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Graphene-Enhanced Fracture Resistance of Epoxy Nanocomposites for Structural Applications

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Abstract: The fracture characteristics of graphene nano-platelets (GNP) in epoxy composite under high strain loadings were investigated using impact test at room temperature. The energy required to create a new surface was measured with load cell and the accompanied plain strain fracture toughness was calculated. The investigation covered the effect of the amount of graphene in graphene/epoxy composite material system on the fracture behavior of the material. The results showed that both the manufacturing and experimental procedures developed were excellent to quickly and precisely characterize the response of GNP/epoxy composite to impact loadings. Furthermore, the results showed clear dependencies of the material response on the amount of GNP in epoxy.

Key words: Fracture, toughness, behavior, graphene, composite.

1. Introduction

Thermosetting polymers such as epoxy are utilized as the matrix phase for fiber-reinforce composites for engineering applications due to their crosslinked network structure. However, due to their inherent properties of brittleness and low fracture resistance, the addition of a small amount of nanoparticles, into the matrix of polymeric composites for the high performance of engineering structures has attracted the attention of engineers and scientist. This practice is gaining more popularity in the development and manufacturing of lightweight materials and products for automotive and aerospace applications as the strength of these composite materials can be increased when the size of the matrix reinforcement is reduced to nano scale.

Furthermore, as the practice of enhancing the mechanical performance of the matrix of the composite structures with the nanoparticles continues to gain a worldwide acceptance, the need to develop and perform a carefully controlled experimental testing of these newly developed nanocomposite materials and structures is also becoming important. The experiment is one of the most reliable methods that can be used to gain an understanding into the effect of nanoparticles on the composite performance under actual loadings. For this reason, researchers [1-3] have been working on the enhancement of the fracture toughness of epoxies that are used in fiber-reinforced composite.

The investigators [4-6] reinforced epoxy with nano-phase rigid fillers and found out that these nano-fillers enhanced the toughness to a larger extent than micro fillers at very low filler loading due to their high surface area. Rafiee et al. [7] compared the effect of GNP, single-walled carbon nanotubes, and multi-walled carbon nanotube nanoparticles on mechanical properties of epoxy nanocomposites and found GPN to be more effective in strengthening the epoxy matrix.

It has been shown [8] that nano-clay can be used to increase the fracture toughness of epoxy at higher filler contents (1.0-10.0 wt%). The investigators [9, 10] have found out that the fatigue life and fracture toughness of glass fiber reinforced composites are enhanced when epoxy matrix is infused with graphene. For nanocomposites in which the dispersed particles are of

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nano dimension, Johnsen et al. [11] have shown that the nano-scaled silica particles increase the toughness of the epoxy matrix through de-bonding of particles that is followed by plastic void growth. For the carbon nanotube reinforced epoxy systems, Hsieh et al. [12] stated that the nanotubes pull-out and de-bonding seems to influence the increase in fracture energy and that the contribution from the plastic void growth is very minimal.

Structurally, graphene nanaoplatelets (GNPs) are short stacks of individual layers of graphite (called graphene) that exhibit high stiffness and strength per unit mass. To document the effect of GNP on the mechanical behavior of composite materials, many researchers [1-13] have carried out both experimental and computational studies on the mechanical behavior of nano-filler based epoxy composites under different loadings. However, there are still few experimental data in the literature to document the effect of weight fraction of graphene on the fracture resistance of GNP based epoxy composite. The objective of this study is to experimentally characterize the toughness behavior of GNP/epoxy systems for different weight fractions of GNP in epoxy. The investigation covers the new methodology for manufacturing the GNP/epoxy composite specimens (Charpy bars), the process for testing, and the techniques used for measuring the output data.

The paper arrangement is as follows: Section 2 documents the methodology of the research work. Section 3 explains the results and discussion. Section 4 presents the conclusions.

2. Methodology

The materials and experimental methods are documented below.

2.1 Materials and Specimen Preparation

A ratio of 2:1 epoxy resin and hardener was mixed thoroughly using a wooden stick for about 5 min to fabricate the pure epoxy. Then the material system was degassed in an oven at a temperature of 50 ºC and 15 in. Hg vacuum for 10 min. It is important to note that the specimens that are fabricated by machining often consist of defects and flaws due to the surface roughness that are caused by the cutting tools. The degree of these defects depends to some extent on the types and the life of the tool used for cutting. Invariably, these geometric discontinuities can cause experimental errors in determining the mechanical behavior of the nanocomposites. A rapid prototyping technique was utilized to make the 3-D printing of Charpy bars pattern out of ABS plastic in order to prevent the errors that can be caused by machining of the specimens. The bars fabricated from 3-D printer were then placed into a boxed frame, and the silicon mold as shown in Fig. 1 to make rapid prototyping tools for the high volume production of the test samples. Immediately after the epoxy was degassed, the epoxy mixture was poured into the mold and the material was allowed to gradually cure at room temperature for five days. It is noteworthy that these bars were made based on the ASTM specifications for the impact test. The ASTM D 6110 standard is defined by a size of 55 mm long, a 12.7 × 12.7 mm section and a central V-notch of 2 mm.

