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Recent Developments in Carbon Based and Graphene Based Thermal Interface Materials: A Review

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ABSTRACT: This review presents a comprehensive and critical evaluation of carbon-based and graphene-based thermal interface materials for advanced thermal management in electronic systems. Conventional thermal interface materials typically exhibit thermal conductivities in the range of 0.1–10 W/m•K, limiting their effectiveness in high-power and miniaturized devices. In contrast, carbon-based TIMs demonstrate significantly enhanced performance, with carbon nanotube composites achieving 8–12 W/m•K and graphene-based composites reaching up to 23.2 W/m•K at 60 wt% loading. We provide a detailed comparative analysis of carbon nanotube and graphene architectures, emphasizing their exceptional intrinsic thermal conductivities (~3000 W/m•K for carbon nanotubes and ~5000 W/m•K for graphene) and addressing practical challenges such as interfacial resistance, dispersion uniformity, and large-scale integration. The review synthesizes fabrication strategies, performance trends, and application-specific considerations, while outlining future directions including hybrid architectures, eco-friendly formulations, and cost-effective, scalable manufacturing techniques. By integrating quantitative comparisons and identifying critical research gaps, this work offers a roadmap for next-generation thermal interface material development aimed at high-power electronics, telecommunications, and computing systems, where efficient thermal management is essential for reliability, energy efficiency, and long-term operational performance.

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1- Introduction

Thermal management has become a critical challenge in high-power electronics, where conventional TIMs fail to meet the demands of miniaturization and increased heat flux. While several reviews exist on carbon-based TIMs, most provide descriptive summaries without quantitative comparisons or actionable insights for future development. This review addresses these gaps by critically analyzing CNT and graphene-based TIMs, presenting comparative performance data, and outlining strategies for hybrid architectures, sustainable materials, and scalable fabrication methods. These contributions aim to guide researchers and industry toward next-generation thermal solutions.

Thermal Interface Materials (TIMs) are specialized materials used to enhance heat transfer between two surfaces, typically between a heat-generating component (e.g., microprocessors, power electronics) and a heat sink. Their primary function is to reduce thermal resistance by filling microscopic air gaps and irregularities, ensuring efficient heat dissipation. Traditional TIMs include thermal greases, phase change materials (PCMs), and metal-based pastes, but recent advancements have introduced high-performance materials like carbon nanotubes (CNTs) and graphene, which offer superior thermal conductivity and mechanical stability. These advanced TIMs are crucial in the electronics, automotive, and aerospace industries, where effective thermal management is essential for performance, reliability, and longevity.

Efficient thermal management is crucial in modern electronics, power devices, and aerospace applications, where excessive heat can degrade performance and reliability. Carbon-based nanomaterials, such as Carbon Nanotubes (CNTs) and Graphene, have been given special attention as high-performance Thermal Interface Materials (TIMs) due to their exceptional thermal conductivity, mechanical flexibility, and stability. Carbon nanotubes (CNTs) possess a unique one-dimensional (1D) tubular structure, enabling high in-plane thermal conductivity (~3000 W/mK) while maintaining flexibility and low density. Their ability to form vertically aligned arrays makes them ideal for bridging heat-generating surfaces with heat sinks, effectively reducing thermal resistance. On the other hand, graphene, a two-dimensional (2D) monolayer of sp^2-bonded carbon atoms, exhibits an even higher intrinsic thermal conductivity (~5000 W/mK) due to superior phonon transport. Graphene-based TIMs, including graphene laminates and hybrid composites, offer high thermal performance while maintaining excellent

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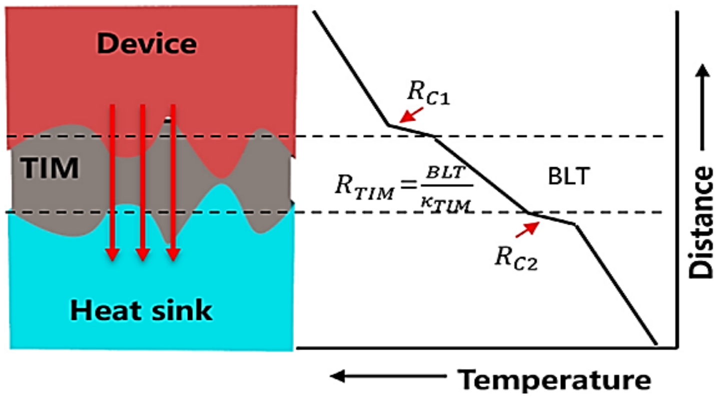

Fig. 1. Schematic representation of a thermal interface material (TIM) filling microscopic air gaps between a heat source and heat sink

mechanical and electrical properties. This review explores the thermal properties, structural advantages, fabrication methods, and real-world applications of CNTs and graphene as TIMs, highlighting their potential to replace traditional materials in high-performance thermal management systems.

