

THERMAL CONDUCTIVITY AND IMPACT PERFORMANCE OF GRAPHENE/EPOXY NANOCOMPOSITES

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Keywords: Graphene, Nanocomposite, Thermal conductivity, Impact performance

ABSTRACT

In this paper, various graphenes are added into epoxy matrix to fabricate graphene/epoxy nanocomposites, and graphene weight fraction is controlled as 1, 2 and 5wt%. There are two kinds of graphenes including GO (Graphene Oxide) and RGO (Reduced Graphene Oxide). During the composite processing, two kinds of ultrasonication (low power) 300W and ultrasonication (high power) 1500W are incorporated to disperse graphenes in epoxy. Effect of processing parameters on two physical properties of graphene nanocomposites is investigated including thermal conductivity and impact performance. Thermal conductivity of composites is measured by thermal interface material (TIM) measurement system, and impact energy and force are measured by Hobbit drop weight impact tester. Among various graphene/epoxy composites, RGO(5wt%)/epoxy by ultrasonication 300W/30 min has highest thermal conductivity (TC) of 0.529 W/m-k, 43% higher than 0.37 W/m-k of epoxy. RGO(5wt%)/epoxy has highest impact force of 1412.08 N, 211% higher than 452.93N of epoxy; while RGO(5wt%)/epoxy by ultrasonication 300W/30 min has highest impact energy of 21.03 KJ/m², 63% higher than 12.88 KJ/m² of epoxy.

1 INTRODUCTION

Nanocomposite materials with nanoscale reinforcements have emerged in the past decades as a promising novel class of materials, which take advantage of greatly increased specific interfacial area, higher attainable loads, controlled interfacial interactions, and higher total compliance. Various graphene materials have very recently emerged as a new class of potential components for advanced nanocomposites with intriguing new opportunities for the integration into polymer matrices. Graphene nanocomposites show not only superior mechanical properties but also impressive functional properties, such as thermal conductivity, unique photonic/optical transportation, anisotropic transport, and low permeability. The most important material characteristics for graphene are [1-4]: the highest elastic modulus of 1 TPa, very high thermal conductivity of $5.1 \times 10^3 \text{ W m}^{-1} \text{ K}^{-1}$, and the highest known intrinsic electrical conductivity of $6 \times 10^5 \text{ S m}^{-1}$. It is noteworthy that thickness of individual graphene sheets of 0.34 nm is the thinnest 2D nanofiller known to date [5].

Graphene oxide (GO) is an oxidized graphene-derived products, which can be widely used as an alternative for graphene materials due to its high dispersibility and processibility in aqueous environment [6-8]. GO can be produced from mineral graphite flakes by thermal oxidation method invented by Hummers [9]. The achieved single atomic layer graphene-like material possesses high density of epoxide and hydroxyl functional groups (functionalities) on both sides of the basal carbon plane and carboxyl groups around their edges [10]. GO can be reduced to graphene-like structures, named reduced graphene oxide (RGO), with similar mechanical and conductive properties but less functionalities by chemical, electrochemical, thermal, hydrothermal, and photothermal reducing techniques [11,12].

Based on the previous literature review, authors propose that study of both functional and mechanical properties of graphene composites are equally important for applications of the

composites. This paper is aimed at clarifying enhanced mechanisms of GO and RGO on epoxy matrix subjected to heat and impact. Thermal interface material (TIM) and drop weight impact tests are conducted to evaluate thermal conductivity and impact performance of GO/epoxy and RGO/epoxy nanocomposites.

