




# Graphene Thermal Interface Materials – State-of-the-Art and Application Prospects

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**ABSTRACT** We provide a summary of the fundamentals of thermal management, outline the state-of-the-art in the field of thermal interface materials, and describe recent developments in graphene-based non-curing and curing composites used for thermal management. The discovery of unique heat conduction properties of graphene and few-layer graphene motivated research activities worldwide focused on creating efficient graphene-based thermal interface materials. While the initial focus of these studies was on obtaining the maximum possible thermal conductivity of the composites, recently the attention has shifted to practical problems of minimizing the thermal contact resistance at interfaces, optimizing the size distribution of graphene as filler, and addressing the issues of scalability, stability, and production costs at commercial scales. We conclude the review with a general outlook for commercial applications of graphene in the thermal management of electronics.

**INDEX TERMS** Composites, graphene, noncured, cured, polymer, power electronics, thermal interface materials, thermal conductivity, thermal contact resistance.

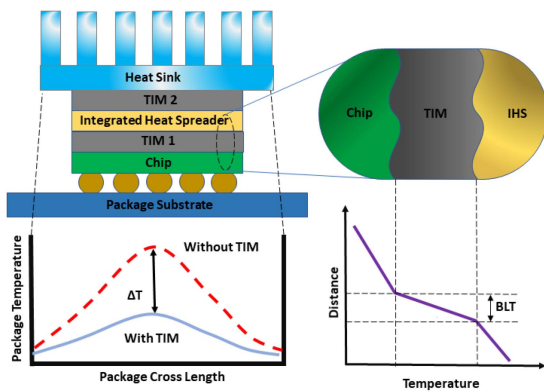
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## I. INTRODUCTION

Advances in modern electronics led to substantially higher transistor density [1], which paved the way for a drastic increase in functionality and, at the same time, the need for better thermal management technologies [2], [3]. The emergence of power electronic devices and their miniaturization [4], [5], [6], [7], [8] also motivate the search for new methods of handling high-generated power [9], [10]. Over the past few years, electronic devices have seen an increase in device instability due to an increase in operating temperature [4], [11], [12], [13], [14]. A study, conducted in 2019, showed a significant increase in the power density in electronics [15] and correlated it with the increasing number of devices integrated into a smaller area [16], [17], [18]. Effective management of heat dissipation in electronic devices is vital for continuing progress in electronics. Although humidity, dust, and vibration can lead to device failure, their influences are relatively minor whereas

an increase in temperature contributes to ~55% of device failures and thus limits technology improvements [15], [19], [20].

The utilization of an effective thermal interface material (TIM) with high thermal conductivity and low thermal contact resistance is important for managing excess heat in electronic devices. Generally, an electronic device with a solid heat sink suffers from thermal interface resistance in the junction. The layers of TIMs are applied between a device and a heat sink where otherwise only 2% or less of the overall surface area is in actual contact [3], [21], [22], [23] (see Fig. 1). The reduced contact surface area between the two solid surfaces is mostly due to the uneven surface roughness at the contacting interfaces [24], [25], [26], [27], [28]. The cavity formed between the two surfaces is filled with air, which has a low thermal conductivity of ~0.026 W/mK at room temperature (RT) [29]. Utilizing a TIM with higher thermal conductivity and low contact resistance improves the thermal management



**FIGURE 1.** Schematic of the use of thermal interface materials in the electronic device packaging.

of electronic devices. The development of TIMs that can provide a lower thermal resistance is an important part of the thermal management approaches for effective heat removal in electronic devices [4], [19], [21], [26], [30], [31], [32].

The performance of the TIM introduced between two uneven surfaces can be evaluated using the equation for the total thermal resistance,  $R_{total}$  [3], [26], [32], [33], [34], [35]:

$$R_{total} = \frac{BLT}{K_{TIM}} + R_{C1} + R_{C2}. \quad (1)$$

Here, the bulk thermal conductivity of the TIM is denoted by  $K_{TIM}$ , and the thermal contact resistance of the two surfaces with TIM is represented by  $R_{C1}$  and  $R_{C2}$ , while the thickness of the TIM layer, also known as the bond line thickness, is denoted by  $BLT$ . The thermal resistance of the TIM layer,  $R_{TIM}$ , is  $BLT/K_{TIM}$  and if the TIM layer is compressed between two identical surfaces, then  $R_C = R_{C1} = R_{C2}$ , which allows one to simplify (1) to:

$$R_{total} = R_{TIM} + 2R_C. \quad (2)$$

The total thermal resistance,  $R_{total}$ , can be reduced by minimizing  $R_C$  and  $BLT$ , which depend on the properties of TIM and the roughness of the connecting surfaces. For high-power-density electronic devices, in modern electronics, the  $BLT$  varies from 15 to 100  $\mu\text{m}$  [4]; typically, larger  $BLT$  is required to fill the voids in high-roughness surfaces. This can be particularly relevant for polycrystalline diamond, used as a substrate or active layer [36], [37]. Thermal management requires TIM with both high thermal conductivity and low thermal contact resistance.