To manufacture the GNP/epoxy composite, a predetermined weight fraction of GNP obtained from XG Science [14] was added to the epoxy resin in a

Fig. 1 The rapid prototyping tool for making the tensile bars.

beaker and the material was mixed thoroughly using 2-in. diameter disperser blade in a Ross high shear mixer HSM-100 LSK-I at a rotating speed of 1,500 rpm for 60 min. Then the hardener is added to the GNP/resin materials and mixed with a stick for 5 min. The mixture was degassed in an oven at a temperature of 50 ºC and 15 in. Hg vacuum for 10 min before it was poured into the mold that had been made based on the ASTM Charpy bars. The GNP/epoxy was allowed to gradually cure at room temperature for five days.

The GNP utilized in this study is xGnP-C-300, and was obtained from XG Sciences. It consists of grade C particles with an average size of 2 μm platelet diameter and a thickness of 2 nm. Its average surface area is 300 m^2/g and has a density of 2.0 g/mL [14].

2.2 Impact Test Method

The rapid prototyping method utilized in making the specimens allowed many specimens to be made into precision of ASTM Charpy bars without any secondary machining operations. The goal of this study is to find the fracture toughness of the GNP-based epoxy for different weight fraction of graphene. To accomplish this, the specimen was tested on the INSTRON impact tester shown in Fig. 2. For precise measurement, the machine was first calibrated prior to the testing and the hammer was raised to 50 J potential energy and then released to impact the specimen to failure. The data obtained from the machine showed how much of the energy was required to fracture the material and the impacted energy. The experimental investigation was done four times on pure epoxy, GNP/epoxy that has 1% GNP, and GNP/epoxy that has 3% GNP and the average results were computed.

2.3 Scanning Electron Microscope (SEM) Examination

To evaluate fracture surface of these composites, the fracture surfaces of several samples including the one for pure epoxy were observed using a JOEL 6330F

Fig. 2 INSTRON impact pendulum.

Scanning Electron Microscope (SEM). The fracture surfaces were sputter coated with gold to allow a better observation of the surface morphology.

3. Results and Discussion

3.1 Impact Test Results

As stated before, four different samples of pure epoxy, GNP/epoxy with 1% of graphene, and GNP/epoxy with 3% of graphene were tested, respectively, on Instron impact pendulum machine. The results for the specific fracture energy are shown in Fig. 3. For the pure epoxy specimens, the fraction of the total energy absorbed by the material ranges from 1.45%-1.64% (0.727-0.820 J) with an average of 1.55% (0.775 J). These correspond to the specific energy within the specimens of pure epoxy ranging from 4.48-5.11 kJ/m^2 with an average value of 4.78 kJ/m^2. The energy absorbed was calculated by multiplying the cross sectional area of the specimen by the specific energy obtained from the test. For the 1% GNP specimens tested, the fractional energy absorbed by the material ranges from 1.51%-1.61% of the impact energy (0.755-0.807 J) with an average of 1.56% (0.781 J). The specific energy ranges from 4.76-5.10 kJ/m^2 with an average of 4.89 kJ/m^2. The 3% GNP specimens absorbed a fraction of the total energy

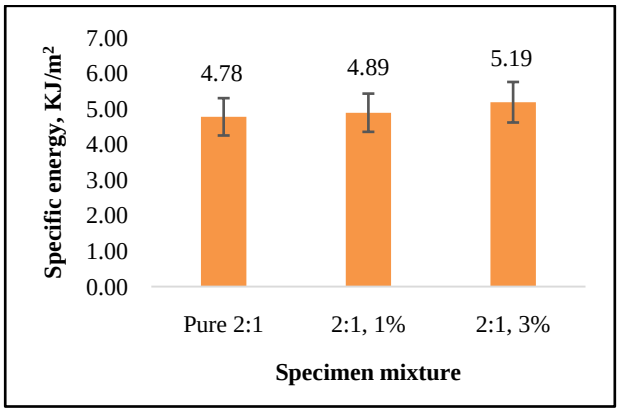


Fig. 3 Average specific fracture energy.