2 EXPERIMENTAL PROCEDURES

2.1 Fabrication of graphene nanocomposites

Graphene oxide (GO) is fabricated by modified Hummers method. First, 10 gram potassium persulfate $K_2S_2O_8$ and 10 gram phosphorus pentoxide P_2O_5 are added to 50 ml Sulfuric acid H_2SO_4 and kept stirring to achieve the solvent. Then 12 gram graphite powders are added to the solvent and the solution are heated to 85 °C and stirred for 4.5 hours. The solution is poured into 2 liter deionized water and kept still for 12 hours. The solution is then vacuum filtered by a filter paper with 0.2 μ m pores and followed by dried in oven at 90 °C for 2 hours in order to achieve the pretreated graphite powders. The powders are added to 460 ml H_2SO_4 and the solution is stirred in an ice bath. Later, 60 gram $KMnO_4$ are slowly added to the solution and heated to 35 °C and stirred for 2 hours. Then 920 ml deionized water is slowly added the solution and the temperature has to be kept under 50 °C. The solution is further diluted in 2800 ml deionized water, and then 50 ml 30% concentration H_2O_2 is added. The solution stands still for 24 hours. The solution is initially set in centrifuge by revolution of 6000 rpm for 1 hour to separate solute and solvent. The solute is further purified as GO by the addition of mixture of water and methanol. The centrifuge and purification processes are repeated for several cycles.

To achieve the reduced graphene oxide (RGO), hexamethyl-enetetramine (HMTA) is used as reduction agent. HMTA is added to the water solution of GO, and weight ratio of HMTA to GO is 1.3 to 3.0. The reduced solution is kept stirring at 95 °C for 12 hours without evaporation. By the centrifuge process similar to that of GO, RGO can be successfully achieved.

To fabricate GO/epoxy or RGO/epoxy nanocomposites, GO or RGO is first mixed with epoxy resin in a container by a stirrer and put on a hot plate at 80°C for 1 hour. Weight fraction of GO or RGO, denoted as W_{GO} or W_{RGO} , versus epoxy is controlled as 1.0, 2.0, 5.0 wt%. The mixture is then subjected to either low power ultrasonication (300 W) for 30 minutes or high power ultrasonication (1500 W) for 1 to 3 minutes in order to uniformly disperse GO or RGO in the epoxy resin. Curing agent is then added into GO/epoxy or RGO/epoxy resin mixture and weight ratio of resin versus curing agent is 5:3. The composite mixture is again uniformly mixed by stirrer. This composite mixture is then poured into two kinds of silicone molds to make specimens for thermal conductivity and impact tests, respectively. The former has round shape with diameter and thickness of 18 mm and 1 mm, and the latter has square shape with length×width×thickness of 55×55×3 mm. The composite specimens are then put in an oven at 90°C for 180 minutes for curing.

2.2 Testing

Thermal conductivity of composites is measured by thermal interface material (TIM) apparatus. The apparatus is designed according to ASTM-D5470 standard. The round shape specimen is clamped at thickness direction and known heat source is provided through thickness direction. According to Fourier's law of heat conduction, thermal conductivity (TC) of composites can be calculated. Impact performance of composites is measured by Hobbit drop weight impact tester. The apparatus is designed according to ASTM-D7136 standard. The drop weight head has semi-sphere shape with diameter of 8 mm and weight of 460 grams. The stroke of the head is 200 mm. The apparatus can record impact force and energy upon impact.

3 RESULTS AND DISCUSSION

3.1 Effect of graphene type and weight fraction

Figure 1 shows effect of graphene type and weight fraction on TC of graphene nanocomposites. Thermal conductivities of both GO/epoxy and RGO/epoxy increase with graphene weight fraction.

Because graphene possesses high TC, higher graphene weight fraction indicates better connecting network among graphene nanosheets, and this leads to higher TC of the nanocomposite. In this figure, highest thermal conductivities for GO/epoxy and RGO/epoxy are 0.45 W/m-k and 0.53W/m-k respectively. TC of RGO/epoxy is all higher than that of GO/epoxy. This is probably due to the undulation nature of RGO nanosheets [13], which result in more connecting networks among graphene nanosheets.