The TIM performance can be improved by using a suitable base polymer with incorporated thermally conductive fillers. In developing TIMs one should keep in mind the existence of the Kapitza resistance, between the polymer and the fillers [38], [39], [40], [41], [42], [43], [44], [45]. There are other performance parameters of TIMs that need to be taken into consideration for practical applications. The electrically insulating and mechanical properties as well as reliability and cost effectiveness play major roles. The most commonly used types of TIMs include non-cured, cured, thermal pads, and

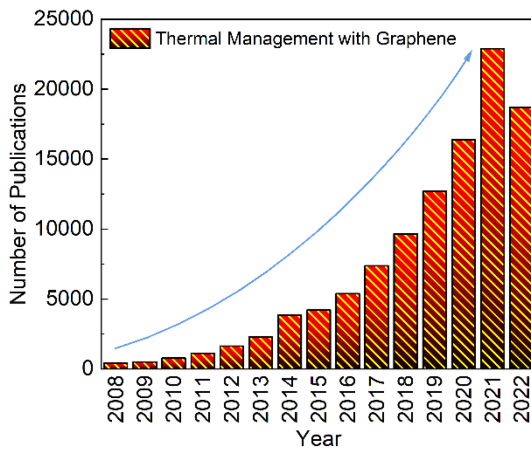
phase change materials. A non-cured TIM is effective in replacing excess air from the structure by filling the voids formed between the two uneven surfaces. The viscosity of the non-cured TIM influences the  $BLT$  and the amount of TIM, which leaks out of the structure, through the process known as pump-out [29], [32], [35], [46], [47]. Commercial TIMs have thermal conductivity, which is no longer sufficient for many modern electronic devices [35], [48], [49], [50], [51], [52], [53], [54], [55]. New TIMs are required for various application domains [4].

## II. GRAPHENE AND FEW-LAYER GRAPHENE AS FILLERS FOR TIMS

Graphene attracted attention by revealing unusual electrical and optical properties [56], [57]. Raman spectroscopy was instrumental in verifying the quality of graphene and determining the number of atomic planes in few-layer graphene (FLG) [32], [58], [59]. The G-peak of graphene is narrow and sensitive to temperature [59], [60]. The unique heat conduction properties of graphene and FLG were discovered with the help of Raman spectroscopy, using the optothermal Raman technique, [61] and later examined further by various other experimental and theoretical studies [62], [63], [64], [65], [66], [67], [68], [69], [70]. The thermal conductivity of high-quality single-crystal graphene is measured to be in the range of  $\sim 2000 \text{ W/mK} - 5000 \text{ W/mK}$  near RT [61], [62], [64], [65], [66]. The thermal transport in FLG is influenced strongly by the lateral dimension and thickness [63], [71], [72], [73], [74], [75], [76], [77], [78]. Overall, FLG retains the excellent in-plane heat conduction properties of graphite while being more flexible owing to its small thickness.

Although single-layer graphene (SLG) has a superior intrinsic thermal conductivity, the use of FLG as fillers for TIMs is more promising for practical applications. This is because FLG has a larger cross-sectional area, less degradation when mixed with a base material matrix, retainable mechanical stability with the base matrix, and low cost of mass production [71], [79]. Initial studies of graphene-FLG filler-cured epoxy TIMs with random orientation of low-loading fillers demonstrated enhanced thermal conductivity by a factor of  $\times 25$  [33], [80]. A similar enhancement in thermal conductivity was also seen in non-cured graphene-FLG composites [26], [32], [35], [81], [82]. Incorporation of graphene fillers in a base polymer increased the thermal conductivity to  $\sim 12.5 \text{ W/mK}$  for cured TIMs [83], [84] and  $\sim 7.1 \text{ W/mK}$  for non-cured TIMs [35], which surpassed the values of commercially available TIMs [35]. When measured in thin-film form by a standard TIM Tester method, the commercial TIMs typically have the thermal conductivity values between  $1 \text{ W/mK}$  to  $6 \text{ W/mK}$ . Utilization of the proper base polymer matrix, which couples well with the fillers, helps to increase the overall thermal conductivity of graphene TIMs. This has also been shown to have a positive influence on the thermal performance of such graphene-FLG filler TIMs throughout power cycling [85].

