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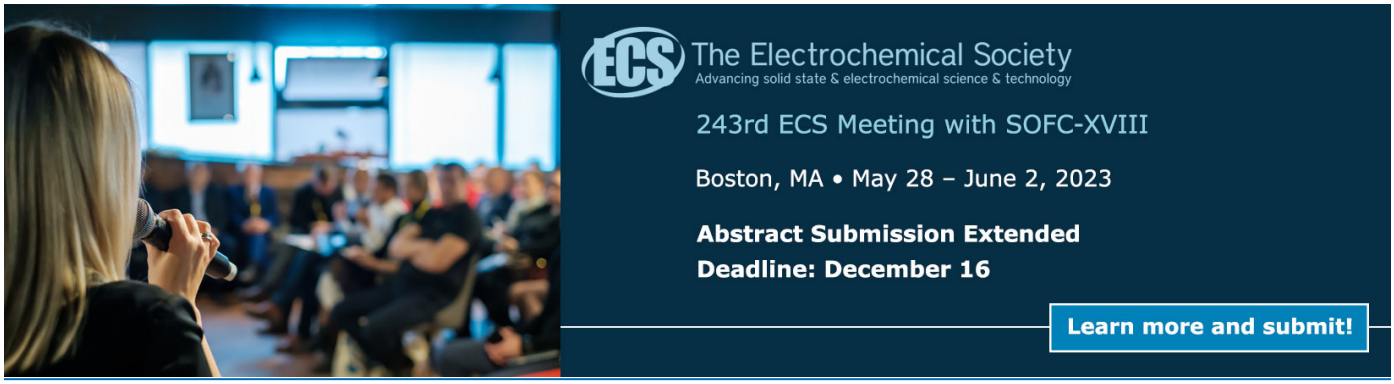

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The effect of graphite flakes (GFs) and hybrid graphene nanoplatelets (GNPs) particles to the mechanical properties of epoxy composites

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Abstract. This paper reports the effects of epoxy when reinforced with graphite flakes (GFs) and was compared to epoxy reinforced by hybrid reinforcements consisted of graphite flakes (GFs) and graphene nanoplatelets (GNPs). DGEBA, the type of epoxy which has been used in this work was added with 2, 4 and 6 wt.% of reinforcement respectively, relative to the total weight of the mixture. Nanocomposites was prepared using mechanical stirrer, stirred at 2000 rpm for 30 minutes followed by curing in the oven. The flexural testing shows that the epoxy/GFs composites has higher modulus compared to epoxy/GFs+GNPs. At 4wt.% filler loading, epoxy containing GFs exhibited 132% enhancement of modulus relative to neat epoxy. At the same filler amount, epoxy containing GFs+GNPs demonstrated slightly lower magnitude than system containing GFs with only 27% increase in modulus. However, addition of higher filler loading to the epoxy resin caused the modulus to decrease in magnitude speculated due to agglomeration of particles within the host matrix. The Charpy impact testing indicated similar pattern with epoxy/GFs system exhibited higher capability in absorbing energy than epoxy/GFs+GNPs where the peak was obtained at 4wt.% filler loading. At this amount of filler, 18.36 J/m increase in energy absorbed was recorded for epoxy/GFs compared to 2.13 J/m increased for epoxy/GFs+GNPs composite. Higher amount of filler loading added into epoxy only deteriorate the impact energy absorb by the sample. The hardness test show similar pattern of result with epoxy/GFs shows higher resistance in scratching compared to epoxy/GFs+GNPs.

1 Introduction

Nowadays, there are many areas in which polymer composite materials has been used due to advantages it can offers for example high specific strength and stiffness, high wear

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resistance, excellent corrosion, chemical resistance, high dimensional stability, reduces noise and flexibility shapes [1]. The best example of polymer composite materials is epoxy resin that widely adapted in many application such as adhesive, aerospace composite, automotive components, used as the binder in countertops or coatings for floors, offshore equipment, boats, pipes, and pressure vessels [2]. Epoxy resin has good in mechanical properties, high adhesion strength, good heat resistance, and high electrical resistance [3].

Nevertheless, despite giving benefit in strength, epoxy resin is known to be disadvantage in ductility. It offers poor brittleness characteristics. Over the years, there are many research works which has been carried out to modify the properties of epoxy by addition of fillers to ameliorate the properties matrix dominated composite [1]. The typical filler content needed for significant enhancement of these properties can be as high as 10-20% by volume [4].