Figure 2 depicts effect of graphene type and weight fraction on impact energy of graphene nanocomposites. Impact energies of both GO/epoxy and RGO/epoxy increase with graphene weight fraction. This indicates that more graphenes in the epoxy matrix may lead to more energy absorption modes such as crack deflection and graphene pull-out. Impact energy of GO/epoxy is all higher than that of RGO/epoxy. This is probably due to weak interface for GO/epoxy. Effect of graphene type and weight fraction on impact force of graphene nanocomposites is depicted in figure 3. Impact forces of both GO/epoxy and RGO/epoxy increase with graphene weight fraction. RGO/epoxy composites all have higher impact forces than those of GO/epoxy composites.

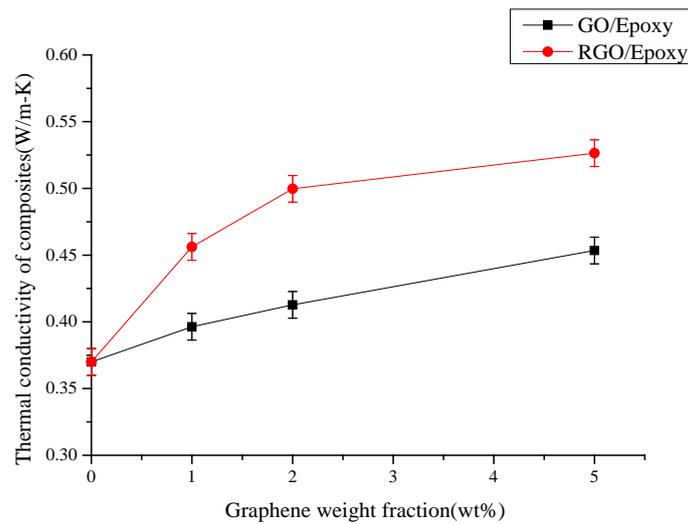


Figure 1: Effect of graphene type and weight fraction on thermal conductivity of graphene nanocomposites.

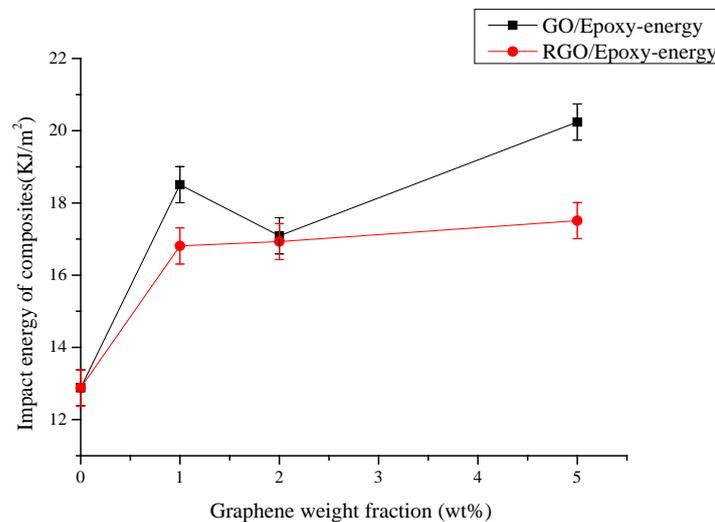


Figure 2: Effect of graphene type and weight fraction on impact energy of graphene

