Composites Science and Technology 71 (2011) 717–723

Contents lists available at ScienceDirect

Composites Science and Technology

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

Interlaminar properties of carbon fiber composites with halloysite nanotube-toughened epoxy matrix

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ARTICLE INFO

Article history:
Received 22 August 2010
Received in revised form 14 January 2011
Accepted 25 January 2011
Available online 1 February 2011

Keywords:
Halloysite nanotube
A. Nanocomposites
A. Hybrid composites
B. Fracture toughness

ABSTRACT

Halloysite nanotubes (HNTs), which are geometrically similar to multi-walled carbon nanotubes, can improve the impact strength of epoxy substantially, according to our previous work [1]. Using a HNT-toughened epoxy as the matrix, a set of hybrid composites was prepared with carbon fiber-woven fabrics. The interlaminar properties of the composites were investigated by a short-beam shear test, a double-cantilever-beam test and an end-notched flexure test. The results showed that the addition of HNTs to the composites improved the interlaminar shear strength and the fracture resistance under Mode I and Mode II loadings greatly. The morphological study of the hybrid composites revealed that HNTs were non-uniformly dispersed in the epoxy matrix, forming a unique microstructure with a large number of HNT-rich composite particles enveloped by a continuous epoxy-rich phase. A study of the fracture mechanism uncovered the important role of this special morphology during the fracturing of the hybrid composites.

1. Introduction

Carbon fiber-reinforced epoxy (EP/CF) composites have been widely used in many areas, including aerospace, automobile, marine, military, etc., due to their unique properties, such as high strength, high modulus and lightweight. However, the strength in the through-thickness direction is a limiting design factor in conventional composites because there are no fibers oriented in the thickness direction to sustain transverse loads. The low strength in the through-thickness direction generally leads to interlaminar failures, such as delamination. To improve the interlaminar fracture toughness, a considerable amount of research has been conducted. Examples of such attempts include the use of Z-pins to connect the laminates, and extending the fibers through the thickness direction by weaving, knitting, braiding or stitching [2–6]. However, these techniques are labor-intensive and require special fabrication processes, which greatly increase the manufacturing cost [5]. Moreover, the complex combination of materials makes it difficult to accurately predict the in-plane mechanical properties, i.e., the tensile, compressive and flexural properties, for a particular stitched composite. For instance, stitching may improve the in-plane properties, or it may leave the properties un-changed; in some instances it may even seriously decrease the in-plane properties [2].

With improvements in nanocomposite technology, many researchers attempted to improve the interlaminar properties of fiber-reinforced composites using nanofillers [7–12]. The incorporation of alumina nanofillers into EP/CF composites resulted in both higher interlaminar shear strength (ILSS) and fracture toughness [12]. Gojny et al. [7] found that the addition of 0.3 wt% double-walled carbon nanotubes (CNTs) to fiber-reinforced epoxy composites increased the ILSS by 20%. By growing aligned CNTs on the surface of SiC fibers, the Mode I (G_{Ic}) and Mode II (G_{IIc}) interlaminar fracture toughness of the 3D composites were improved by 348% and 54%, respectively [8]. Siddiqui et al. [10] also increased the value of G_{Ic} by 60% by adding 3 wt% organoclay to an epoxy. All these studies have shown just how promising the applications of nanofillers are in fiber-reinforced composites. Moreover, the introduction of nanofillers does not increase the weight of the components manufactured with EP/CF composites.

Halloysite is a fine clay mineral consisting of tubular particles with a multi-layered wall structure. In recent years, there have been great interests in the application of HNTs in polymer materials [13–15]. Our previous work [1,16] has shown that HNTs, as low-cost nanotubes, can improve the mechanical properties of an epoxy significantly. The addition of 2.3 wt% HNTs to the epoxy increased its Charpy impact strength by about four times with slight improvements in the flexural modulus and strength. The underlying toughening mechanisms were identified as massive bridging, pull-

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doi:10.1016/j.compscitech.2011.01.018

