



Superior enhancement in thermal conductivity of epoxy/graphene nanocomposites through use of dimethylformamide (DMF) relative to acetone as solvent [☆]

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ABSTRACT

This method article describes the fabrication of graphene-epoxy nanocomposites using two different solvents, dimethylformamide (DMF) and acetone, and validates the resulting thermal conductivity improvements. The study compared the two solvents at a filler composition of 7 wt% and found that DMF resulted in more uniform dispersion of graphene nanoparticles in the epoxy matrix, leading to a 44% improvement in thermal conductivity compared to acetone. Laser scanning confocal microscopy (LSCM) imaging showed that DMF-based composites had more evenly dispersed graphene nanoplatelets than acetone-based composites, which exhibited larger graphene agglomerations. Effective medium theory calculations showed that DMF led to almost 35% lower interface thermal resistance between graphene and epoxy compared to acetone. The validated fabrication method and findings provide new possibilities for developing high thermal conductivity graphene-epoxy nanocomposites for various thermal management applications.

- This article describes methods for fabricating graphene-epoxy composites using acetone and DMF as solvents, and validates that DMF is better for achieving higher thermal conductivity in the composite.

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*Method details

Introduction

Graphene, since its discovery in 2004 [1], has been thoroughly studied due to its extraordinary properties like extremely high electrical conductivity, high thermal conductivity, high mechanical strength, and modulus [2]. This study focuses on the role of solvents in enabling superior dispersion of graphene in epoxy composites for achieving higher thermal conductivity values. High thermal conductivity of graphene ($2000 - 5000 \text{ W m}^{-1} \text{ K}^{-1}$) makes it an ideal filler material for obtaining thermally conducting polymer composites [3–7]. Incorporation of graphene nanoplatelets into polymer matrix has been shown to yield significant enhancement in physical properties of polymers [8–15]. Uniform dispersion of graphene into the polymer is of utmost importance to achieve maximum improvement in properties. Uniform dispersion leads to superior thermal contact between graphene nanoplatelets and polymer matrix leading to lower thermal interface resistance between the two, thus enabling higher composite thermal conductivity. Uniform dispersion can also help in forming efficient graphene percolation networks in the polymer matrix by reducing gap between the graphene nanoparticles [8,16,17].

Commonly used polymers in engineering applications are mostly soluble in organic solvents. This makes dispersion of graphene sheets into organic-solvents, an important parameter to prepare a homogeneous composite. Graphene also cannot be simply mixed mechanically with the polymer as it tends to reaggregate due to strong Van der Waals forces between graphene sheets. Furthermore, the high viscosity of typical polymer melts prevents direct uniform dispersion of graphene into polymer. Therefore, using organic solvents compatible with the common polymers has been widely accepted and regarded as the simplest method for uniformly dispersing graphene sheets [16–18]. Such organic solvents primarily enhance the dispersion of graphene in the polymer matrix without altering any other properties.

Studies have been conducted for the effect of solvents on dispersion of graphene in regards with improvement of mechanical properties [19]. However, no results have been reported for the effect of solvents on improvement of thermal conductivity. Acetone, one of the commonly used solvent for preparing graphene-polymer composites has shown short-term stability of graphene dispersions when compared to many other organic solvents like dimethylformamide (DMF), tetrahydrofuran (THF), N-methyl-2-pyrrolidone (NMP) and ethylene glycol, which show long term stability of dispersions [20]. The improved dispersion of graphene in certain solvents compared to acetone, has been shown to also lead to improved mechanical properties such as mechanical strength. Mishra et al. compared the effect of three solvents (ethanol, acetone, and toluene) on the dispersion of polyhedral oligomeric silsesquioxane (POSS) in epoxy and the subsequent enhancement of mechanical properties of POSS-epoxy nanocomposite [21]. They found that composite prepared using ethanol showed increase in elastic modulus and fracture toughness values due to the better dispersion of POSS in epoxy resin in ethanol solvent. While the above studies addressed the effect of improved dispersion in certain solvents on mechanical properties like mechanical strength, the present study focuses on the effect of solvents on thermal conductivity.