Graphene-FLG mixtures are preferred over carbon nanotubes (CNTs) as fillers for TIMs. In contrast to CNTs, FLG

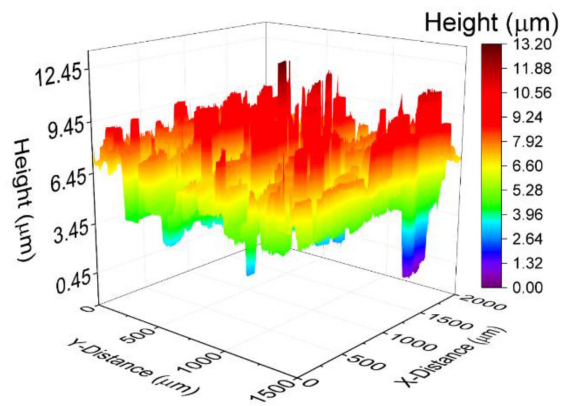


**FIGURE 2.** Statistics of the publications on thermal management with graphene during 2008 - 2022 (Source: Google Scholar).

does not exhibit severe agglomeration issues during mixing processes yielding a better overall thermal performance [74]. Obtaining graphene fillers by synthesis using the technique of liquid-phase exfoliation (LPE) is cheap and can easily be scaled up [32], [86], [87], [88], [89], [90], [91]. The process of LPE can be performed by breaking down the van der Waals (vdW) bonds between graphite with compatible solvents and sonication to obtain FLG. The graphene fillers for composites and TIMs can also be produced by electrochemical exfoliation [92]. This technique utilizes bulk graphite and interaction between electrodes and solvent ions to produce FLG. There are multiple studies on graphene-FLG filler TIMs for various applications [5], [26], [32], [33], [34], [35], [40], [42], [64], [80], [81], [82], [83], [84], [85], [93], [94], [95], [96], [97], [98], [99], [100] and composites with dual fillers including graphene-FLG [38], [40], [80], [98], [100], [110], [111], [112], [113], [114], [115], [116], [117], [118], [119], [120], [121], [122], [123], [124], [125], [126], [127], [128]. Since the discovery of the thermal conductivity of graphene in 2008, multiple studies have been published on thermal management with graphene. Fig. 2 illustrates the interest in this research topic using the number of publications as an indicator.

### III. INFLUENCE OF SURFACE ROUGHNESS ON THE PERFORMANCE OF TIMS

In the thermal management of electronics, one often looks for the best TIM based on thermal conductivity and cost. However, the selected TIM may not be the best choice for specific applications and the materials of the electronic device and heat sink. The bulk thermal conductivity of the TIM layer is an important factor of performance, but one should also address the issues of conformability and spreading of TIMs on particular surfaces. Utilizing a TIM with high thermal conductivity and low thermal contact resistance (TCR) is crucial for the thermal performance of TIM and, ultimately, the extended life of an electronic device [23], [26], [85], [129], [130], [131], [132], [133]. The parameter of total thermal resistance,  $R_{total}$  can be calculated by measuring the contact resistance of the



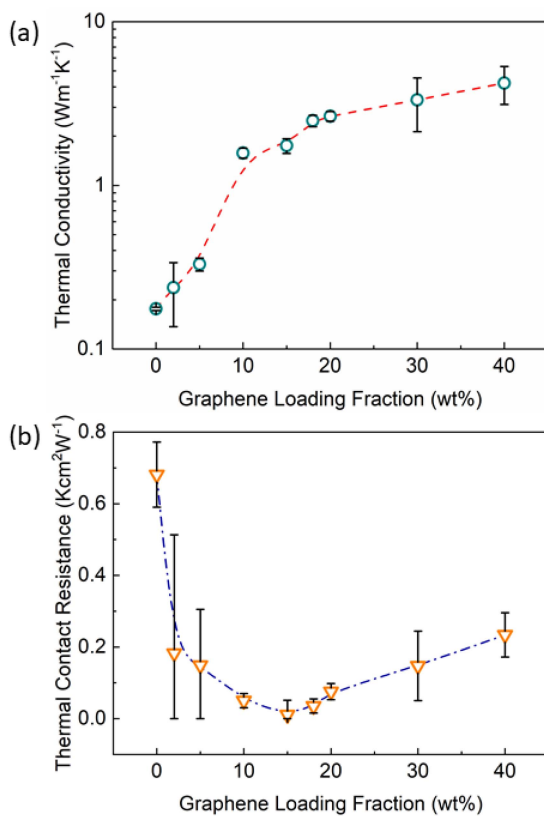
**FIGURE 3.** RMS surface roughness of polished 1 in<sup>2</sup> copper plates determined by optical profilometer for one of the used copper plates after polishing for ~1 minute with  $S_q = 1.2 \mu\text{m}$ . Reproduced from Ref. [26] under CC-BY-4.0.

TIM layer with the mating surfaces,  $R_C$  as expressed in (1) and (2).

Multiple studies have been conducted with graphene TIMs [26] and other conductive filler TIMs [27], [28], [134], [135], [136] while varying the surface roughness of the material to understand the correlation of TCR with the surface roughness. Some of us performed a detailed investigation of TCR of non-curing TIMs with graphene fillers with surfaces characterized by different roughness [26]. The study used PDMS and silicone oil as the base polymer and square copper plates with an area of 1 in<sup>2</sup> and a thickness of ~1 mm as the connecting surfaces. The surface roughness of the plates was adjusted using a polisher and silicon carbide sandpaper to produce copper plates of different roughness. To determine the root mean square (RMS) roughness,  $S_q$ , an optical profilometer was used. Fig. 3 illustrates the  $S_q$  roughness of the copper plates determined with the optical profilometer, following the standard procedures [26], [137].