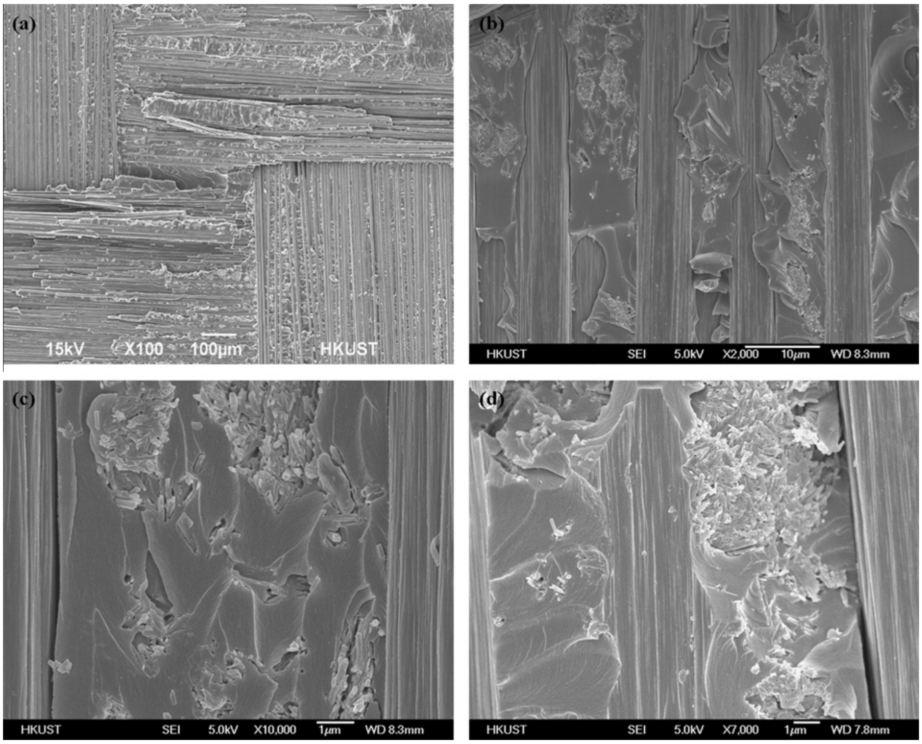

Fig. 1. SEM micrographs of EP/HNT/CF composites with 3 wt% HNTs.

out, and breaking of nanotubes, micro-cracking, as well as main-crack deflection. Considering that the HNT crack-bridging capability and the low damage resistance of conventional EP/CF composites are largely a result of the propagation of internal defects (e.g., micro-cracks) under external loadings, we used the EP/HNT nanocomposite as the matrix in the fabrication of the CF composite in the present study. We anticipated that the EP/HNT/CF hybrid composites would benefit from the high impact toughness due to the presence of the HNTs, leading to a new class of CF composites. The microstructure of the EP/HNT/CF hybrid composites was examined using a scanning electron microscope (SEM). The mechanical properties and the failure mechanisms of the hybrid composites were studied.

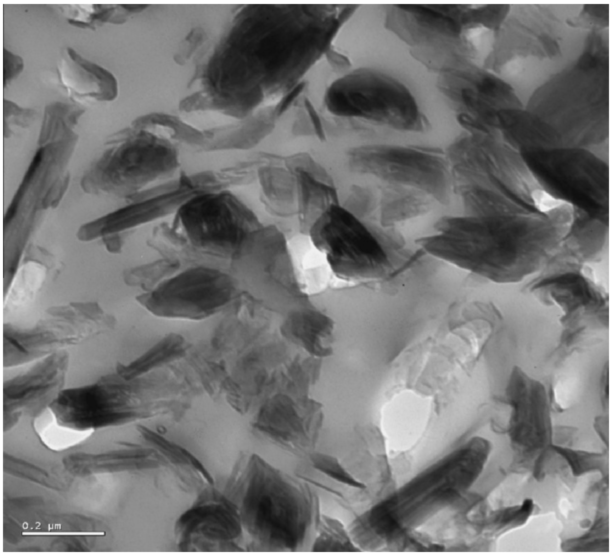

Fig. 2. TEM micrograph showing the HNT-rich region.

2. Experimental work

2.1. Fabrication of composites

The EP/HNT/CF hybrid composites were prepared with carbon fiber-woven fabrics and a HNT-filled epoxy by the hand lay-up process. Plain woven carbon fibers (TI3101 supplied by Taiwan Electrical Insulators Co.) with a unit weight of 200 g/m^2 were used as the major reinforcement. The materials and processing conditions for the EP/HNT nanocomposite solution were the same as those reported previously [1]. The nanocomposite was prepared using halloysite nanotubes (supplied by Imerys Tableware New Zealand Limited), and EPON Resin 828 (Bisphenol A, supplied by Resolution Performance Products) with a curing agent, 4, 4′-methylene dianiline (MDA, Aldrich), at a 100/27 weight ratio. A certain amount of halloysite nanotubes was first dispersed in acetone and

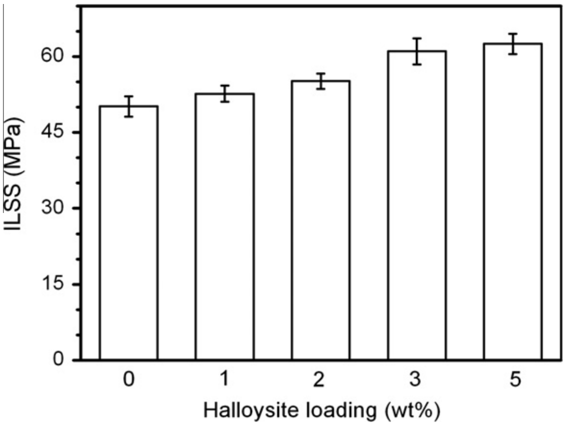

Fig. 3. Interlaminar shear strength of EP/CF composites with different HNT contents.