Wang et al. prepared graphene nanoplatelet/epoxy samples using acetone with $0.45 \text{ W m}^{-1} \text{ K}^{-1}$ thermal conductivity at 5wt% loading [22]. Guo et al. demonstrated a thermal conductivity close to 0.3 and $0.45 \text{ W m}^{-1} \text{ K}^{-1}$ for graphene/epoxy composite samples made with acetone at 5wt% and 10wt% respectively [23]. Han et al. showed thermal conductivity of $0.33 \text{ W m}^{-1} \text{ K}^{-1}$ for epoxy/graphene nanoplatelets at 4wt% loading prepared using acetone [24]. One finds multiple such studies for thermal conductivity of graphene/epoxy composites prepared using acetone. There are, however, no reported studies on thermal conductivity of such composite samples prepared using DMF and this work aims to fill that gap and provide a more efficient way to make high thermal conductivity graphene/epoxy composites.

The dispersion capability of a solvent can be inferred from their ‘Hansen Solubility Parameters’ (HSP). These parameters include a dispersion cohesion parameter (δ_d), polarity cohesion parameter (δ_p) and a hydrogen bonding cohesion parameter (δ_h) [25]. Park et al. showed that highly reduced graphene (HRG) was well dispersed in solvent mixtures (DMF/H₂O mixed with either acetone, acetonitrile, THF, DMF, NMP, DMSO and ethanol) having the sum $\delta_p + \delta_h$ in the range of 13–29. They further showed that the solvent having the sum lower than 10 (DMF/H₂O mixed with either DCB, diethyl ether and toluene) and higher than 30 (water) exhibit poor dispersion of HRG [26]. The HSP sum ($\delta_p + \delta_h$) of acetone and DMF are 17.4 and 25 respectively, implying they both are viable candidates for dispersing HRG [27].

Another important parameter related to dispersion is the zeta potential (ζ). Zeta potential is the electric potential difference between the dispersion medium and the stationary layer of fluid attached to the dispersed particle [28]. It is denoted using a numerical value with a positive or negative sign. A strong positive or negative zeta potential result in high repulsive forces between the dispersed particles, indicating good stability of dispersions [29]. Ghazvini et al. have shown that the zeta potential for graphene in acetone is -22.6 mV [30]. S. Gambhir et al. have shown that graphene in different forms has zeta potential lower than -30 mV in DMF [31]. These values suggest a stronger negative zeta potential of graphene in DMF which leads to better stability of the dispersion over time. Villar-Rodil et al. have demonstrated stable suspension of unreduced and chemically reduced graphene in DMF over several months [18].

In general, there are many methods to improve the thermal conductivity of polymer composite. Some of these methods include graphene functionalization [32,33] (to enhance thermal interface interactions), graphene alignment [34] (to enhance in-plane thermal conductivity) and graphene dispersion (to reduce thermal interface resistance due to agglomeration). The present study is conducted to enhance polymer composite thermal conductivity by improving the graphene dispersion. Three graphene concentrations- 3 wt%, 5 wt% and 7 wt% are studied. The dispersion effect is characterized using laser scanning confocal microscopy which provides optical images of the graphene sheets dispersed in the polymer matrix, enabling a visual comparison of achieved dispersion.

Materials

Graphene nanoplatelets were purchased from Graphene Supermarket. Characterization of graphene is provided in the supplementary information. Epoxy resin used for the study was EPIKOTE RESIN MGS RIMR 135 and the hardener used was EPIKURE CURING AGENT MGS RIMH 137, both purchased from Hexion. The organic solvents N-N, Dimethylformamide (DMF) and Acetone were purchased from the University of Oklahoma chemical stock room.

Epoxy-graphene composite fabrication methods

Preparation of epoxy/graphene composite with acetone

0.15, 0.25 and 0.35 g (corresponding to 3 wt%, 5 wt% and 7 wt% compositions) of graphene nanoplatelets (3–7 nm thick and ~5 μm in lateral size) were dispersed in 80 mL acetone and tip sonicated for one hour in an ice bath to prevent heating and evaporation of acetone. The resin was then added to the solution and tip sonicated in an ice bath for another two hours. After sonication, the solution was heated to 80 °C and stirred continuously using a mechanical mixer to remove the solvent. The mixture of graphene and epoxy was weighed until the required weight was reached related to removal of acetone. This mixture was then spread on a PTFE sheet and placed in a vacuum oven at 65 °C for 15 h to further ensure complete removal of acetone. The hardening agent was then added to the composite mixture and then transferred into aluminum molds and cured at 90 °C for 20 h.