Graphene-FLG fillers with an average lateral dimension,  $L$ , of ~25  $\mu\text{m}$  were added to the base polymer and acetone was used to avoid agglomeration during the shear mixing process at low speed. The final non-cured TIMs, at different filler loadings,  $f$ , were then experimentally tested for their thermal properties with an ASTM-D5470 TIM tester at ~80 °C and ~80 psi pressure using plastic shims to acquire data at different thicknesses.

Fig. 4(a)–(b) illustrates the thermal performance of the graphene-FLG non-cured TIMs. The thermal conductivity of the graphene-FLG TIMs at different  $f$  is shown in Fig. 4(a). The thermal conductivity of silicone oil itself was measured to be 0.18 W/mK, in agreement with literature [138], [139], [140]. At low graphene loading, TIMs are below the percolation threshold [83]. As the loading increases, one observes a transition to thermal transport in the percolated regime. The specific dependence of the thermal conductivity on the loading can differ for the cured and non-cured TIMs. In curing epoxy TIMs the thermal conductivity shows super-linear trends after percolation is reached. In non-curing TIMs,



**FIGURE 4.** Thermal performance of non-curing TIMs with graphene-FLG as fillers. (a)  $K_{TIM}$  of TIMs as a function of  $f$ . (b) TCR of TIMs shows a non-monotonic response with an increase in  $f$ . Reproduced from Ref. [26] under CC-BY-4.0.

the thermal conductivity reveals a sub-linear increase, with a significant enhancement at first as fillers are added as a result of thermal percolation. The thermal conductivity does not show significant enhancement as more fillers are introduced, possibly due to agglomeration or the dominance of filler-polymer and filler-filler contact resistance in thermal transport characteristic of the TIM. The slow increase in the thermal conductivity after  $f \sim 10$  wt.% is consistent with the previously studied non-cured graphene-FLG TIMs [35]. The variations in this trend in different graphene-FLG filler composites have been studied in detail [33], [34], [83], [84], [85], [98], [99], [100]. This can also be observed with soft and nano-fluid TIMs [31], [141], [142], [143].

Fig. 4(b) shows the TCR of non-cured TIMs with graphene-FLG filler. The measured  $R_C$  reveals a non-monotonic trend as  $f$  increases. Theoretically, the thermal contact resistance,  $R_C$ , would depend on  $K_{TIM}$  and shear modulus of the TIM and the surface roughness of the mating surfaces and the pressure applied. Here,  $R_C$  is affected by both shear modulus and  $K_{TIM}$  where an increase in bulk thermal conductivity would decrease TCR whereas an opposite dependency can be seen for shear modulus [31]. Although one wants to improve  $K_{TIM}$  by increasing  $f$  to achieve the lowest  $R_{Total}$ , special attention must be given to viscosity. The increase in  $f$  affects both viscosity and the minimum attainable  $BLT$ .

#### IV. THE ROLE OF THE LATERAL DIMENSION OF THE FILLERS ON THE PERFORMANCE OF TIMS

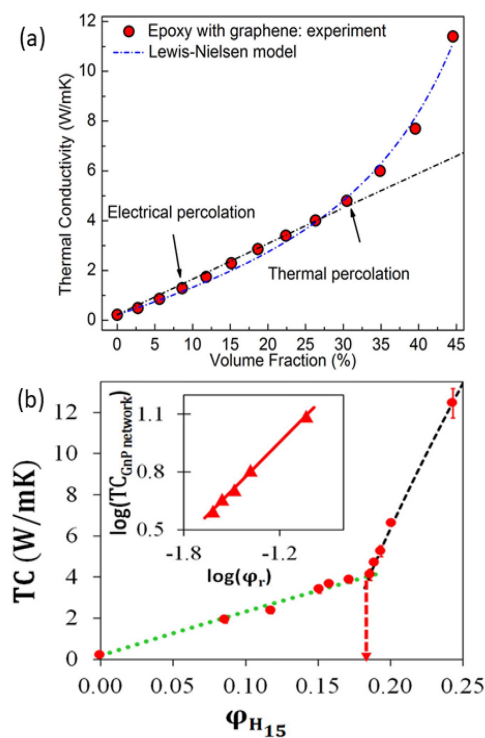
Multiple studies of graphene filler TIMs focus on  $K_{TIM}$  and the thermal diffusivity of the prepared composites as a function of  $f$  [71], [73], [74]. Fewer studies focus on fundamental questions such as the existence of thermal percolation (similar to electrical percolation) [83], [144] and synergistic effects, when using various fillers or fillers of the same type with different size distributions [100], [111], [145], [146], [147]. For the electrically conductive filler composites, it is well documented that the electrical percolation threshold depends on the filler aspect ratio and size [148], [149], [150]. A limited number of studies on the influence of  $L$  on the thermal conductivity of synthesized graphene filler composites have been documented [144], [151], [152], [153]. Some of us reported a detailed investigation of the influence of  $L$  on  $R_C$  in the context of graphene TIMs [32]. The scarcity of such data can be attributed to the difficulty in producing graphene fillers with consistent average  $L$  to synthesize the required composites.