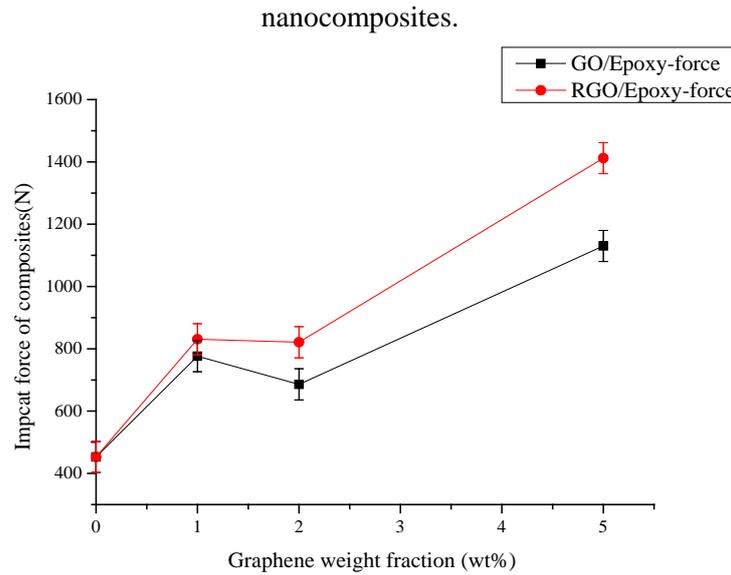


Figure 3: Effect of graphene weight fraction on impact force of graphene nanocomposites.

3.2 Effect of ultrasonication

Figure 4 indicates effect of ultrasonication (low power) on TC of GO/epoxy and RGO/epoxy. Results show that ultrasonication can increase TC of both GO/epoxy and RGO/epoxy at lower graphene weight fraction. This infers that ultrasonication can well disperse graphenes in epoxy resin, and thus leads to better connecting network among graphene nanosheets and higher TC. In figure 5, another ultrasonication method with high power of 1500 W is adopted for 1 minute to study its effect on TC of GO/epoxy and RGO/epoxy. Best TC occurs for both composites with graphene weight fraction of 2.0wt%.

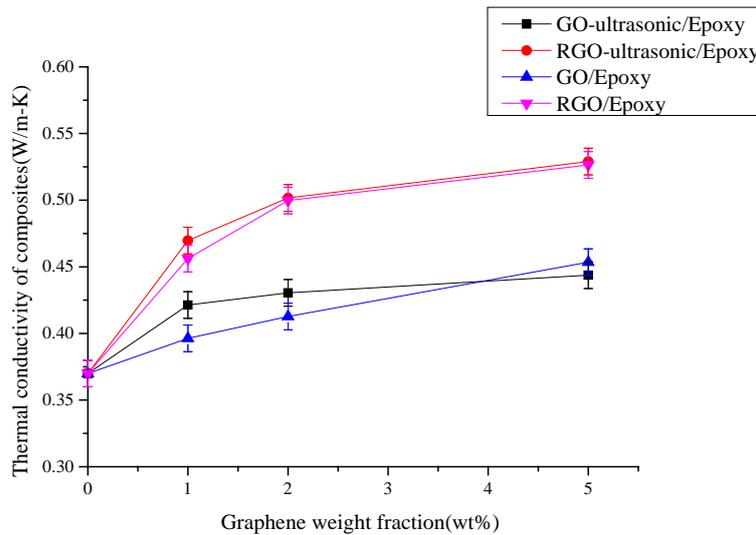


Figure 4: Effect of ultrasonication dispersion (300W/30 min) on thermal conductivity of graphene nanocomposites.

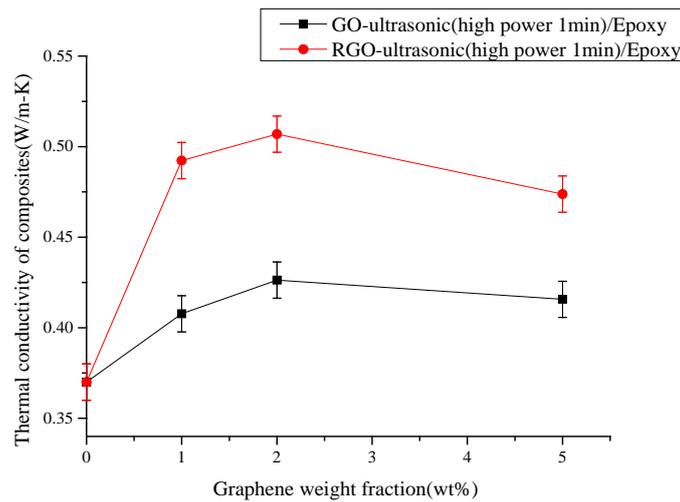


Figure 5: Effect of high power ultrasonication dispersion (1500W/1 min) on thermal conductivity of graphene nanocomposites.