Preparation of epoxy/graphene composite with DMF

0.15, 0.25 and 0.35 g of graphene nanoplatelets (3–7 nm thick and ~5 μm in lateral size) were dispersed in 80 mL DMF and sonicated in an ice bath for 30 min. Epoxy resin was then added to the solution and sonicated for another one hour. After sonication, the solution was heated and stirred at 150 °C to remove the solvent. The graphene-epoxy mixture was then spread on a PTFE sheet and placed in a vacuum oven at 140 °C for 15 h to ensure complete removal of the DMF. The hardening agent was then added to the mixture and then transferred into aluminum molds and cured at 90 °C for 20 h. Fig. 1 shows the schematic of the sample preparation process.

Characterization methods

Thermal conductivity of the composite samples was calculated using the thermal diffusivity values measured using the NETZSCH LFA 467 which works on the principle of laser flash analysis. In this process, a light pulse beam heats the lower surface of the sample and the temperature increase on the upper surface of sample is measured using an infrared detector. The temperature rise on the upper surface of sample is recorded as a function of time. Thermal diffusivity is then calculated using the equation:

$$\alpha = 0.1388 \frac{d^2}{t_{\frac{1}{2}}} \quad (1)$$

where α is the thermal diffusivity, d is the sample thickness and $t_{\frac{1}{2}}$ is the time taken to reach the half of the maximum temperature. Using this thermal diffusivity, the thermal conductivity is calculated using the equation

$$k = \alpha \rho C_p \quad (2)$$

where k is the thermal conductivity, ρ is the density and C_p is the specific heat of the sample respectively.

Method validation

Thermal conductivity measurements

Fig. 2 shows the thermal conductivity values of the graphene-epoxy composites with 3wt% (1.54 vol%), 5wt% (2.6 vol%) and 7wt% (3.68 vol%) concentration for the two organic solvents - acetone and DMF. Thermal conductivity of pure epoxy sample is measured to be 0.17 $\text{Wm}^{-1}\text{K}^{-1}$. At 3wt%, DMF and acetone samples show identical thermal conductivity value of 0.34 $\text{Wm}^{-1}\text{K}^{-1}$. However, at higher concentrations, DMF-based composite samples show significantly higher thermal conductivity values relative to acetone-based composite.

The solvent thus has a clear effect on thermal conductivity of epoxy-graphene nanocomposites. This enhancement in thermal conductivity at higher concentrations can be explained in terms of improved dispersion of graphene at higher concentrations in the epoxy composite prepared with DMF (discussed below).

Confocal microscopy imaging for dispersion characterization

The dispersion effect of the two solvents was characterized using confocal microscopy imaging. Confocal microscopy or laser scanning confocal microscopy (LSCM) is an optical imaging technique. It uses a spatial pinhole to block out the out of focus light