The gray mean-free path (MFP) of graphene is known to be  $\sim 800$  nm [63], [72], [75], [76], [77], [78], [154]. This suggests that the lateral dimensions of graphene – FLG fillers should be larger than the MFP value to preserve the intrinsic heat conduction properties of the fillers. On the other side, technological considerations, e.g., bending and rolling of fillers, should also be considered as these processing hassles deteriorate the intrinsic thermal properties of FLG. Apart from the influence of  $L$  on the thermal conduction of composites, other key factors are the specific surface area, defect density [62], [110], [112], [155], Kapitza resistance between filler-matrix [156] and interface area between filler-matrix [157], [158], [159], [160].

Fig. 5(a)–(b) illustrates the thermal conductivity and thermal percolation in graphene TIMs based on the average size of the graphene – FLG fillers. Fig. 5(a) shows the thermal conductivity of cured epoxy-based graphene filler composites with  $L$  ranging from  $\sim 2$ –8  $\mu\text{m}$ . An increase in the thermal conductivity can be seen as more fillers are introduced into the base polymer matrix and a clear thermal percolation can be observed at  $f \sim 30$  vol%. The results were also confirmed with the help of the semi-empirical Lewis-Nielsen (L-N) model [161], [162]. The L-N model accounts for the packing of fillers, the shape of the fillers, and the orientation of the particles with respect to the heat flux. Fig. 5(b) represents the thermal conductivity obtained for cured epoxy-based graphene filler composites with  $L \sim 20 \pm 7$   $\mu\text{m}$ . The increase in thermal conductivity can be seen as filler content increases and the thermal percolation threshold was experimentally obtained at  $f = 17$  vol%. The obtained results can also be theoretically interpreted with the Nans' model [163], [164], [165]. Table 1 summarizes the thermal conductivity obtained by multiple research groups which helps to illustrate the influence of  $L$  on graphene-FLG filler composites. The acronyms used in the table are GNP–graphene nanoplatelets, SBR–Styrene butadiene, DGEBA–Bisphenol-A diglycidyl ether, GN sheet–graphene nanosheet, EGNP–Expanded

**TABLE 1. Thermal Conductivity of Graphene Composites**

Graphene Filler Type	Average Filler Lateral Dimension, $L$ ( $\mu\text{m}$ )	Polymer Matrix	Composite Type – Cured/ Non-Cured (C/NC)	Filler Loading		Thermal Conductivity ( $\text{Wm}^{-1}\text{K}^{-1}$ )	Reference
				wt%	vol%		
GNP	nm- $\mu\text{m}$	Silver Epoxy	C	-	5	9.9	[38]
GNP	25	Epoxy	C	-	30	9.3	[85]
GNP	2-8	Epoxy	C	-	45	12	[83]
GNP	0.05-0.5	Epoxy	C	-	10	5.1	[33]
GNP	15	Cyanoacrylate	C	-	0.5	0.35	[166]
GNP	-	SBR Rubber	C	-	15	2.922	[167]
GNP	2-8	Epoxy	C	-	43.6	7.9-9	[98]
GNP	1.5-10	Epoxy	C	50	-	8	[99]
GNP	2-8	Epoxy	C	50	-	3.2	[99]
GNP	3-7	DGEBA Epoxy	C	10	-	0.668	[168]
GNP	5-10	Epoxy	C	-	10	3.35	[169]
GNP	10	Epoxy	C	8	-	1.18	[170]
GNP	25	Epoxy	C	10	-	0.65	[171]
GNP	10	Epoxy	C	-	10	3.87	[172]
GNP	30	Epoxy	C	-	25	6.44	[173]
GNP	15	Epoxy	C	-	24	12.4	[80]
GNP	70	Epoxy	C	10.1	-	4.01	[174]
GNP	-	Polyamide	C	5	-	0.41	[175]
GNP	15	Lauric Acid	C	-	1	0.49	[176]
GNP	0.55	Paraffin	C	20	-	45	[101]
GNP	5	Polystyrene	C	20	-	0.48	[121]
GNP	5-10	Eicosane	C	10	-	2	[177]
GNP	-	Polyolefin	C	10	-	5.6	[131]
GNP	5	Cyclic butylene terephthalate	C	30	-	4	[153]
GNP	15	Cyclic butylene terephthalate	C	30	-	6	[153]
GNP	25	Cyclic butylene terephthalate	C	30	-	8	[153]
GNP	<2	Polycarbonate	C	20	-	0.75	[151]
GNP	5	Polycarbonate	C	20	-	1.2	[151]
GNP	15	Polycarbonate	C	20	-	1.4	[151]
GNP	5	Polycarbonate	C	20	-	2.2	[151]
GNP	25	Polycarbonate	C	20	-	2.3	[151]
GNP	15	Mineral Oil	NC	50	27	7.1	[35]
GNP	25	Mineral Oil	NC	40	-	6.74	[96]
GNP	25	Silicone Oil	NC	30	-	4.2	[26]
GNP	1.2	Silicone Oil	NC	20	9.55	0.428	[32]
GNP	0.8	Silicone Oil	NC	20	9.55	0.359	[32]
GNP	0.4	Silicone Oil	NC	20	9.55	0.285	[32]
GN Sheet	3-5	Natural Rubber	C	-	5.78	10.64	[178]
EGNP	-	Epoxy	C	2	-	0.51	[179]
GO	150-500	DGEBA Epoxy	C	2	-	0.242	[180]
GO	44	Epoxy	C	1	-	0.65	[181]
rGO	-	Polyamide	C	10	-	0.416	[182]
rGO	320	Polyamide	C	10	-	3.5	[183]
rGO	44	TPU	C	1.04	-	0.8	[184]
rGO	0.45	Cellulose	C	30	-	0.07	[185]
rGO	25	PVDF	C	0.25	-	2.35	[186]
GF	0.014	Natural Rubber	C	-	5.78	3	[178]
GF	-	PDMS	C	0.5	-	0.4	[187]
f-FLG	0.2	Epoxy	C	10	-	1.53	[188]
3D-GNs	2-10	Epoxy	C	-	17	4.9	[42]
Graphene woven fabric	105	Polyimide	C	12	-	0.41	[189]