Figure 6 compares effect of ultrasonication (1500 W) time from 1 to 3 minutes on thermal conductivities of both GO/epoxy and RGO/epoxy. For most composites, longer ultrasonication (1500 W) time does not result in higher TC of composites. 1 minute ultrasonication time on RGO (2.0wt%)/epoxy leads to its highest TC among six kinds of composites. For longer ultrasonication time more than 1 minute, ultrasonication vibration may cause temperature increase of graphene solution, which induces negative effect for dispersion of graphene in epoxy resin. Compared figure 6 with figure 4, only one high power ultrasonication condition (1500W/3 min) leads to higher TC of GO/epoxy than the counterpart, regular ultrasonication (300W/30 min). Figure 7 shows effect of ultrasonication dispersion (300W/30 min) on impact energy of graphene nanocomposites. Ultrasonication leads to increase of impact energy of RGO/epoxy as the graphene weight fraction increases. However, for most GO/epoxies ultrasonication does not improve their impact energy. The undulation nature of RGO may benefit the better dispersion of RGO in epoxy resin by ultrasonication. Figure 8 depicts effect of ultrasonication dispersion (300W/30 min) on impact force of graphene nanocomposites. It can be observed that the trend of impact force is in reverse compared with that in figure 7 for impact energy. For example, impact force of RGO/epoxy is higher than that of RGO-ultrasonication/epoxy. Higher impact force of RGO/epoxy infers that RGO/epoxy is more brittle than that of RGO-ultrasonication/epoxy when subjected to impact. Therefore, RGO/epoxy absorbs less impact energy than RGO-ultrasonication/ epoxy.

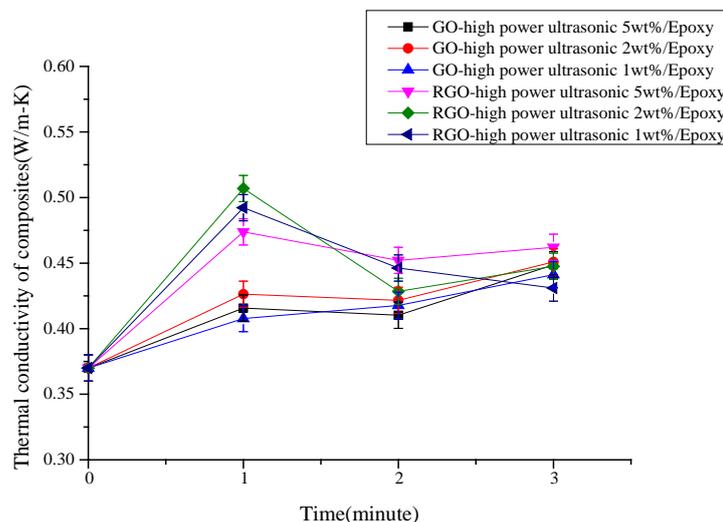


Figure 6: Effect of high power ultrasonication dispersion time on thermal conductivity of graphene nanocomposites.