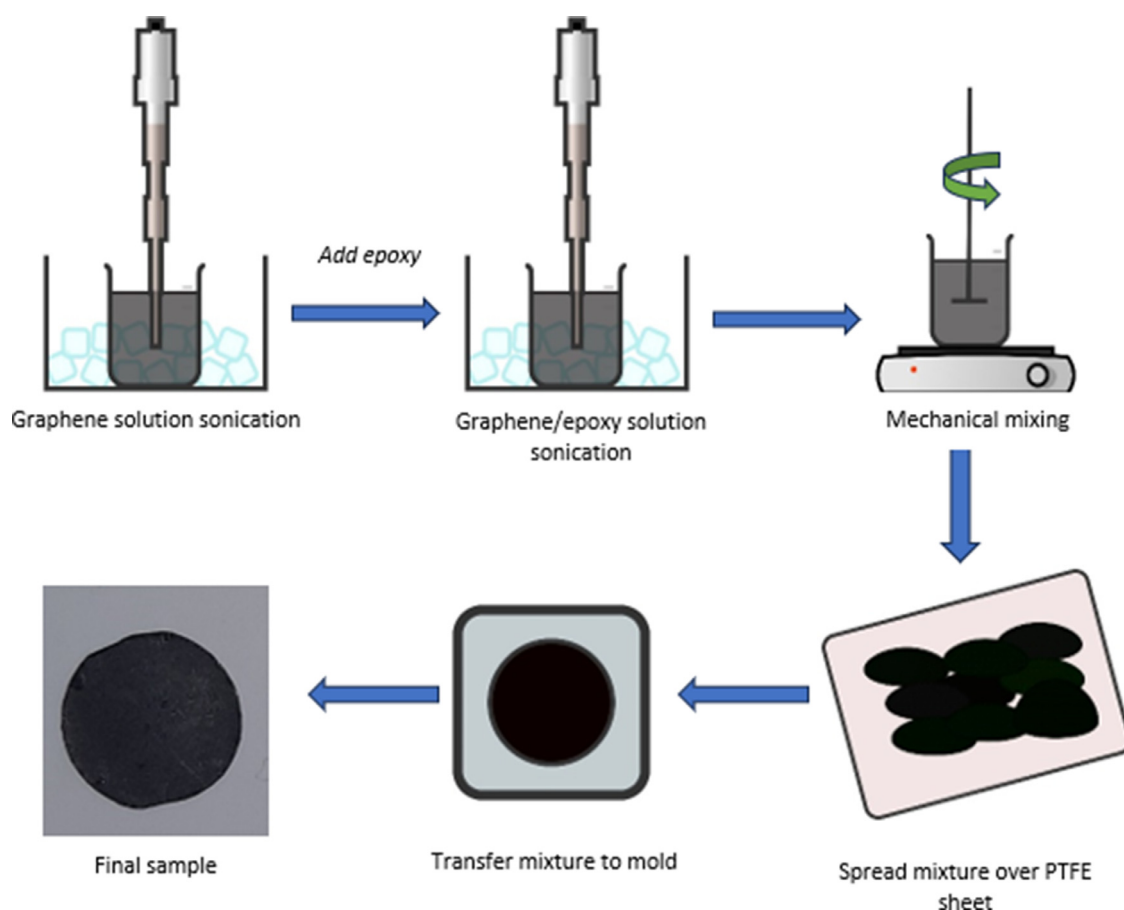


Fig. 1. Schematic for sample preparation.

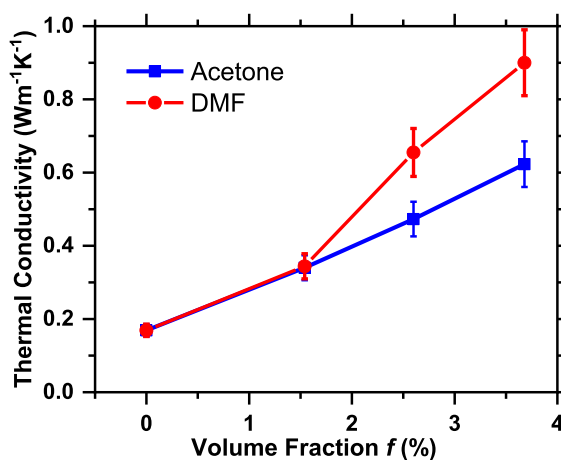


Fig. 2. Thermal conductivity of graphene/epoxy composites prepared using two different solvents, with increasing graphene concentration.

which aids in increasing the optical resolution of a micrograph. LSCM generate images with lesser haze and better contrast than a conventional microscope and can be focused on a thin cross-section of a sample [34–36]. It can also be used to generate a three-dimensional image of the sample. This makes LSCM an effective tool to visualize the dispersion of graphene within the composite samples. Fig. 3 shows the LSCM images of the graphene-epoxy composites with 3 wt%, 5 wt% and 7 wt%. There is a clear visible difference in the dispersion of graphene in the epoxy composites prepared using DMF and acetone solvents. Acetone based samples