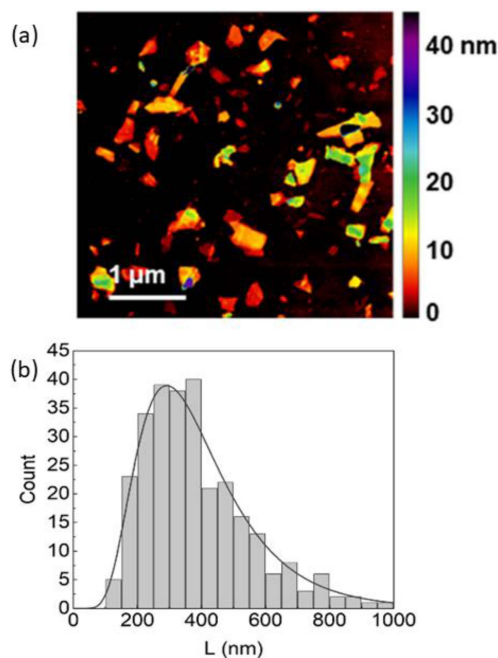


**FIGURE 5.** Thermal conductivity of graphene-FLG composites as a function of the filler loading, demonstrating the onset of the thermal percolation. (a)  $L \sim 2 - 8 \mu\text{m}$ . Reprinted with permission from Ref. [83]. Copyright (2018) American Chemical Society. (b)  $L = 20 \pm 7 \mu\text{m}$ . Reprinted with permission from Ref. [144]. Copyright (2015) American Chemical Society.

graphene nanoplatelets, GO–graphene oxide, rGO–Reduced graphene oxide, TPU–Thermoplastic polyurethane, PVDF–Poly (vinylidene fluoride), PDMS–Poly(dimethyl siloxane), GF–Graphene foam, f-GF–Functionalized graphene flakes, 3D-GNs–Three-dimensional graphene networks.

As seen from Table 1, little information is available for graphene-FLG filler composites with  $L$  which are near the *gray* MFP of graphene. The influence of  $L$  on the thermal properties of non-cured graphene-FLG filler composites with  $L \sim \text{MFP}$  is addressed [32]. The fillers with different  $L$  were produced from graphite using liquid phase exfoliation (LPE) [86], [87], [88], [90], [91]. Graphite was introduced to the process of LPE in an aqueous surfactant solution using liquid cascade centrifugation [190], [191] to select the size of graphene fillers. This enabled us to synthesize fillers with controlled different lateral dimensions and thicknesses. The fillers were introduced to the base polymer matrix to synthesize the required composites. Details on the LPE process can be found in Ref. [32]. The composites were prepared with a similar procedure as reported in Ref. [26] apart from the usage of acetone and thermal treatment for evaporation of the solvent.