Nanomaterials are basic of nanoscience and nanotechnology [6]. They are materials or chemical substances which function at a very small scale. Nanoscale materials are substances where at least one dimension is less than approximately 100 nanometers. Nanometer is approximately 100,000 times smaller than diameter of a human hair which means of one millionth of a millimetre[5]. It can be found in sporting goods, stain resistant clothing, tires, electronics, sunscreens, cosmetics, as well as many other items, and are also used in medicine, imaging and drug delivery. Nanomaterials have several advantages compared to the same materials without nanoscale features such as high strength, high chemical conductivity and reactivity [6].

Graphene is a two-dimensional platelet consisting of carbon atoms, arranged in a hexagonal shapes or honeycomb structure that have attracted the great deal of attention due to excellent physical and electronic properties [7]. GNPs consists of small stack of graphene that can be replace carbon fiber, nano clays, carbon nanotubes or any other compounds in many composite applications. GNPs is also known as graphite nanosheets, graphene nanosheets (GNs), graphite nanoplatelets and graphite nanoflakes (GNFs) [8]. The improved mechanical properties of nanocomposites is due to the uniform dispersion of functionalised graphene and strong interfacial bonding between modified graphene and epoxy resin where this can be confirmed by microscopy observations [9]. Graphene have the incredibly high specific surface area, unique graphitised plane structure and extremely high charge mobility[9]. Because of their unique nanoscale size, shape, and material composition, graphene nanoplatelets can be used to improve the properties of a wide range of polymeric materials, including thermoplastic and thermoset composites, natural or synthetic rubber, thermoplastic elastomers, adhesives, paints and coatings [10]. The extraordinary properties of graphene could only happen if graphene nanoplatelets are well spread in matrix and that there is a strong interfacial adhesion between graphene nanoplatelets and polymer matrix [9].

GFs is a three-dimensional form of carbon atoms [11]. Basically, it is a layered material and also can be observe as a two-dimensional graphene crystal that weakly joined [12]. Graphite colour is dark gray to black and became black gray colour when it was crushed to powder. The graphite crystal lattice consisted of sp2-hybridized carbon atoms which bonded covalently in hexagonal rings [13]. The layers between atom which eventually forms graphite are bonded to each other by weak Van der Waals forces. Graphite has been widely used in variety of the areas of industry, transport, energy, defense and medical. It is due to its prominent structural, electrical and mechanical properties.

From the previous works, various particles and effect of nanoreinforcements to the properties of polymer matrix had been studied. For example, Chatterjee [7] studied the mechanical reinforcement and thermal conductivity in expanded graphene nanoplatelets reinforced epoxy composites. Their group processed the particles from natural graphite flakes (NG) and converted into expanded graphene nanoplatelets (EGNPs) using

acidification procedures. Embedded in the bisphenol A type of epoxy as matrix, they later characterised the performance of the composite by mechanical testing. The mechanical testing that they have used for characterisations are three-point bending test and fracture toughness test. They demonstrated that significant improvement in mechanical properties for all samples with EGNP addition compared to neat epoxy. By increasing the concentration of EGNP in the composite, fracture toughness (K_{1C}) increases steeply, reaches a highest at some amount of loading and decreases subsequently at higher EGNP loadings due, speculated due to agglomeration. In addition, the load transfer from the matrix to the EGNP fillers shows increase in flexural modulus when stress is being applied [7] [14].

A composite can be termed as hybrid, if two or more types of fillers are combined in a common matrix to produce a composite that drives benefits from each of the individual fillers and exhibits a synergetic response [15]. Basically, it will increase the elastic modulus, decreases brittleness controls cracks initiation and its subsequent growth and propagation. The advantage of hybrid reinforced composite is to provide an arrangement where one type of filler, which is stronger and stiffer, ameliorate the first cracks stress and ultimate strength, and another type of filler, which is more flexible, and ductile, leads to reinforced toughness and strain in the existed cracking zone [16]. Previously, the effects of hybrid nanoreinforcements to the properties of polymer matrix were reported. For example, Subhani et al [17] developed composites containing multiwalled carbon nanotubes (MWCNTs) and nanodiamonds (NDs) to explore the hybridized effect of nanoreinforcements on the mechanical performance of nanocomposites [17]. Hardness test was carried out to investigate the mechanical properties of nanocomposites. It was discovered that neat epoxy showed a hardness value of 11.5 ± 0.4Hv, which increased to 14.1 ± 0.3 Hv (22% rise) by adding 0.1 wt.% MWCNTs and 12.8 ± 0.6 Hv (11% rise) by the addition of 0.1wt.% NDs. The effect of the incorporation of low content of MWCNTs in epoxy matrix is more pronounced in comparison to NDs. The combined effect of each NDs and MWCNTs at a loading of 0.05 wt.% showed a rise in the hardness value, 13.6 ± 0.6 Hv (18% rise), which is greater than the effect of NDs and less than the individual influence of MWCNTs on hardness but it shows the hardness values is continuously increase with contents of MWCNTs and NDs, 0.1 wt.% and 0.2 wt.% each which are 16.4 ± 0.7 Hv (42% rise) and 17.3 ± 0.8 Hv (50% rise). However, they stated that, generally, increasing the concentration of nanoreinforcements will cause the formation of particles agglomeration [17].