By reducing the thermal resistance between heat sources and sinks, thermal interface materials (TIMs) are essential for improving heat dissipation in electronic devices [1]. The need for high-performance thermoelectric coolers (TIMs) with reduced thermal resistance and better thermal conductivity is rising as electronics power densities rise [2]. The efficiency of TIMs, which are available in a variety of forms such as pastes, solders, and resilient sheets, is contingent upon their capacity to adjust to surface topographies and displace insulating air gaps [3]. Recent developments in TIM technology include fusible particles for enhanced thermal conductivity and the production of continuous metal phase TIMs and carbon nanotube array TIMs [4]. Understanding the relationship between manufacturing processes, material structure, and contact resistance is crucial for enhancing thermal conductivity across interfaces and progressing the creation of smaller microsystems. Figure 1. Schematic representation of a thermal interface material (TIM) filling microscopic air gaps between a heat source and heat sink. TIM reduces interfacial thermal resistance by replacing low-conductivity air (0.026 W/m·K) with a higher-conductivity medium, improving heat dissipation.

There is a certain production of intense heat in devices due to the increasing demands for high power and miniaturization, which can lead to malfunctions or even failure. Thus, it is highly desirable to have efficient heat dissipation. As shown in Figure 1, the rough surfaces of heat sinks and high-power devices prevent complete contact between the two surfaces, resulting in unavoidable air gaps. Air has a thermal conductivity of nearly 0.026 W/m-K, which is roughly four orders of magnitude lower than that of metals. As a result, hotspots will form in the devices, reducing their efficiency and acting as a barrier to future advancements in microelectronics development [5-8].

Consequently, thermal interfacial materials (TIMs) are gaining a lot of attention [9-13] and are frequently used in thermal management and electronics device packaging [14], including computers, consumer electronics, telecommunications infrastructure, LEDs, automobiles, military/industrial equipment, medical equipment, and aerospace. TIMs can reduce the interfacial thermal resistance.

Depending on the material, thermal interface materials have generally been described as having thermal conductivities in the range from 0.1 to 10 W/m-K. Still, higher values are possible (up to 20 W/m·K) with high-performance TIMs composed of metal particles or phase-change materials. Regarding elastic modulus, a large variability exists for TIMs as a function of composition and form. Greases and gels exhibit low elastic modulus values, less than 1 MPa, with the ability for these materials to be flexible with high conformability. But there are also solid TIMs, such as gap fillers, adhesives, or pads, whose elastic moduli can lie in a wide range from a few MPa up to several GPa because of their polymeric or metallic nature. Commercial, or conventional, TIMs that are widely used include solders, phase change materials, thermal gel, grease that conducts heat, thermal pads, and thermal gel. Solder is known for having a high thermal conductivity and low thermal resistance, making it an ideal material for thermal interfaces. However, stress in addition to the high modulus of solder joints may cause stress-induced cracking. The pertinent process's expense and complexity present another problem. Thermally conductive fillers and matrix materials constitute the majority of the other commercial TIMs. Figure 6. Comparative analysis of carbon nanotubes (CNTs) and graphene for TIM applications. The plot shows intrinsic thermal conductivity, elastic modulus, and density on a logarithmic scale. Graphene exhibits higher thermal conductivity, while CNTs offer superior mechanical flexibility and lower density.However, greater proportions of thermally

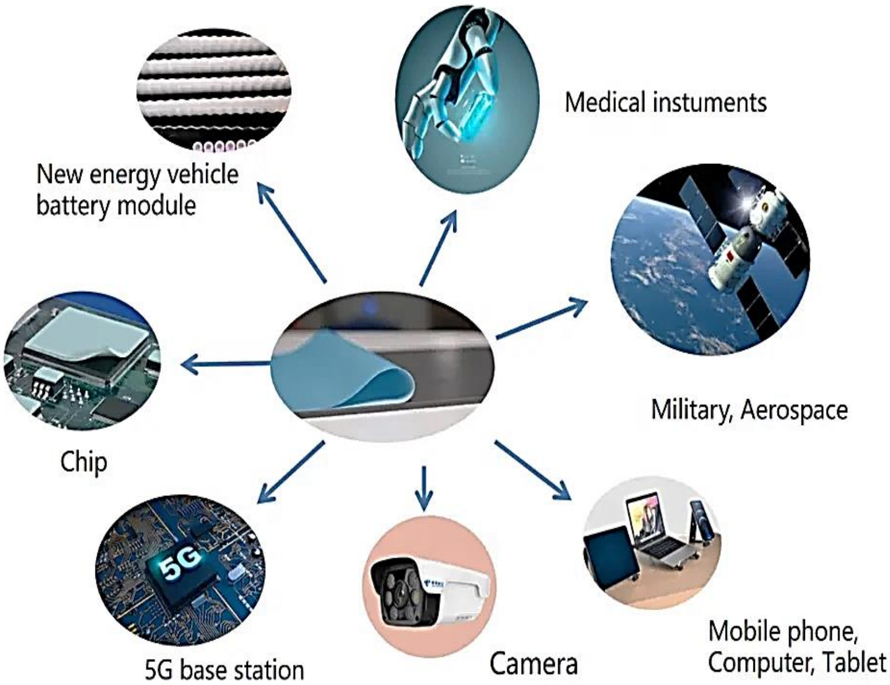

Fig. 2. Common applications of thermal interface materials (TIMs) in electronics packaging

