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Flexural fatigue behavior of synthesized graphene/carbon-nanofiber/epoxy hybrid nanocomposites

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ABSTRACT

In the present research, effects of adding a combination of synthesized graphene nanosheets and carbon nanofibers (CNFs) on the flexural fatigue behavior of epoxy polymer have been investigated. Graphene nanosheets are synthesized based on a changing magnetic field. The flexural bending fatigue life of 0.5 wt.% of graphene/CNF/epoxy hybrid nanocomposites has been considered at room temperature. The samples were subjected to different displacement amplitudes fatigue loadings. Due to the addition of hybrid nanoparticles, a remarkable improvement in fatigue life of epoxy resin was observed in comparison with results obtained by adding 0.25 wt.% graphene or 0.25 wt.% CNF into the resin. Experimental observations show that at a strength ratio equal to 43% by using 0.5 wt.% of hybrid nanoparticles; 37.3-fold improvement in flexural bending fatigue life of the neat epoxy was observed. While, enhancement of adding only graphene or CNF was 27.4 and 24-fold, respectively.

1. Introduction

The flexural fatigue behavior of composites and nanocomposites has been carried out by many researchers [1–4]. For composites under displacement-controlled condition, Paepegem and Degrieck developed an experimental setup for bending fatigue loading [1]. They adopted a residual stiffness model which describes the fatigue damage behavior of the composite material [2]. Also, Paepegem and Degrieck [3] used a finite element approach for composites fatigue life prediction. El Mahi et al. [4] studied the flexural fatigue behavior of the sandwich composite materials using three-point bend test and the derived approach permitted to predict the fatigue life of the sandwich composite materials while avoiding the large number of experiments that would normally required in fatigue testing. A survey in the available literature reveals that the addition of nanoparticles can improve the fatigue behavior of composites under displacement control loading and has been carried out by many researchers [5–8]. Ramkumar and Gnanamoorthy [5] studied the stiffness and flexural fatigue life improvements of polymer matrix reinforced nanocomposites with nanoclay. They described the effect of adding nanoclay fillers on the flexural fatigue response of Polyamide-6 (PA6). Rajeesh et al. [6] considered the influence of humidity on the flexural fatigue behavior of commercial grade polyamide-6 granules and hectorite clay nanocomposites. Timmaraju et al. [7] considered the influence of the environment on the flexural fatigue behavior of polyamide 66/hectorite nanocomposites. They also found the effect of initial imbibed moisture content on the flexural fatigue behavior of polyamide 66/hectorite nanocomposites conducted under deflection control method using a custom-built, table-top flexural fatigue test rig at a laboratory condition [8].

In the literature, it was also found that the presence of multi-nanoparticles in composites improves the properties of nanocomposites. Some researchers used hybrid fillers in order to have a perfect potential of both fillers. For instance as a first group, a combination of micro rubber and nanosilica has been used to improve the fracture toughness and fatigue behavior of [9–15]. Liang and Pearson [9] used two different sizes of nanosilica (NS) particles, 20 nm and 80 nm in diameter, and carboxyl terminated butadiene acrylonitrile (CTBN) which was blended into a lightly cross-linked, DGEBA/piperidine epoxy system in order to investigate the toughening mechanisms. It was shown that addition of small amount of NS particles into CTBN, caused increase of the fracture toughness. Manjunatha et al. [10–15] investigated the fatigue behavior of reinforced composites by adding a combination of micro rubber and nano-silica particles into epoxy matrix in several states. For instance, they [10] studied the tensile fatigue behavior

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of modified micron-rubber and nano-silica particle epoxy polymers. They [11] also addressed the tensile fatigue behavior of a glass-fiber reinforced-plastic (GFRP) with participation of rubber micro-particles and silica nano-particles. They [12] also observed the enhanced capability to withstand longer crack lengths, due to the improved toughness together with the retarded crack growth rate, to enhance the total fatigue life of the hybrid-modified epoxy polymer. Also, Manjunatha et al. [13] enhanced the fatigue behavior of fiber reinforced plastic composites by means of 9 wt.% of rubber microparticles and 10 wt.% of silica nanoparticles and showed the fatigue life under WISPERX load sequence was about 4–5 times higher than that of the neat composites. Manjunatha et al. [14] also used another hybridization of carboxyl-terminated butadiene–acrylonitrile rubber microparticles and silica nanoparticles to increase the tensile fatigue behavior of GFRP composites at a stress ratio equal to 0.1. Manjunatha et al. [15] conducted fatigue crack growth test on a thermosetting epoxy polymer which was hybrid-modified by incorporating 9 wt.% of CTBN rubber micro particles and 10 wt.% of silica nano-particles. The fatigue crack growth rate of the hybrid epoxy polymer was observed to be significantly lower than that of the unmodified epoxy polymer.

