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Manufacturing and characterization of carbon fibre/epoxy composite prepregs containing carbon nanotubes

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ABSTRACT

Prepregs of carbon fibre-reinforced polymer (CFRP) composites containing carbon nanotubes (CNTs) have been successfully produced based on a solventless prepregging process. This work provides one of the most comprehensive evaluation to date of the processing and cure parameters affected by the incorporation of CNTs in CFRP prepreg manufacturing. The results confirmed that high-speed shear mixing and functionalisation of CNT are effective in lowering the viscosity of the CNT-epoxy nanocomposites. Based on the findings obtained from the rheological studies, suitable processing parameters are chosen for lab-scale production of CFRP composite prepregs. The effects of surfactant-treated CNTs on cure behaviour of epoxy and CFRP prepreg are evaluated. The catalytic activity of CNTs is negligible for a 0.5 wt% filler content whereas it becomes prominent when the CNT content is increased to 1.0 wt%. The degree of cure is lower for the prepregs containing 0.5–1.0 wt% CNTs than those without or containing a lower CNT content.

1. Introduction

Carbon nanotubes (CNTs) possess exceptional mechanical, electrical and thermal properties, making them ideal fillers for polymer nanocomposites for various structural and functional applications [1,2]. During the last two decades, various techniques such as in situ polymerization [3,4], solution casting [5,6], polymer crystallization [7] and melt processing [6] have been used for the preparation of CNT-polymer nanocomposites to explore these unique properties of CNTs. The factors influencing the properties of nanocomposites such as CNT structure [8,9], dispersion [10,11], alignment [12,13] and interfacial adhesion between CNTs and matrices have been extensively studied [14]. The development of nanocomposites with much improved mechanical and functional properties have been reported [4,10,15].

Incorporation of CNTs into a polymer matrix along with long fibre reinforcements to produce hybrid composites has attracted significant attention in recent years. To impregnate the long fibres or fabrics with a CNT-resin mixture, resin transfer moulding (RTM), vacuum-assisted resin transfer moulding (VARTM) [16–20] or wet hand lay-up process [21–24] were commonly used. In all these processes, the resin integration and consolidation of the composites were achieved in one single process. A more systematic route of mass-producing fibre reinforced composites (FRPs) uses prepregs in which fibres are pre-impregnated with a resin in a latent or beta state. Fabrication of FRP components using prepregs has unmatched benefits that include readiness to use with no further treatment, ease of handling, uniform fibre alignment, accurate control of the resin content, ability to conform to intricate shapes, optimum polymeric composites integration and very low void contents in the final products. As a consequence, the highest material qualities and performance characteristics are achieved by prepreg-based manufacturing processes. Over the past years, FRP prepregs have been successfully used for manufacturing high-performance composites for structural applications such as airplane components, defence, marine, offshore, mass transit and speciality sporting goods.

Very few studies have so far been reported on the fabrication of CFRP hybrid composites via manufacturing of prepregs [25], which were modified by cup-stacked carbon nanofibres, instead of CNTs that are much more difficult to process because of the issues associated with agglomeration and lack of interfacial interactions with polymers. None of the previous studies, nevertheless, have given due attention to the influence of adding CNTs on the prepregging process. It is necessary to understand the effects of adding CNTs on viscosity of the matrix material, which will largely affect the impregnation of fibres and prepreg processing conditions. The change in cure kinetics might also influence the prepreg storage life and the characteristics of the fabricated composites.

This paper is part of a larger project aimed at developing CFRP composites containing CNTs for speciality applications. Our

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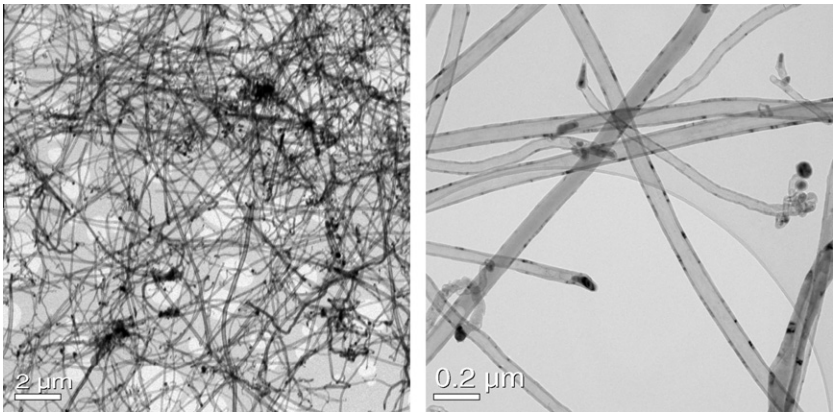

Fig. 1. TEM micrographs showing morphologies of CNTs.

previous studies have focused on functionalisation and uniform dispersion of CNTs in polymer, including epoxy resins [4,26–29]. In this study, a special emphasis has been placed on studying the effects of resin type and CNT content on various parameters in a solventless prepregging process and on the changes in curing behaviour of epoxy and prepreg.