conducting particles, which enhance thermal conductivity, typically cause the composite to become stiffer. Good thermal conductivity and Flexibility thus becomes mutually exclusive. Most modern state-of-the-art thermoelectric heat modules have a through-plane thermal conductivity of less than 30 W/mK, and most of them are less than 5 W/mK. On the other hand, their inherent high thermal conductivity and additional optimization are facilitated by novel materials under research, like carbon-based TIMs, which have high through-plane thermal conductivity and flexibility.

The potential of carbon-based thermal interface materials (TIMs) to improve electronic device heat dissipation has drawn a lot of interest [16]. TIM based on carbon has been developed in various forms, including carbon pastes, TIMs based on carbon nanotubes, and TIMs based on graphene. The high thermal conductivity of these materials makes them highly sought after, but thermal contact conductance is becoming more widely accepted as a more accurate indicator of TIM performance [17]. Researchers are looking into carbon-based TIMs for a variety of other industries besides electronics. They have potential, for example, in applications related to construction, especially in the production of artificial timber. In these situations, flexural strength can be significantly increased, and durability can be improved by adding a small amount of epoxy binder to carbon-based materials [18].

The table 1 lists the different Thermal Interface Materials (TIMs) that are frequently used to enhance heat dissipation in high-tech electronics. Graphene-Carbon Nanotube composite films are perfect for high-performance electronics where effective heat management is essential because of their exceptionally high thermal conductivity (~627 W/m-K in-plane). With a thermal conductivity of more than 200 W/m-K, metal-based TIMs like silver nanowire find use in high-power industries like defence and automotive electronics. Polymer-based TIMs filled with carbon provide moderate conductivity (1-10 W/m-K) and are cost-effective, making them suitable for less demanding applications like computer processors. These materials are pivotal in reducing interfacial thermal resistance and enhancing device performance in environments with high heat fluxes

To increase thermal transfer efficiency, TIMs are made of thermally conductive fillers, such as metal or ceramic particles, distributed throughout a polymer matrix. Although bulk thermal conductivity is frequently taken into account first when evaluating TIM performance, other elements like interface impedance and boundary resistance have a big impact on the system's overall capacity for heat dissipation [19]. For high-power electronic systems like server CPUs, phase change materials (PCMs) are often employed as TIM2, which is applied in between the integrated heat distributor and the heatsink. Surface warpage and component flatness have an intense impact on PCM performance as TIM2, as they can result in irregular contact and increased thermal resistance [20].

On a broad note, two thermal management analytical tools, the Transient Interferometric Mapping (TIM) and others, are employed to evaluate the heat dissipations of semiconductor

Table 1. Different TIMs and their application according to their thermal conductivity.

Material Type	Thermal Conductivity (W/m-K)	Applications	Reference
Graphene–Carbon Nanotube Composite	627 (in-plane)	High-performance electronics, improving heat dissipation	Yu et al. (2023) [62]
Silver Nanowire (Metal-Based TIM)	More than 200	Automotive, defense, and high-power electronics.	Xing et al. (2022) [63]
Polymer-Based TIM with Carbon Fillers	1 to 10	Computer processors moderate heat transfer at low cost.	Xing et al. (2022) [63]

devices that include GaN-based high-electron-mobility transistors (HEMT) and ESD protection components. [21] Researchers have the tools and know-how to construct thermal maps of the different thermal profiles, which is crucial in the design of thermal structures for high-frequency and high-power systems. Another context in which the acronym TIM is encountered is that TIM also stands for Technology Integration Matrix, a tool used to examine the integration of technology in K–12 education. This framework allows for organized comprehension of how instruction includes the use of technology; it evaluates teaching strategies based on five attributes and their levels of technology [22].

Reported intrinsic thermal conductivity values for carbon-based nanomaterials, such as ~3000 W/m·K for carbon nanotubes and ~5000 W/m·K for single-layer graphene, represent ideal conditions for defect-free, isolated structures measured under controlled laboratory environments. These values are typically obtained using techniques such as Raman spectroscopy and applying in-plane heat conduction. In practical thermal interface material (TIM) applications, actual performance is significantly lower, often in the range of 8-12 W/m·K for CNT-based composites and 10-23 W/m·K for graphene-based composites due to factors such as interfacial resistance, agglomeration, and imperfect dispersion. Therefore, these intrinsic values should be considered theoretical benchmarks rather than achievable figures in real-world systems [65].