The lateral dimensions and thickness of the fillers were determined with atomic force microscopy (AFM), Raman spectroscopy, scanning electron microscopy (SEM), and optical extinction spectroscopy. Fig. 6(a)–(b) illustrates the



**FIGURE 6.** AFM characterization of LPE exfoliated fillers. (a) A 2-D image of small  $L$  size fillers. (b) Histogram to determine the average  $L$  of the exfoliated small fillers. Reproduced with permission from [32]. Copyright (2021) American Chemical Society.

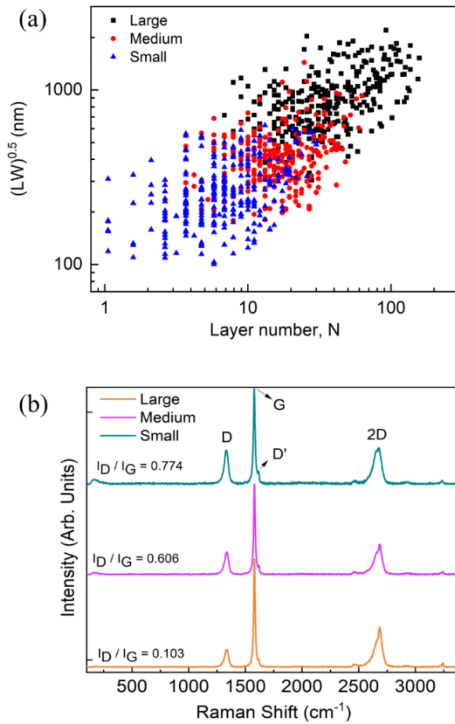
size distribution of a representative batch after LPE. The determined average  $L$  for each filler set were  $1.2 \mu\text{m}$ ,  $0.8 \mu\text{m}$ , and  $0.4 \mu\text{m}$  while the number of average atomic planes was 40, 19, and 8. For simplicity, these fillers would now be further labeled as “large”, “medium” and “small”, respectively. Table 2 summarizes the characteristics of the fillers used in the study.

One should note that it is difficult to obtain FLG fillers with fixed  $N$  and different  $L$  because there is always a correlation between the two. This can be detailed by the process of exfoliation during delamination by tearing [192]. The centrifugation helps the process of size selection which helps correlate the factors of  $L$  and  $N$  by providing a meaningful relationship between them [190], [191], [192] (see Fig. 7(a)). The characteristic lateral length can be represented as  $L_* = (L \times W)^{0.5}$  [32]. Each data point in the plot represents a graphene filler imaged and analyzed with AFM. We found that  $L$  influences the thermal transport greater than the thickness of the fillers. Fig. 7(b) shows the Raman spectra of the fillers with different sizes, indicating the correlation of Raman signature peaks with the filler sizes [59], [63], [75], [191], [193]. One can see that the intensity of the D peak increases as filler size decreases [59]. The  $I_D/I_G$  ratio can be used to determine the average characteristics of the fillers since this ratio decreases with the increase in filler size [194], [195], [196].

The  $K_{TIM}$ ,  $R_C$ , and  $R_{total}$  values of the composites with different filler characteristics were determined following the industry-standard method (ASTM-D5470) [197]. Fig. 8(a)–(b) presents the measured thermal properties of the synthesized composites with different filler sizes and loading

**TABLE 2. Characteristics of LPE Exfoliated graphene-FLG fillers**

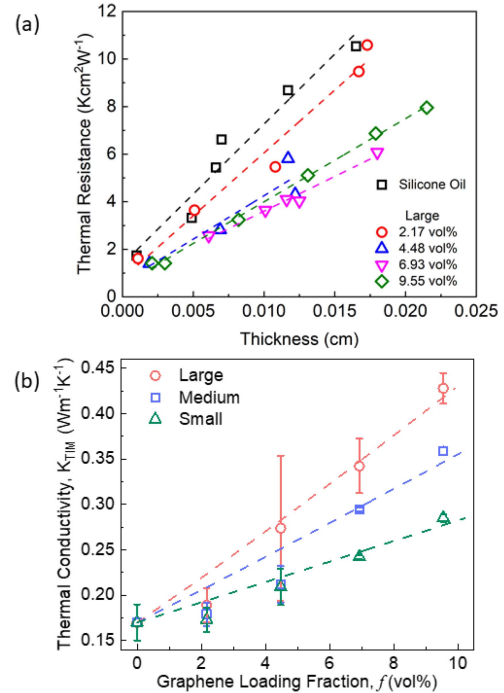
	Symbol	Large	Medium	Small
Average length (nm)	$L$	1200	800	400
Aspect ratio (length/width)	$L/W$	1.8	2.1	2.3
Aspect ratio (length/thickness)	$(L/t)$	110	120	200
Average layer number	$N$	40	19	8
Characteristic length $(L \times W)^{0.5}$ (nm)	$L_*$	900	430	270



**FIGURE 7. (a) Correlation between  $L$  and  $N$  of the fillers used with the data acquired from multiple AFM measurements. (b) Raman spectra of liquid-phase exfoliated graphene-FLG fillers with different characteristics. Reproduced with permission from [32]. Copyright (2021) American Chemical Society.**

fractions,  $f$ . Specifically, Fig. 8(a) shows the thermal resistance,  $R_{total}$ , of the large graphene-FLG filler composites as a function of thickness,  $BLT$  for different  $f$ . As expected,  $R_{total}$  increases with an increase in  $BLT$  for all filler loadings,  $f$ . This was consistent with all the filler-size composites. The total thermal resistance was the highest for lower loading fractions of graphene-FLG filler composites. The acquired data from these plots help us to extract the  $K_{TIM}$  and  $R_C$  of the synthesized composites.