2 Experiment

2.1 Materials

The first stage of this study consists of selection of materials and preparation of the materials. GNPs was purchased from Sigma Aldrich. The density of graphene is from 0.03-0.1 g/cm^3. Graphite bulk was ground by using mortar to turn into GFs. Epoxy resin (Araldite LY 556) was used as matrix combined with hardener, Triethylenetetramine (TETA) to give great binding properties between the filler and the matrix [18]. Curing of epoxy resin was at room temperature in this work. Types of epoxy resin were Araldite LY 556 and Diglycidal Ether of Bisphenol A (DGEBA), and the hardener or curing agent was HY 951, Triethylenetetramine (TETA). This type of hardener was employed to improve the interfacial adhesion and impart good strength to the composite [19]. The mixing ratio of

resin to hardener is 2:1 as suggested by manufacturer to obtain the optimum degree of crosslinking.

2.2 Composites Fabrication

Fig. 1. The filler (a) Graphene Nanoplatelets (GNPs) and (b) Graphite bulk (GFs).

The composite was prepared in different compositions of filler (GNPs and GFs) which were 0wt.%, 2.0wt.% (1.0wt.% GNPs/1.0wt.% GFs), 4.0wt.% (2.0wt.% GNPs/2.0wt.% GFs) and 6.0wt.% (3.0wt.% GNPs/3.0wt.% GFs). For single filler, the composition will be the 2.0wt.% GFs, 4.0wt.% GFs and 6.0wt.% GFs. Firstly, the desired amount of epoxy and hardener were added into a cup. After that, the certain amount of compositions of filler was placed on the mixture of epoxy and hardener. Then, the mixture was stirred by using mechanical stirrer at 2000 rpm for 30 minutes. After that, the mixture was degassed using vacuum oven for 15 minutes at 50°C to eliminate the air bubbles [20]. It is important to remove the air bubbles because their presence will significantly affect the mechanical properties of the composite. Afterwards, the mixture of epoxy and filler was poured into the mould. Before fabrication of composite, the mould was sprayed with the mould release as a release agent to ensure that the composite and the mould will not stick and get hard for withdrawal. The mould would also be coated by the plastic wrap tape. The moulding will be left in the mould with load placed on top for about 24 hours to allow the settlement within mould container and for appropriate curing at room temperature. After the composite was cured, the casted product was withdrawn from the mould. The samples were cut to the required dimensions as per individual test requirements and example of the specimens are indicated as shown in Fig. 2a and 2b.

Fig. 2. Sample for (a) Flexural Test (b), Impact Test.

2.3 Mechanical testing

There are three different types of mechanical testing which was carried out in this work which includes hardness test, impact test and flexural test. The specimen is conducted in the standard laboratory atmosphere at temperature 24 ± 5 °C.

2.3.1 Hardness Test

The Vickers hardness test was carried following standard ASTM E384. It uses a square base diamond pyramid as the indenter. The included angle between the opposite faces of the pyramid is 136º. As a result of the indenter's shape, the impression on the surface of the specimen will be a square. The loads are between 1 and 1,000 g which referring to micro hardness test. In this hardness testing, 100g load was used with 5 seconds of dwell time.

2.3.2 Impact Test

This test provides a measure of energy required to break a material under impact loading. The test consists essentially of a hammer with a given amount of energy striking a notched test piece of fixed dimensions and recording the energy required to fracture the specimen at a specific temperature and recording whether the fracture mode was ductile or brittle. This test is used for determining behavior of material subjected to a shock load. Here the quantity measured is the energy absorbed by the specimen after being struck by a pendulum in a single blow. Normally, the impact strength of the samples is largely determined by the filler composition and strength regardless of the resin used. An impact testing machine with the Charpy arrangement is employed to perform the test having a striking angle of hammer is 124.4° and use 1J hammer strike. The specimens testing are prepared according to ASTM D256 [21] with dimension of 80 mm length, 10 mm width and 4 mm thickness with v-shaped notch depth 2mm.