In the next category, applying carbon nanotubes (CNTs) with different nanoparticles as hybrid fillers were taken into account in the literature [16–19] to improve the fatigue behavior, mechanical and electrical properties of reinforced composites. Böger et al. [16] appointed silica and MWCNT hybrid nanoparticles to increase the high cycle fatigue life of epoxy laminates and finally reported that the life was increased by several orders of magnitude in number of load cycles. Fritzsche et al. [17] investigated the CNT based elastomer-hybrid-nanocomposites prepared by melt mixing and showed promising results in electrical, mechanical and fracture-mechanical properties. Witt et al. [18] improved mechanical properties such as tensile strength and strain to failure of a conductive silicone rubber composite using both CNTs and carbon black (CB). Al-Saleh Mohammed and Walaa Saadeh [19] fabricated a nanostructured hybrid polymeric materials based on CNTs, CB and CNFs and investigated electrical properties and electromagnetic interference shielding effectiveness in the X-band frequency range.

The other various hybrid nanoparticles were discussed in the literature are considered here as the last category [20,21]. Jen et al. [20] applied hybrid Magnesium/carbon fiber to increase the fatigue life of nanocomposite laminates. On the other hand, applying carbon nanotubes (CNT) and graphite nanoplatelets (GNPs) to epoxy nanocomposites was shown by Li et al. [21]. It was represented that the flexural mechanical as well as electrical properties of the neat resin was marginally changed by hybridization.

This survey reveals that the effect of hybrid particles is mostly positive and can improve the static and dynamic properties of composites. However, it is figured out that in case of displacement control fatigue loading condition, there is a lack of research on this issue for hybrid nanofillers/epoxy nanocomposites. Therefore, in the present research, the flexural fatigue behavior of graphene/CNF/epoxy hybrid nanocomposites under displacement control flexural loading is investigated and compared with those of the pure epoxy resin.

2. Materials specification

2.1. Epoxy resin

In the present research, ML-526 (Bisphenol-A) epoxy resin was selected because of its low viscosity and extensive industrial applications to fabricate the specimens. The low viscosity of the matrix makes the dispersion of additives easier. Physical and mechanical properties of ML-526 epoxy resin are shown in Table 1. The curing agent was HA-11 (Polyamine). The ML-526 resin and the HA-11 polyamine hardener were supplied by Mokarrar Company, Iran.

2.2. Nanoparticles

In this research, graphene nanoplatelets (GPL) and CNF are utilized as carbon based nanofillers. The graphene nanoplatelets (GPL) were synthesized with a stirring grinding driven by changing the magnetic field as shown in Fig. 1. The steel needles with a weak magnetism are used as grinding media and four NdFeB permanent magnets are inserted into a motor-driven disc (Fig. 1). When the disc is made of steel, the magnetic stainless steel needles are attracted by the permanent magnets (Fig. 1a and b). By increasing the rotational speed, the magnetic stainless steel needles fly up and collide with each other with a high frequency under the changing attraction and repulsion forces of the high speed rotating permanent magnets (Fig. 1c). When a rigid grinding chamber filled with a certain amount of graphite powder is set on the disc, there are high frequent collisions and shears between the grinding chamber and magnetic stainless steel needles, which can finally result in a strong collision and shear forces. Graphite in the chamber will be crushed into ultra-fine powder under the action of these strong forces and then the powder will be prepared efficiently. Physical properties of synthesized graphene powders are shown in Table 2. The TEM image of the synthesized GPL powder is shown in Fig. 2. The D, G and 2D bands of Raman spectra of the synthesized GPLs powder are demonstrated in Fig. 3.

The CNF was supplied by Grupo Antolin SL, Spain. The physical properties of CNF are represented in Table 3. The Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) images of CNF nanoparticles are illustrated in Fig. 4.

3. Specimen preparation

The polymer nanocomposite reinforced with 0.5 wt.% of graphene/CNF hybrid nanoparticles/epoxy nanocomposites was prepared as described below. Firstly, epoxy resin was mixed with 0.25 wt.% CNF and stirred for 10 min at 2000 rpm and then the mixture was sonicated via 14 mm diameter probe-sonicator (Hielscher UP400S) at output power of 200 W and 12 kHz frequency. The mixture was sonicated for 60 min. It is worth mentioning that during the sonication, the mixture container was kept by the aid of ice-bath to prevent the overheating of the suspension to keep the temperature around 40 °C. Secondly, suspension was mixed with 0.25 wt.% GPL under same condition within 30 min by the sonication. After sonication, the hardener at a ratio of 15:100 was added to the mixture and stirred gently for 5 min. Then, it was vacuumed at 1 mbar for 10 min to remove any trapped air. Six samples were prepared and cured at room temperature for 48 h and followed by 2 h at 80 °C and 1 h at 110 °C for post curing.

The approach was used to disperse GPL/CNF hybrid nanoparticles into epoxy resin, is adopted from a combination of supplementary research [22]. Time for sonication depends on the filler contents and has been defined based on experiments until fillers remain intact. For CNF fillers, Shokrieh et al. [22] investigated the suitable time for sonication versus contents of the filler and pointed out for 0.25 wt.% CNF materials, the optimum value of sonication with regard to Fig. 5, was found around 90 min with the same compartment and conditions. Also, the optimum sonication time for 0.25 wt.% GPL was equal to 30 min. In addition, to inspect the dispersion state of nanofillers, a new technique based on scanning electron microscopy, which utilizes the burn-off test, was introduced to visualize the dispersion state of nanofillers [23].