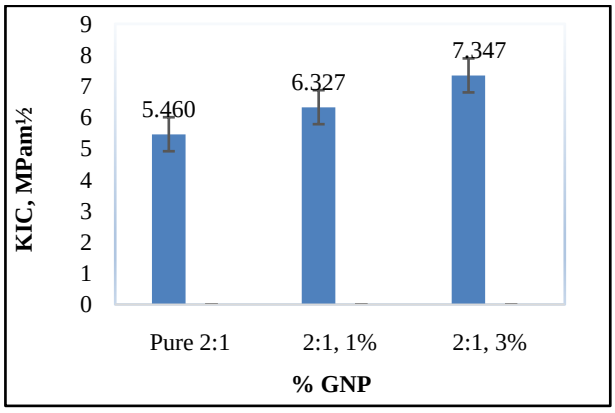


Fig. 4 Average fracture toughness.

ranging from 1.61%-1.80% (0.806-0.89) with an average of 1.70% (0.849 J). The specific energy ranges from 4.92-5.69 kJ/m^2 with an average of 5.19 kJ/m^2. The above results showed that the infusion of GNP into the matrix of epoxy increases the fracture resistance of epoxy.

The equivalent value of fracture toughness was also computed using Eqs. (1) and (2) expressed as:

$$\sigma_c = \left(\frac{2E\gamma_s}{\pi a}\right)^{\frac{1}{2}} \quad (1)$$

$$K_{Ic} = Y\sigma_c\sqrt{\pi a} \quad (2)$$

where,

E = modulus of elasticity;

γ_s = specific surface energy;

a = the length of an external crack;

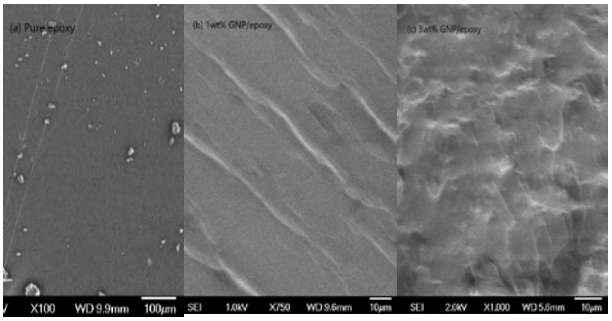
Y = 1.1 [15];

K_{Ic} = fracture toughness.

It should be noted that the values of E utilized for the nanocomposites and pure epoxy to obtain the fracture toughness were taken from Ref. [13]. Fig. 4 shows the fracture toughness of the pure epoxy, 1% GNP in epoxy, and 3% GNP in epoxy. The investigation showed that the strengthening effect of graphene on epoxy composites increases with the weight fraction of graphene. This suggests that the increase in fracture toughness of GNP/epoxy composites is due to the crack pinning by the nano fillers and the higher the weight fraction of GNP in the epoxy network, the more the crack growth was inhibited on the nanocomposites.

3.2 Fracture Surface Examination through SEM

Fig. 5 shows the micrograph of the fracture surfaces of a sample made with pure epoxy, GNP/epoxy sample with 1 wt% GNP, and GNP/epoxy sample with 3 wt% of GNP. It is clear that the facture surface of the sample made with pure epoxy is smooth and very similar to the typical fracture surfaces observed by other investigators. In contrast, the fracture surfaces of the samples containing 1 wt% or 3 wt% GNP are much rougher than those of pure epoxy. It is clear that for samples containing GNP, a large number of fracture surfaces are present. These large numbers of fracture surfaces are the major contributing factor for improving the mechanical integrity of this composite structure. It is also interesting to point out that some of these fracture surfaces have different heights and show some GNP platelets are separated at the crack boundary.



(a) (b) (c)

Fig. 5 SEM micrograph of the fracture surfaces of three different samples: (a) pure epoxy, (b) sample with 1 wt% GNP, and (c) sample with 3 wt% GNP. Samples (b) and (c) show the fracture surfaces at higher magnification for 1% GNP and 3% GNP, respectively.

4. Conclusion

The fracture resistance of graphene based epoxy nanocomposites was experimentally investigated using charpy bar specimens. The specimens utilized in this study were fabricated using rapid prototyping tool to circumvent the surface roughness effect that can be caused by machining. The epoxy nanocomposites were toughened with GNP. The impact tests and the micrographic assessments were carried out on pure epoxy as well as GNP-reinforced epoxy composites. The impact energy values and the equivalent fracture toughness for the pure epoxy and GNP/epoxy material systems were measured and compared. The addition of graphene to the pure epoxy network showed an increase in the fracture resistance of the epoxy nanocomposites, but this only increases with GNP contents due to an increase in crack-pinning sites.

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