2.3.3 Flexural Test

Flexural testing or also known as bending test is used to determine the bending properties of a material as well as known as three-point bending that consists in applying a point load at the centre of the material specimens. According to the ASTM D790 standard the dimensions of specimen used are 125 mm length 12.7 mm width and 3.2 mm thickness [23]. The span distance (distance between two support points) is 52 mm. The test speed was 2 mm/min [23].

3 Results

3.1 Mechanical Testing Results

3.1.1 Hardness Test

Fig. 3 shows the graph for micro vickers hardness test result of epoxy resin with different content and different composition of particles. From Fig. 3, the hybrid nanoreinforcements shows the smaller vickers hardness than the epoxy/GFs composite reinforcements. It shows that the 2wt.% of epoxy/GFs+GNPs (15.69HV) increase than the value of neat epoxy (13.86HV). The highest value for the epoxy/GFs+GNPs for 4wt.% is 18.525HV. The hybrid nanoreinforcement for 6wt.% of epoxy/GFs+GNPs shows the decreases in vickers hardness test which is 16.15HV. This may be due to, at higher wt.%, the reinforcements act as flaws and it were not perfectly aligned with matrix. For the epoxy/GFs, the vickers hardness test also increase at 2wt.% (16.385HV) GFs compared to neat epoxy (13.86HV). The vickers hardness value remain increase when 4wt.% of GFS was added yield 23.61HV but decreased when 6wt.% of GFs was added. It shows that adding a small amount of

reinforcements into epoxy resin could potentially enhance the scratch resistance which optimum at 4wt.%. Adding more particles beyond 4wt.% indicated that the hardness was dropped potentially due to agglomeration of particles which caused uneven dispersion within epoxy hence indentation becomes unstable during testing. Subsequently, lower reading was recorded when particles more than 4wt.% was added into the epoxy.

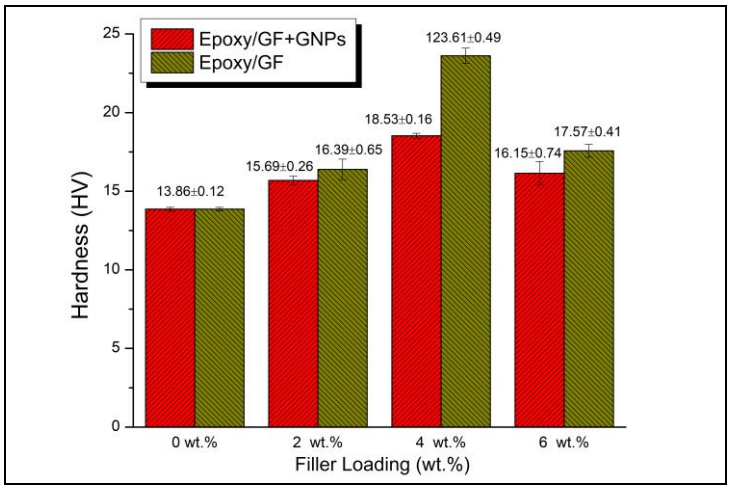

Fig. 3. Graph of micro vickers hardness test.

3.1.2 Flexural Test

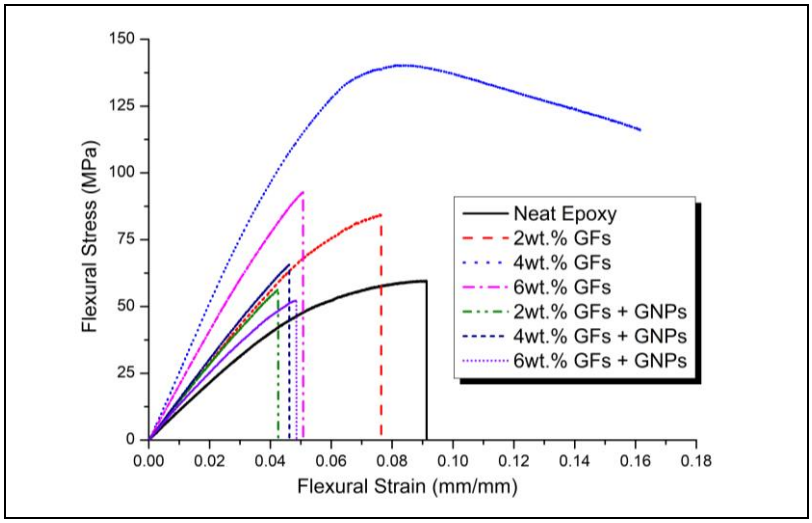

Fig. 4. Stress versus strain for each specimen as a function of reinforcement content.