2- Methodology of the Review:

This review was conducted using a structured approach to ensure comprehensiveness and relevance. Literature was collected from Scopus, Web of Science, and IEEE Xplore databases using keywords such as “thermal interface materials,” “graphene TIM,” and “carbon nanotubes TIM.” The search covered publications from 2010 to 2025. Only peer-reviewed journal articles and experimental studies reporting thermal conductivity, fabrication techniques, and application insights were included; patents and non-English papers were excluded. Data extraction focused on quantitative performance metrics (e.g., thermal conductivity, interfacial resistance) and fabrication strategies. Comparative analysis was performed between CNT- and graphene-based TIMs under similar loading conditions. Unlike previous reviews, this study synthesizes quantitative benchmarks, identifies critical challenges, and outlines future research directions, including hybrid architectures, eco-friendly alternatives, and scalable manufacturing methods.

2- 1- Carbon-Based Thermal Interface Materials

Carbon-based thermal interface materials (TIMs) have grabbed significant attention due to their exceptional thermal properties, which hold the potential to hone heat dissipation in electrical devices [23]. Figure 3 shows the structural relationship between graphene, graphite, and carbon nanotubes (CNTs). Graphene consists of a single layer of sp^2-bonded carbon atoms arranged in a hexagonal lattice. Multiple graphene layers stack to form graphite. When a single graphene sheet is rolled up, it forms a single-walled carbon nanotube (SWCNT), whereas rolling multi-walled carbon nanotubes (MWCNTs). This structural evolution highlights the dimensional transition from 2D (graphene) to 1D (CNTs) and its impact on thermal and mechanical properties relevant to TIM applications.

The high mechanical strength and heat conductivity

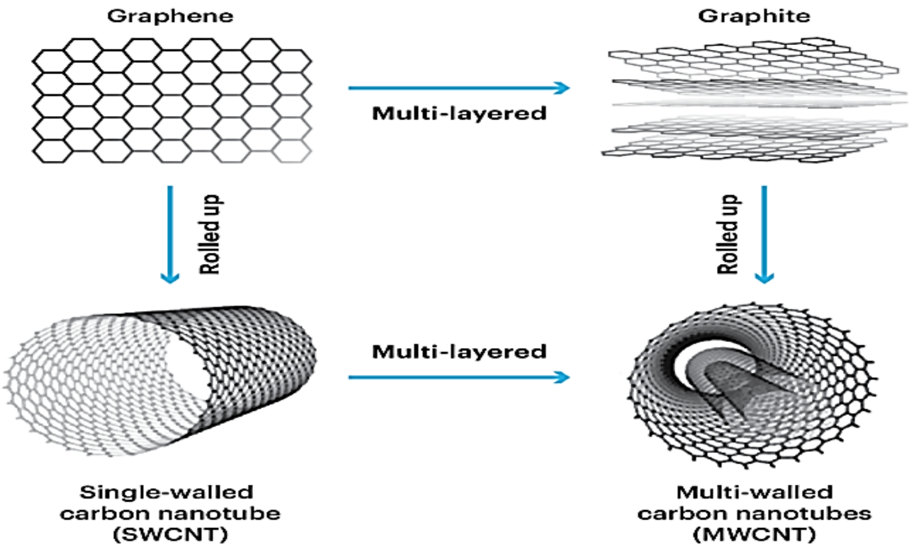

Fig. 3. Structural relationship between graphene, graphite, and carbon nanotubes (CNTs).

of carbon-based thermoelectric materials (TIMs) make carbon fibers, graphene, and carbon nanotubes (CNTs) great examples. Despite these materials' good qualities, high costs and performance issues significantly prevent practical implementation. On the other side, carbon black paste is still a reasonably priced option with good solid-state conformability; moreover, its thermal conductivity rises with more solid content, potentially reducing its conformability. Therefore, finding a balance between usefulness and performance requires careful optimization.

2- 1- 1- Carbon Nanotubes (CNT)

Carbon-related nanomaterials such as carbon nanotubes (CNTs), graphene oxide, and graphene quantum dots are gaining popularity in the biomedical fields, in addition to their use in heat management. Their unique chemical and physical properties make them perfect for many uses, including drug delivery, cancer medication, and biosensing [24, 25]. Due to these materials' versatility, they can be used in electronics as well as cutting-edge healthcare solutions. The objective of current research is to overcome existing challenges and enhance the efficiency of carbon-based TIMs for broader commercial application. The ongoing advancements in material science aim to increase the effectiveness and economy of these TIMs, expanding the range of industries in which they can be employed.