2. Experimental

2.1. Materials and preparation of CNT modified matrices

The multiwalled CNTs (NK-50 supplied by Nanokarbon, Korea) used in this work were of bamboo-like structure and were produced by a vapour grown method. The outer diameters ranged between 40 and 60 nm and the lengths were about 20 m. Typical TEM images of these CNTs are presented in Fig. 1. Both pristine and functionalised CNTs were employed in this work. CNTs were functionalised using a combination of processes chosen based on extensive studies reported previously [26–29]. The CNTs were first subjected to oxidation in a UV/O_3 chamber (Jelight 144AX-220) for 30 min, followed by treatment with a surfactant. A nonionic surfactant, polyoxyethylene phenyl ether (Triton X-100, supplied by VWR International, UK) with the critical micelle concentrations (CMC) value of 0.2 mM at 25 °C was used to treat the CNTs to improve dispersion in the resin. The procedure adopted for the treatment was basically similar to that reported previously [29]. A desired amount of CNT was dispersed in acetone containing 10CMC of surfactant, equivalent to a Triton weight to acetone volume ratio of approximately 12.5 mg/1000 ml. The mixture was subjected to sonication in a bath (Branson 150) for 60 min.

Two different epoxy resins were used to study the rheological behaviours: one (EP1) was made from Epon 828 (supplied by Shell), a DGEBA epoxy, and the other (EP2) was Araldite LY556 (supplied by Huntsman), a reaction product of bisphenol A (epichlorohydrin). The carbon fibre roving (Pyrofil TR 30S, supplied by Mitsubishi Rayon, Japan) with a filament count of 6 K was used as the main reinforcement. The Araldite LY556 resin system was eventually selected for producing prepregs, which were composed of Araldite LY556, Aradur 5021 and hardener XB 3403 (all supplied by Huntsman) in the ratio of 100:25:12 parts by weight. A desired amount of epoxy (EP1 or EP2) was added into the suspension of treated CNT/acetone solution to obtain CNT/epoxy composite mixtures with varying CNT contents. The initial dispersion was achieved by ultrasonication at 60 °C for 30 min, followed by degassing in a vacuum oven at 70 °C overnight to ensure complete removal of acetone. The dispersion of CNTs was further improved by mixing for 30 min using a Ross high speed shear mixer [10]. Two different speeds, 3000 and 4000 rpm, were used to study their effects on dispersion and viscosity of the suspensions.

2.2. Rheological studies and cure behaviour

Rheological studies were carried out to select a suitable matrix material and identify optimal prepregging parameters. Four types of resin blend containing 0 wt%, 0.5 wt%, 0.7 wt%, and 1.0 wt% CNT were prepared with the epoxy resins. The viscosity changes were measured on an oscillatory rheometer (CSL500, TA instrument) using a parallel plate geometry (with 40 mm flat plate and 300 m gap). The effects of shear mixing speed, type of epoxy and CNT functionalisation were evaluated on the viscosity changes. Temperature sweep was also carried out for the finally chosen resin system to determine the most suitable temperature for the prepreg process; the viscosity measurements were taken over a temperature range from room temperature to 50 °C. The effects of CNT on curing behaviours of epoxy resin and CFRP prepregs were evaluated using the dynamic differential scanning calorimetry (Q1000, TA instrument) at a ramping rate of 10 °C/min. The glass transition temperatures, T_g, of CFRP prepregs were measured from the DSC thermograms of the specimens obtained from the single layers of prepregs cured at 120 °C for 2 h followed by post-cure at 130 °C for 1 h. A minimum of five specimens were tested for each set of conditions.

2.3. Prepreg process of CNT-CFRP

CNT-CFRP hybrid composite prepregs were prepared on a lab-scale prepregger (Model 40 Research Tool Corp., USA). A photograph and a schematic of the prepregger system are presented in Fig. 2. Based on the outcome of the rheological studies, the temperature of the resin bath was set at 37 °C, which was optimised to maintain the viscosity of the CNT-resin mixture within the required limits (1.5–2.4 Pa s) and to achieve good wetting of the carbon fibre tow. The flattening pins and dies were also maintained at the same temperature. Various combinations of line speed and exit die gap were tried to control the resin content in the prepreg. The resin contents of the prepregs were determined by matrix extraction using acetone as a solvent according to the specification, ASTM standard C613. The prepreg thickness was determined using a small piece of prepreg (5×5 cm^2), which was pressed between two parallel metal plates at 2 N before taking measurements. The overall quality of the prepregs was determined by observing their polished cross sections under a scanning electron microscope (SEM, Jeol 6390F).