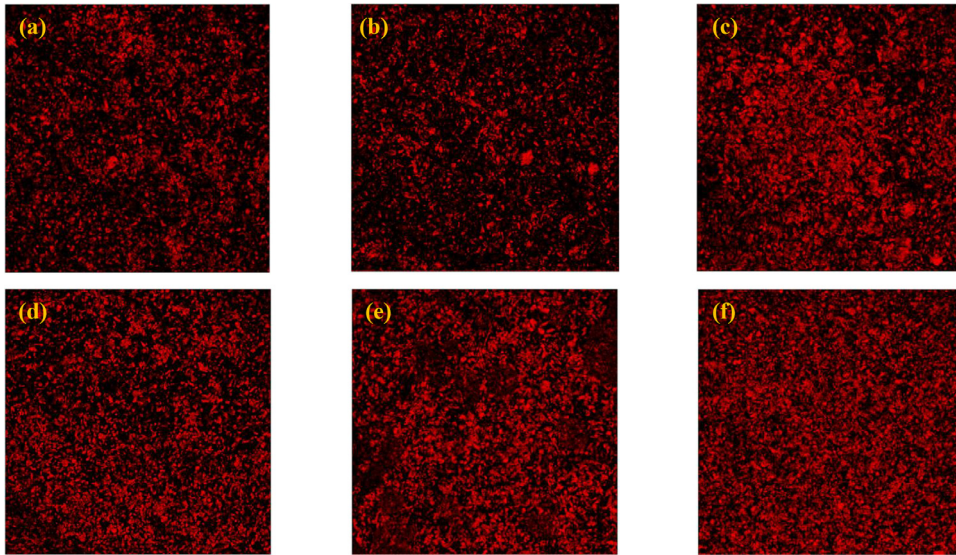


Fig. 3. Confocal microscopy images for acetone-based samples for concentrations (a) 3wt%, (b) 5wt%, (c) 7wt% and DMF-based samples for concentrations (d) 3wt%, (e) 5wt%, (f) 7wt%.

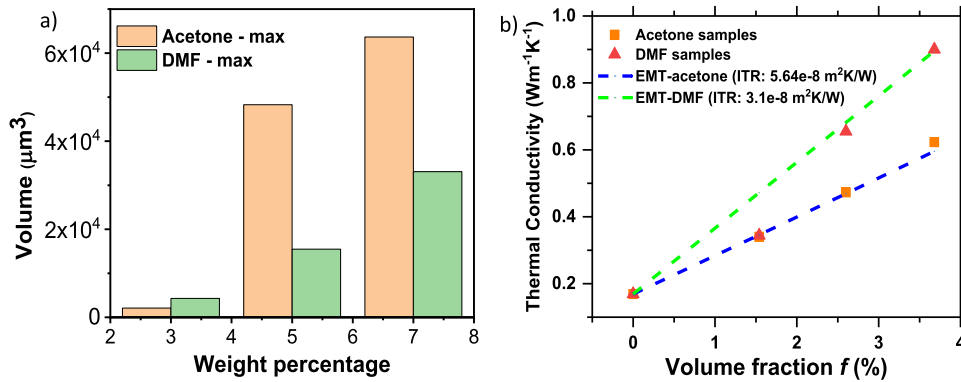


Fig. 4. (a) Maximum graphene agglomeration volume comparison, (b) Effective medium theory and measured thermal conductivity values.

(Fig. 3a–c) show larger gaps between graphene agglomerates indicating poor dispersion. Composites prepared using DMF, however, show relatively more uniform distribution of graphene sheets leading to higher thermal conductivity (Fig. 3d–f).

Fig. 4 shows the maximum graphene agglomerate size comparison between the acetone and DMF based samples. The calculation of agglomerate size was performed using the Fiji (also known as ImageJ) image analysis software. The confocal microscopy images were analyzed for 3d particle volumes using the software. Results show that at 3wt% the agglomeration sizes are identical which explains the similar thermal conductivity values at this concentration. At higher concentrations, agglomerate size in acetone-based samples is 211% and 93% higher at 5wt% and 7wt% respectively. Such high graphene agglomerate volumes in acetone-based samples explains the high thermal interface resistance leading to lower thermal conductivity.

Effective medium theory

The effect of graphene dispersion on the thermal conductivity of graphene-epoxy nanocomposite was theoretically studied using the effective medium theory presented by Nan et al. [34,37]. The theory focuses on the interfacial thermal resistance between the dispersed particles and the matrix material. Interfacial thermal resistance is an essential parameter in determining the thermal conductivity of any composite material. Using the theory presented by Nan et al. and the measured thermal conductivity values, we calculate the interface thermal resistance for acetone and DMF based samples.