Carbon atoms arranged in a hexagonal pattern create carbon nanotube (CNTs) structures, which are cylindrical in form at the nanoscale. The two primary kinds are single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). MWCNT and SWCNT are shown in Figure 3. To create SWCNTs, one graphene sheet is wrapped into a tube with a typical diameter of 0.4 to 2 nanometers. On the other hand, MWCNTs consist of more than two layers and have a thickness of up to 100 nanometers. Because of their superior mechanical strength, elevated thermal conductivity, and unique electronic features, these kinds are more useful in various fields than all the other SWCNTs. SWCNTs show excellent electronic characteristics whereby, depending on their chirality, they can exhibit metallic or semiconducting characteristics, whereas MWCNTs are mostly metallic. Due to their strong physical and chemical properties, these carbon-based nanostructures have found applications in a wide array of areas such as energy storage, electronics, composite materials, and biomedical engineering.

2- 1- 1- 1- Properties of CNT as TIMs

Carbon nanotubes (CNTs) are exceptional, nanoscale tubular structures owing to their unique cylindrical architecture, which also has great mechanical, electrical, and thermal properties [26]. Their tensile strength is high at up to 63 GPa, and their Young's modulus is also very high at up to 1 TPa, which makes them highly suitable for reinforcing composite materials [27, 28]. As CNTs can be used in both metallic and semiconducting applications, such as field-effect transistors and single-electron transistors, they possess greater prospects in electronics. The mechanical strength and stiffness of CNTs have been the focus of several investigations, which demonstrate that they are capable of enhancing the performance of a wide range of materials.

Apart from their remarkable mechanical characteristics, CNTs also have interesting electrical, magnetic, and optical properties. These properties create interesting opportunities for therapies involving carbon nanotubes (CNTs) for novel

Table 2. Thermal conductivity of CNT-based polymer composites' thermal conductivity.

Material	Matrix	Thermal Conductivity (W/m-K)
CNT	EP	4
CNT	EP	1
CNT	Polyethylene	63
CNT/Gr	PVDF	1
SWCNT	PMMA	2
MWCNT	PMMA	3
CNT	Polyamide	16

biological applications such as drug delivery, biosensing, and targeted therapy for cancer. The compliance of CNTs is illustrated by their ability to be integrated into various matrices such as metals, polymers, and ceramics. Our hope for integrating them is to be able to enhance the functionality of advanced materials for commercial applications by exploiting their superior properties.

There is still being conducted research about the mechanical bulk properties, such as defect formations, fracture development, and interface interactions of carbon nanotubes (CNTs), in an effort to enhance and improve CNTs' performance in practical applications. CNTs are also being investigated for their potential in the development of innovative drug delivery systems and treatments due to their unique properties and ability to interact at the nanoscale with biological systems [29]. The goal of further research is to maximize the functionality and integration of carbon nanotubes (CNTs) in a variety of domains to realize their full potential and spur innovation in a range of industries.

2- 1- 1- 1- Materials Based on Carbon Nanotubes that are used in Thermal Management

Because of their extraordinary mechanical, electrical, and thermal properties, carbon nanotubes (CNTs) have become a possible new material for heat control [30, 31]. Research has focused on emerging carbon nanotube (CNT)-based composites, namely carbon/carbon composites and aligned CNT arrays, which have low thermal expansion coefficients and high thermal conductivity, correspondingly, making them good for governing heat in electronic components [32]. A range of production techniques have been inspected to improve these composites' thermal performance. By improving heat flow paths, electrospinning, for example, has been used to progress CNT alignment inside polymer matrices, resulting in a significant increase in thermal conductivity.

Carbon nanotubes (CNTs) have been combined into numerous thermal management applications, such as polymer composites with greater thermal conductivity, membrane materials showing anisotropic heat conduction, and composite thermal interface materials (TIMs). These CNT-improved TIMs expand heat dissipation by creating a conductive pathway between heat-producing elements and heat-dissipating components. Moreover, CNT-reinforced metal-matrix composites have been planned to modify the thermal expansion coefficient, honing their compatibility with other system components. CNT-based thermal greases using natural oils have also shown excellent thermal conductivity and long-term stability, offering an environmentally friendly option for high-performance thermal solutions [33]. Table 2 depicts an overview of CNT-based polymer composites' thermal conductivity.

Ongoing research in this field is centered on developing high-density aligned CNT films and large-diameter CNT array composites to achieve even greater thermal management efficiency for future electronic devices. Table 2 gives information about CNT-based polymers and their thermal conductivities with respect to the matrix. By tackling challenges related to manufacturing scalability and interface compatibility, these advancements are anticipated to unlock the full potential of CNTs in thermal management applications, providing solutions for high-power and miniaturized electronics [34, 35, 36, 37].