Fig. 8(b) presents the extracted  $K_{TIM}$  as a function of  $f$  for the three filler characteristics and their comparison with a theoretical model. The thermal conductivity increases as  $f$  increases for all filler sizes as expected;  $K_{TIM}$  of the composites increase as the filler size increase. The findings can be rationalized considering the ratio of the graphene lateral size,  $L$ , to graphene's *gray* phonon MFP. The enhancement of  $K_{TIM}$  as filler size increases can also be explained by the increase in the thermal conduction path in composites as filler size

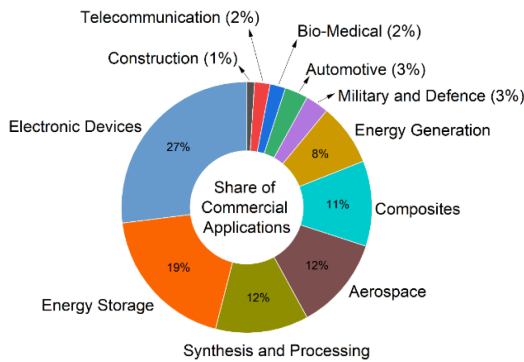


**FIGURE 8. Thermal properties of non-curing composites with different  $f$  of graphene-FLG fillers. (a) Thermal resistance as a function of composite  $BLT$  for different volume fractions of filler loading for large fillers. (b) Experimentally acquired thermal conductivity of synthesized non-curing composites with different  $L$  and  $N$  of the fillers compared with the theoretical Kanari model. Reproduced with permission from [32]. Copyright (2021) American Chemical Society.**

increases. The lower  $K_{TIM}$  at all  $f$  for the smaller filler size can be explained by its lower intrinsic thermal conductivity due to the reduced conduction path length as compared to larger length fillers. This estimate can be explained by the simple model based on kinetic theory of gases where the thermal conductivity of the fillers can be expressed as  $K \sim Cv\Lambda$ . Here, the average phonon group velocity is represented as  $v$ , the average phonon MFP is represented as  $\Lambda$  and  $C$  is the volumetric specific heat capacity of graphene fillers. If  $L < \Lambda$ , then  $K$  decreases linearly with  $L$  [83]. Therefore,  $\Lambda$  of fillers with lateral dimensions smaller the MFP exhibit lower thermal conductivity due to the phonon-edge scattering.

## V. COMMERCIAL APPLICATIONS OF GRAPHENE

While the initial interest in graphene originated from its unique electronic properties, it is now generally accepted that the high thermal conductivity of graphene and few-layer



**FIGURE 9.** Illustration of graphene commercial applications in various industries.

graphene is also extremely interesting and promising for practical applications. The use of graphene and FLG in composites is not limited to thermal management. It is also being utilized for electromagnetic interference (EMI) shielding [84], [99], [103], [108], [125], [198], [199]. Often graphene composites can perform dual functionalities in EMI shielding and thermal management. One should keep in mind that the term graphene in non-electronic applications typically means the mixture of graphene and FLG. The requirements for graphene's purity and crystallinity for applications in thermal management are not as strict as for applications that utilize the electronic or optical properties of graphene. For this reason, scaling up to the industrial level of graphene and FLG production via LPE processes is feasible. Fig. 9 summarizes the present application of graphene in various industries and its impact. The illustration is based on the data reported in Ref. [200], [201], [202].

Industrial reviews and forecasts from leading business analysts show that the graphene market would grow from less than \$100 million in 2020 to more than \$1 billion by 2032. The major application can be found in energy storage, thermal management, coatings, inks, adhesives, textiles, concrete, asphalts, sensors, photonics, and other applications [203], [204], [205], [206], [207], [208], [209], [210], [211]. The market size of TIMs in 2022 is estimated to be \$2.05 billion and is projected to grow up to \$4.86 billion or beyond by 2030 [212]. The target thermal conductivity varies, depending on the industry and intended application, with some aiming at  $\sim 10 \text{ Wm}^{-1}\text{K}^{-1}$  while others target more than  $25 \text{ Wm}^{-1}\text{K}^{-1}$  [212]. There is also a specific goal of reducing the thermal contact resistance [213], [214]. These industry needs and market trends open a wide application domain for graphene and FLG as fillers in thermal composites.

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