Table 1. Flexural Properties of Composite (n = 3 to 6).

Flexural Test	**Modulus (MPa)**			
Sample	**0wt.%**	**2wt.%**	**4wt.%**	**6wt.%**
Epoxy/GFs+GNPs	1110.07 ± 65.96	1410.41 ± 80.88	1572.54 ± 157.57	1367.39 ± 82.49
Epoxy/GFs	1110.07 ± 65.96	1523.44 ± 103.09	2577.56 ± 26.62	2174.81 ± 147.49

Fig. 4 shows the flexural stress – strain curve of each different composition while Table 1 displays the summary of flexural modulus. From the graph and table, the 4wt.% epoxy/GFs shows the highest value of the modulus, 2577.56MPa. similarly, for epoxy/GFs+GNPs, 4wt.% is the highest amount of the weight percentage which is 1572.53MPa. if more reinforcement was added into epoxy beyond 4wt.%, the modulus of the composite decreased perhaps due to agglomeration. Amount of the GFs and GNPs is excessive as well as the epoxy cannot combined well and start to re-aggregate. Same with epoxy-GFs, the modulus will decrease if the parameter is more than 4wt.% which is 6wt.% at 2174.81MPa. Experimental results show that, micro sized particles have effect on modulus and strength of epoxy such as increasing modulus and strength but at certain limit it will decrease the mechanical properties of material. In addition, the flexural strength will increase if the number of filler increases. This is due to the increasing number of filler will increase the barrier in the composite to resist the crack [21, 22]. At certain percentage of fillers, the flexural strength of the specimen will decrease due to the agglomeration of fillers.

3.1.3 Impact Test

Table 2 shows the summary of impact strength of epoxy resin with different content and different composition of particles. From the Table 2, the composite containing hybrid particles shows the smaller impact strength than the composite containing GFs alone. For the epoxy/GFs+GNPs, the impact strength increases when 2wt.% (11.25J/m) of filler was mixed to the epoxy compared to neat epoxy (10.29J/m). The impact strength value was also increased when the hybrid reinforcement composites increased for 4wt.% (12.42J/m) but decreased for 6wt.% (11.13J/m) of hybrid composite. For epoxy/GFs, the value of impact strength also increases when 2wt.% of GFs (19.53J/m) was added into the composite of compared to the neat epoxy (10.29J/m). The energy absorbed was found increased continuously for reinforcements of epoxy/GFs at 4wt.% (28.65J/m) but decreased at 6wt.% (17.13J/m). The highest value was achieved for epoxy/GFs at 4wt.% which is 28.65J/m. For the epoxy/GFs+GNPs, the highest value is from 4wt.% specimen which is 12.42J/m. This mechanical testing shows that the epoxy/GFs has higher impact strength than the epoxy/GFs+GNPs. The hybrid particles of GFs+GNPs can improve the mechanical properties of neat epoxy on the impact strength but not as much as in the case for epoxy/GFs. As the conclusion for impact test, the epoxy/GFs absorbs more impact strength than epoxy/GFs+GNPs.

Table 2. Result of impact strength.

Impact Test	Impact Strength (J/m)			
Sample	0wt.%	2wt.%	4wt.%	6wt.%
Epoxy/GFs+GNPs	10.29 ±0.31	11.25±1.13	12.42±0.31	11.13±0.66
Epoxy/GFs	10.29±0.31	19.53±0.78	28.65±1.95	17.13±2.18

4 Conclusion

This study shows that GFs and hybrid particles of GFs and GNPs can affect the properties of epoxy. The present of hybrid particles and nanoreinforcements composite exhibited enhancement in mechanical properties of the epoxy. The finding indicate that these particles are effective to become obstacles to resist crack/failures. The higher the weight percentage of particles within the system, the better the mechanical properties of the composite but there was an optimal limit in which beyond the optimum limit, the particles will deteriorate the mechanical performance of composites probably due to agglomeration.

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