Carbon nanotubes (CNTs) and graphene exhibit remarkable intrinsic thermal conductivities of approximately 3000 W/m·K for CNTs and 5000 W/m·K for graphene, making them highly attractive for thermal interface materials (TIMs). CNTs bring benefits such as mechanical flexibility and low density, which facilitate their integration into polymer matrices. However, interfacial thermal resistance at the CNT–CNT and CNT–matrix junctions remains a significant barrier; in situ microscopy shows that the thermal contact resistance (TCR) can vary by two orders of magnitude and is highly sensitive to nanoscale morphological features. Furthermore, large-scale synthesis of aligned, defect-free CNT networks with consistent performance poses a significant challenge [66].

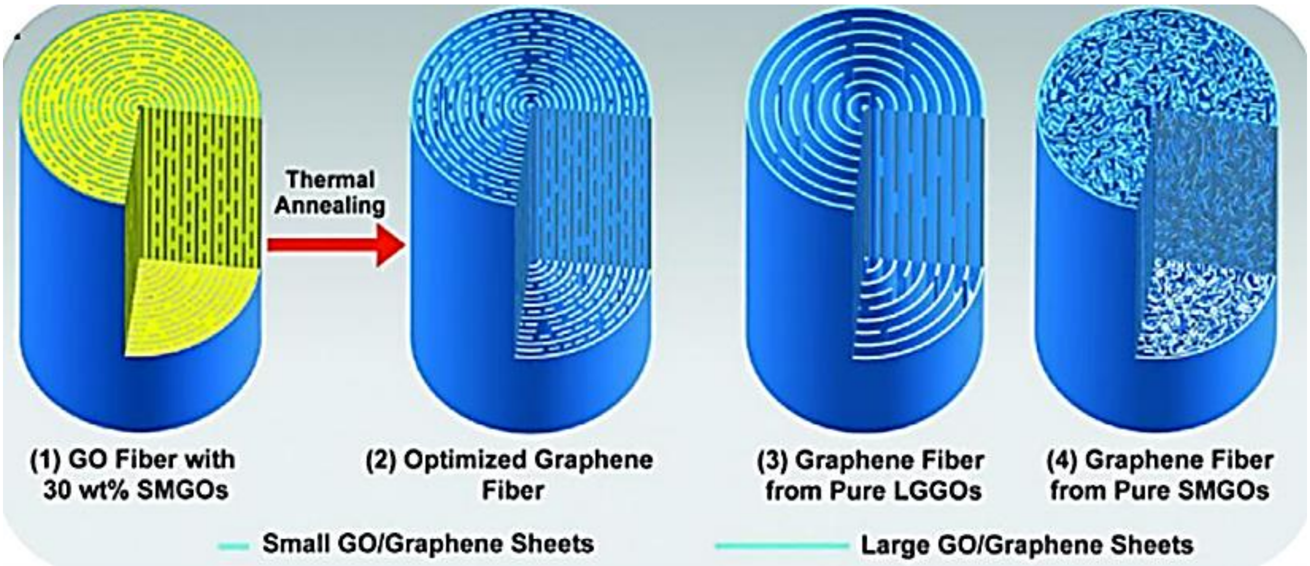

Fig. 4. Strong, mechanically strong, and highly thermally conductive Gr fibers with an "intercalated" structure of large and small-sized Gr sheets.

2- 1- 2- Graphene

Because of graphene's extraordinary thermal conductivity, which can surpass 5000 W/m·K in pure single-layer forms, graphene-based thermal interface materials (TIMs) have become extremely promising candidates for thermal management in contemporary electronics [38]. Dispersed graphene/polymer composites, graphene framework/polymer composites, and inorganic graphene-based monoliths are the three main categories into which these materials fall [39]. Of these, graphene-epoxy composites have shown exceptional gains in thermal conductivity; improvements in thermal conductivity have exceeded 1000% at volume loadings as low as 5%. Graphene composites are extremely effective at dissipating heat because they perform far better than traditional filler materials.

According to [40], few-layer graphene (FLG) composites have also demonstrated remarkable thermal properties. These include low thermal interface resistance and notable increases in thermal conductivity, with enhancement factors reaching up to 17 at concentrations of 10 vol. Furthermore, hybrid fillers that combine graphene with other materials, like boron nitride (BN), have the advantage of providing both electrical insulation and efficient thermal conduction, which makes them perfect for applications that need independent control over these properties [41]. Due to their adaptability, graphene-based thermoelectric coolers (TIMs) can meet the demanding requirements of high-speed, high-power electronic devices, where reliable and efficient heat dissipation is important for optimal performance. Figure 4 depicts Strong, mechanically strong, and highly thermally conductive Gr fibers with an "intercalated" structure of large and small-sized Gr sheets

2- 1- 2- 1- Properties of Graphene as TIMs

Graphene has emerged as a promising material for heat interface applications due to its exceptional thermal properties. Studies have shown that graphene-based composites can significantly enhance thermal conductivity, even at low volume fractions. One study reported a fivefold increase in thermal conductivity at 5% volume loading, while another study [42] achieved 12.8 W/m-K with 30 wt% graphene. [43] demonstrated a 500% increase in thermal conductivity using hybrid graphene-metal nanocomposites. The exceptional improvement is due to several factors: graphene's inherently high thermal conductivity, its strong interaction with matrix materials, and the diverse range of filler sizes spanning from nano to microscale. These graphene-incorporated thermal interface materials (TIMs) surpass traditional TIMs, which necessitate high filler volume fractions (approximately 70%) to achieve comparable conductivities. The research indicates that graphene-based TIMs show considerable promise for heat management in various applications, including electronics, optoelectronics, and photovoltaics.