The theoretical effective thermal conductivity of the graphene-epoxy composites is given by

$$k_{effective} = k_m \frac{2 + f[\beta_{11}(1 - L_{11})(1 + \langle \cos^2 \theta \rangle) + \beta_{33}(1 - L_{33})(1 - \langle \cos^2 \theta \rangle)]}{2 + f[\beta_{11}L_{11}(1 + \langle \cos^2 \theta \rangle) + \beta_{33}L_{33}(1 - \langle \cos^2 \theta \rangle)]} \quad (3)$$

where $k_{effective}$ and k_m are the effective thermal conductivities of composite and pristine epoxy matrix respectively, for f volume fraction of graphene nanoparticles. The $\langle \cos^2 \theta \rangle$ term considers the orientation of the filler material. For the present scenario, a random orientation of graphene nanoparticles is considered ($\langle \cos^2 \theta \rangle = 1/3$).

The geometrical parameters of the oblate graphene nanoparticles such as L_{ii} are obtained by using the following equations (p is the aspect ratio),

$$L_{11} = L_{22} = \frac{p^2}{2(p^2 - 1)} + \frac{p}{2(1 - p^2)^{3/2}} \cos^{-1} p \quad (4)$$

$$L_{33} = 1 - 2L_{11}$$

In Eq. (3), β_{ii} is computed using the equation:

$$\beta_{ii} = \frac{K_{ii}^c - k_m}{k_m + L_{ii}(K_{ii}^c - k_m)} \quad (5)$$

where K_{ii}^c are the effective thermal conductivity values of the graphene nanoparticles considering the effect of thermal interface resistance. The in-plane effective values K_{11}^c , K_{22}^c and the through-plane effective value K_{33}^c are given by equations:

$$K_{11}^c = K_{22}^c = \frac{k_{pi}}{1 + \gamma L_{11} k_{pi} / k_m} \quad (6)$$

$$K_{33}^c = \frac{k_{pt}}{1 + \gamma L_{33} k_{pt} / k_m} \quad (7)$$

$$\gamma = (1 + 2p)\alpha \quad (8)$$

$$\alpha = \frac{Rk_m}{t} \quad (9)$$

where R is the thermal interface resistance and k_{pi} and k_{pt} are the in-plane and through-plane thermal conductivities of graphene, respectively.

Fig. 4(b) shows the effective medium theory results. Using the effective medium theory and measured thermal conductivity data, respective interface thermal resistance resistances for the acetone and DMF based samples were calculated. Results show that the thermal interface resistance for acetone-based samples is 82% higher than the DMF based samples. This difference in the thermal interface resistance is key in understanding the higher thermal conductivity of the DMF based samples.

Summary

Dispersion of graphene into epoxy matrix and its effect on the thermal conductivity of the graphene-epoxy composite was investigated for two organic solvents - acetone and DMF. Laser Scanning Confocal Microscopy was used to obtain optical images of the composite samples to visualize the dispersion of graphene in the epoxy matrix. In comparison to the DMF-based samples, acetone-based samples showed poor dispersion with agglomeration of graphene visible at various locations. Improved dispersion of graphene in composites prepared with DMF as solvent was found to lead to 40% and 44% higher thermal conductivity in DMF based samples than the acetone-based samples at 5wt% and 7wt% respectively. These results provide new pathways for developing graphene-epoxy nanocomposites with high thermal conductivity.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Swapneel Danayat: Conceptualization, Methodology, Validation, Writing – original draft. **Avinash Singh Nayal:** Methodology, Validation, Data curation. **Fatema Tarannum:** Methodology, Validation, Data curation. **Roshan Annam:** Methodology, Validation, Data curation. **Rajmohan Muthaiah:** Writing – review & editing, Supervision. **Madhan K. Arulanandam:** Writing – review & editing. **Jivtesh Garg:** Supervision, Funding acquisition, Conceptualization, Methodology, Writing – original draft, Writing – review & editing.

Data availability

Data will be made available on request.

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