTM D3800 for sample details) was

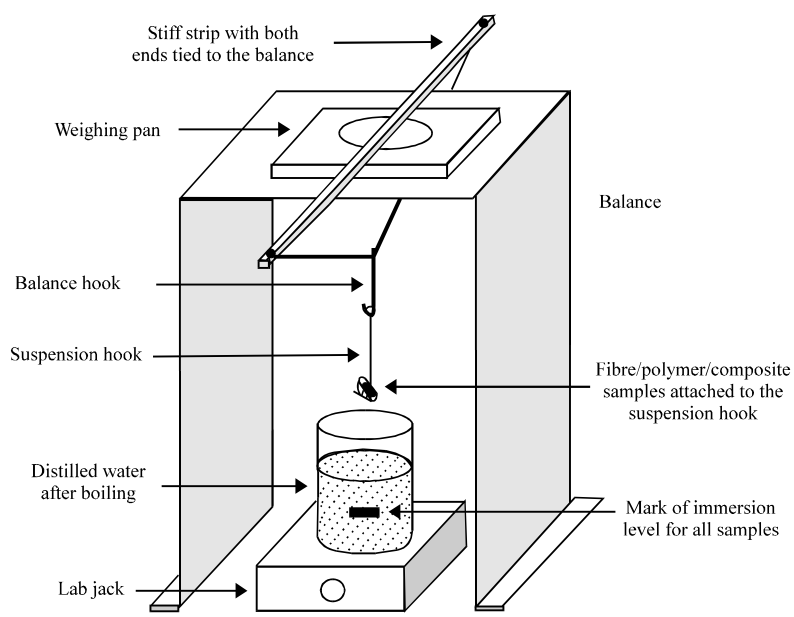

Fig. 1: Apparatus for measuring densities of samples using Archimedes' principle

attached to the suspension hook and weighed in air. A beaker filled with degassed distilled water was then raised by a lab jack such that the sample was thoroughly immersed in the water. All samples were immersed at the same level for consistency. Air bubbles entrapped on sample surfaces were removed by removal of samples from the water and reimmersion for a few cycles to increase wetting. Any bubbles left visually detected were removed by a thin wire before the final weight of the sample in water was measured. The sample volume was obtained by dividing the weight difference of the sample in air and in water (buoyancy force) with water density and hence the sample density (equal to sample weight in air divided by the sample volume) was calculated.

RESULTS AND DISCUSSION

Viscosity of Epikote-828/PBE blend: At 80°C, all polymer blends with different concentrations of PBE up to 20 wt% eventually showed no sign of PBE after different intervals of stirring. Viscosity analysis using a Brookfield DV-II Digital Viscometer showed that increasing concentration of PBE increased the viscosity of polymer blends (Table 1); viscosity of pure epoxy was increased from 0.07-3.90 Pa.sec when 10 wt%. The PBE was blended-in, whereas viscosity of polymer blends with 20 wt%. The PBE (Epikote-828/20 wt% PBE) was 8.32 Pa.sec, twice as viscous as Epikote-828/10 PBE. During trial fabrication, it was found that polymer blends with viscosity greater than 5 Pa.sec were not workable for fabrication by hand lay-up. It was therefore essential to increase the temperature of such polymer blends during fabrication.

Assessment of void distribution in laminates by density measurement: The average content ratio of fibre/resin was found to be 58.97/41.03 (±2.20 wt%) for the UD-composite laminates. From the Archimedes' buoyancy method, density of carbon fibres from the Gurit UT-C200 UD-fabric was found to be 1718.0±20.7 kg m^{-3}. Densities for resin matrices were found to be 1195.5±2.8 kg m^{-3} for Epikote-828/without PBE, 1193.9±1.7 kg m^{-3} for Epikote-828/10 PBE and 1194.4±3.9 kg m^{-3} for Epikote-828/20 PBE; this data was used to calculate the theoretical density of laminates according to Eq. 2. The results were listed in Table 2.

A summary of mean void content data of all different groups of laminates were tabulated in Table 3, showing that void content distribution trends for respective laminate groups were unanimous. It can be seen that higher concentrations of PBE resulted in greater percentages of void content. Also, when the data of samples obtained from central area and near the edges of laminates were assessed separately, the near-edge samples give a higher void content than the centre samples. The reason of void content being higher in outer areas of laminates may be due to entrapped air bubbles moving outwards during vacuum-degassing prevented from total evacuation by the viscous resins and accumulating at the edges of the laminates.

Table 1: Viscosity data of polymer blends with different concentrations of healing agent PBE

Temperature of mixture (°C)	Mean viscosity (Pa.sec) of polymer blend with PBE concentrations (%wt): 0	5	10	15	20
80	0.07	0.97	3.90	4.95	8.32
90	0.06	0.56	2.21	2.67	5.67
100	0.04	0.30	1.19	1.66	4.27

Table 2: Density data of fibres and resin matrices, along with the corresponding calculated theoretical density of unidirectional (UD) Gurit UT-C200 composite laminates

Sample materials	Mean density (kg m^{-3})
Reinforcing fibres	
UD Gurit UT-C200	1700.9
Resin matrices	
Epikote-828/without PBE	1195.5
Epikote-828/10 wt% PBE	1193.9
Epikote-828/20 wt% PBE	1194.4
***Composite laminates**	
(UD) Epikote-828/without PBE	1449.5
(UD) Epikote-828/10 wt% PBE	1448.5
(UD) Epikote-828/20 wt% PBE	1448.8

*Theoretical density data of composite laminates calculated according to Eq. 2

Table 3: A summary of mean void content data of UD-laminates

	Comparison of void content distribution in edges and central area of laminates					
	Epikote-828/without PBE		Epikote-828/10 wt% PBE		Epikote-828/20 wt% PBE	
Composite laminates	Central	Edges	Central	Edges	Central	Edges
1	1.30	1.57	2.24	3.75	2.56	4.71
2	1.29	1.64	2.42	3.76	2.96	4.07
3	1.06	1.76	2.25	3.51	3.19	5.59
Average	1.22	1.66	2.30	3.67	2.90	4.79
*Stdev	0.14	0.10	0.10	0.14	0.32	0.76

*Stdev: Standard deviation

CONCLUSION

Self-healing composites using thermoplastic PBE as healing agent were fabricated by hand lay-up. The quality of laminates was assessed by examining the void content and distribution using density measurement based on the Archimedes' principle. It was showed that composite laminates containing healing agent PBE exhibit greater void content at near laminate edges than at more central areas. This may be caused by entrapped air bubbles moving outwards during vacuum-degassing slowed by the viscous resins and accumulating near laminate edges. In general, applications in advanced dynamic aerospace structures require the void content of composite laminates not more than 1.5%. Laminates containing void content up to 6% as fabricated in this research are often acceptable for industrial applications such as ground vehicle components and rudimentary composite items. However, with higher levels of fabrication methods such as the autoclave process used in the aerospace industry, the void content of self-healing laminates may be reduced to the tolerable level and hence diminish the influence of healing agent on quality of composites to the minimal.

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