2- 1- 2- 2- Materials Based on Graphene used in Thermal Management.

Graphene-based materials continue to show exceptional potential for thermal heat management in electronics due to high thermal conductivity and versatility. When incorporated into phase change materials, graphene significantly enhances thermal conductivity by more than two orders of magnitude while retaining the material's ability to store latent heat [44]. Additionally, thick graphene networks grown via chemical vapor deposition (CVD) on ceramic substrates

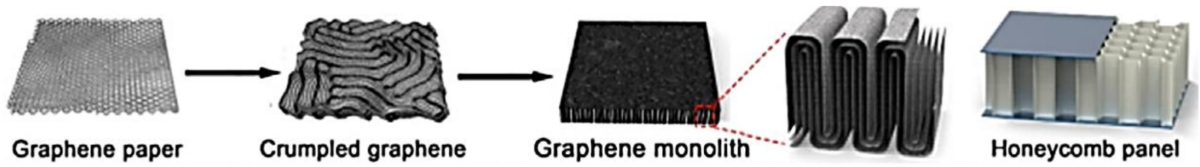

Fig. 5. A Schematic diagram of the structural change for Gr based on the proposed method.

Table 3. Thermal conductivities of Gr-based polymer composites.

Material	Matrix	Thermal Conductivity (W/m-K)
Gr	EP	33.5
rLGO	PVDF-HFC	19
Thermally reduced Gr sheets	poly(p-phenylene benzobisoxazole)	5
Gr Laminate	polyethylene terephthalate	4
Graphite nanoplatelet	polyethylene	4.62
GR flakes	EP	1.5
Cellulose/Gr aerogel	PCM	1.3

have demonstrated superior performance in power switching devices, reducing hot spot temperatures by over 25°C. Compared to conventional methods [45]. Figure 5 shows a schematic diagram of the structural change for Gr based on the proposed method.

Table 3 shows thermal conductivities of different Gr-based polymer composites.

Graphene-enhanced phase change materials also outperform other carbon materials and metal nanoparticles in managing heat for high-power-density battery systems [46]. Furthermore, hybrid composites that combine graphene with ceramic fillers have achieved extremely high thermal conductivities, exceeding 12 W/m-K, while preserving strong mechanical properties and electrical insulation. These materials, when used for potting, can significantly lower the operating temperatures of electronic components from 110°C to 37°C and reduce thermal resistance by up to 90%. [47] This highlights the growing importance of graphene in advanced thermal management solutions.

The thermal conductivity of Gr fiber can reach 1290 W/m-K [48] when small-sized Gr sheets, as seen in Fig. 4, fill in gaps and micro-voids while large-sized Gr sheets are arranged in a highly ordered manner. Through mechanical machining, Jiang et al. created a Gr-based microstructure that is primarily vertical Gr with a thin cap layer of horizontal Gr layers on the top and bottom sides (Fig. 5). The resulting Gr monolith has a through-plane thermal conductivity of 143 W/m-K, which is ultra-high compared to many metals, and a low compressive modulus of 0.87 MPa [49]. Compared to the most advanced commercial TIMs, a system that uses the Gr monolith function as TIM has a cooling output that is three times higher [50-56].

Graphene-based TIMs are synthesized using several approaches: Chemical Vapor Deposition (CVD), which produces high-quality, large-area graphene films suitable for direct integration on substrates. However, CVD is expensive and requires high temperatures, limiting scalability. Liquid-phase exfoliation, which involves sonication or shear mixing of graphite in solvents to produce graphene flakes. This method is cost-effective but often yields non-uniform flake sizes. Reduction of Graphene Oxide (rGO): In this process, graphene oxide is chemically or thermally reduced to restore conductivity and thermal properties. rGO offers easier processing but has lower intrinsic conductivity compared to pristine graphene.