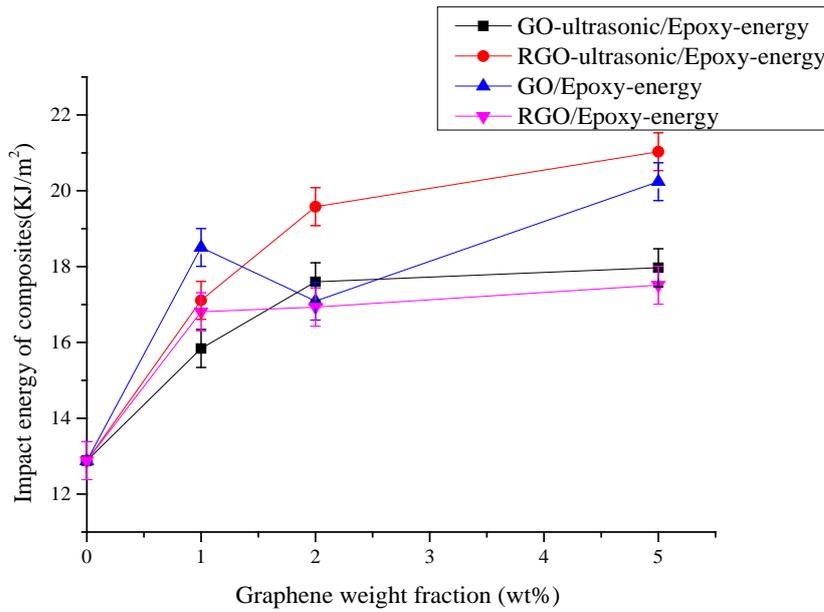


Figure 7: Effect of ultrasonication dispersion (300W/30 min) on impact energy of graphene nanocomposites.

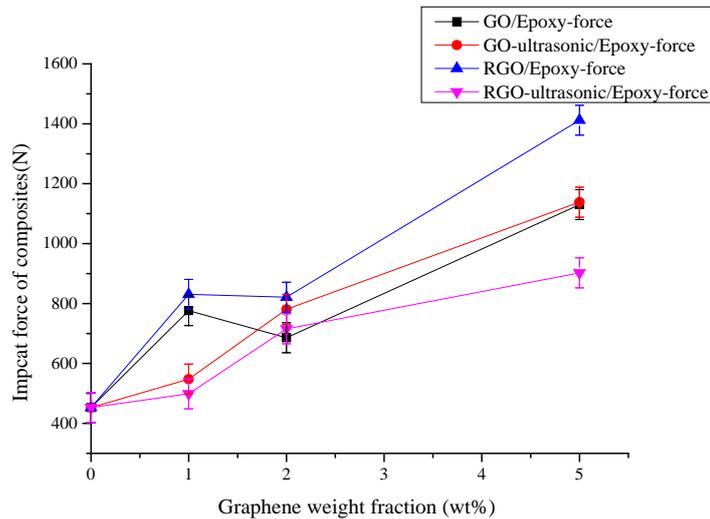


Figure 8: Effect of ultrasonication dispersion (300W/30 min) on impact force of graphene nanocomposites.

4 CONCLUSIONS

In this paper, effects of graphene type, graphene weight fraction, and ultrasonication on thermal conductivity (TC) and impact performance of graphene nanocomposites are revealed. Both TC and impact energy of composites increase with graphene weight fraction, indicating that addition of graphene does enhance both properties. RGO/epoxies all have higher TC than those of GO/epoxies due to more connecting networks among undulated RGO nanosheets. Regular ultrasonication improve TC of GO/epoxy and RGO/epoxy at lower graphene fraction. 1 minute high-power ultrasonication time applied on composites leads to improvement of TC for most composites, indicating efficiency of

this method. Regular ultrasonication significantly improves impact energy of RGO/epoxy. Among various graphene/epoxy composites, RGO(5wt%)/epoxy by ultrasonication 300W/30 min has highest thermal conductivity (TC) of 0.529 W/m-k. RGO(5wt%)/epoxy has highest impact force of 1412.08 N; while RGO(5wt%)/epoxy by ultrasonication 300W/30 min has highest impact energy of 21.03 KJ/m².

ACKNOWLEDGEMENTS

Authors are grateful for financial support from Ministry of Science and Technology in Taiwan on this work under contract MOST 105-2221-E-035-028.

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