Figure 6 shows a clear positive correlation between graphene loading and thermal conductivity in thermal interface materials (TIMs). At a low loading of 5%, the thermal

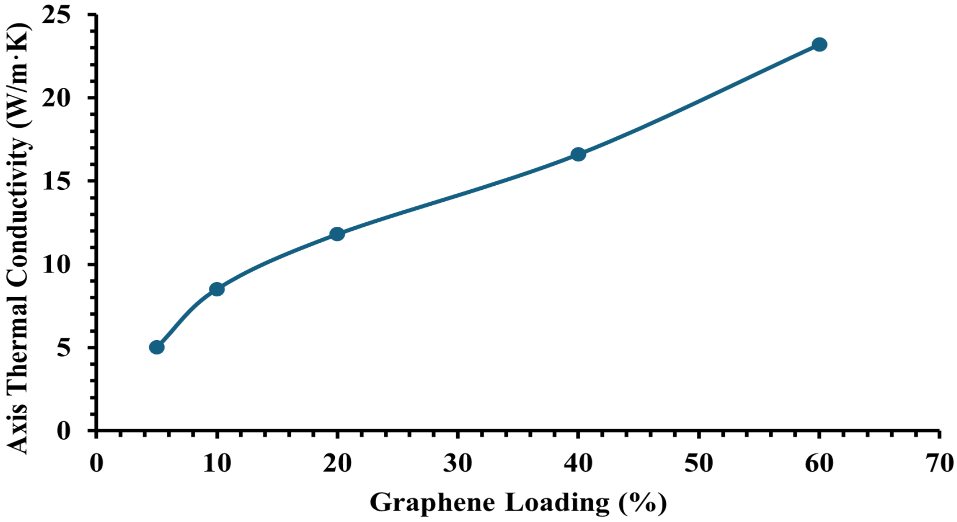

Fig. 6. Variation of thermal conductivity with graphene loading.

conductivity reaches 5.0 W/m·K, which already represents a significant improvement over conventional TIMs. As the loading increases to 10%, conductivity rises to 8.5 W/m·K, and at 20% it reaches 11.8 W/m·K, surpassing most CNT-based TIMs. Further increments to 40% and 60% result in thermal conductivities of 16.6 W/m·K and 23.2 W/m·K, respectively, demonstrating graphene's superior ability to enhance heat dissipation. This trend highlights graphene's potential for high-performance applications, although higher loadings may introduce challenges such as agglomeration, reduced mechanical flexibility, and increased cost. Therefore, while graphene offers exceptional thermal performance, practical implementation requires balancing conductivity gains with manufacturability and economic feasibility. [64]

Graphene offers even greater advantages through its superior in-plane heat conduction, enabling thermal conductivities of 10–23 W/m-K at moderate filler loadings. Hybrid rGO/CNT composite films achieved an in-plane thermal conductivity of approximately 627 W/m-K, with CNTs enhancing through-plane conduction by bridging adjacent graphene layers. Despite these advances, limitations remain—graphene particles tend to agglomerate in polymer matrices, and through-plane heat transfer is typically much less efficient than in-plane, limiting overall thermal performance. Additionally, the high cost of pristine graphene and its electrical conductivity can pose insulation risks unless appropriately addressed through surface functionalization or coatings [67].

Ultimately, both CNT- and graphene-based TIMs require advanced strategies such as surface engineering, hybrid filler integration, and orientation control to fully exploit their intrinsic properties. Realizing high-performance TIMs at an industrial scale depends on integrating these processing techniques into scalable manufacturing workflows.

3- Comparison of CNT and Graphene as TIMs

Figure 7 presents a comparative analysis of Carbon Nanotubes (CNTs) and Graphene in terms of their key physical properties relevant to their use as Thermal Interface Materials (TIMs). The y-axis represents the logarithmic scale of property values, while the x-axis lists different properties. The solid blue line represents CNTs, and the dashed red line represents graphene. Here, in-plane thermal conductivity values are taken for comparison.

Carbon nanotubes (CNTs) and graphene as thermal interface materials (TIMs) have been thoroughly compared in recent studies, with graphene-based TIMs exhibiting better thermal conductivity performance. According to Mohamed et al. (2020), graphene-based TIMs outperformed CNT-based TIMs, which only managed 12.2 W/m-K at the same loading, with a thermal conductivity of 23.2 W/m-K at a 60 wt% graphene loading [57]. Commercial TIMs can benefit from even tiny (0.5–1%) CNT additions to increase their thermal conductivity [58]. In this work they brought in even more innovation when they created a covalently bonded graphene-CNT hybrid [59]. This hybrid significantly outperformed few-layer graphene, especially in c-axis heat transfer, where it achieved improvements in performance that were more than two orders of magnitude.

Various nano-TIM technologies, including flexible graphite films, metal nano springs, laminated solder, and carbon nanotube-based materials [60]. Among these, copper nanospring TIMs consistently demonstrated thermal interface resistivities as low as 1 mm^3K/W, outperforming other nano-TIMs such as CNT and flexible graphite-based materials, which achieved resistivities below 10 mm^3K/W. The study highlighted ongoing efforts to further reduce thermal interface resistance, with the goal of reaching 0.1 mm^2-K/W. Achieving this target would significantly enhance thermal management

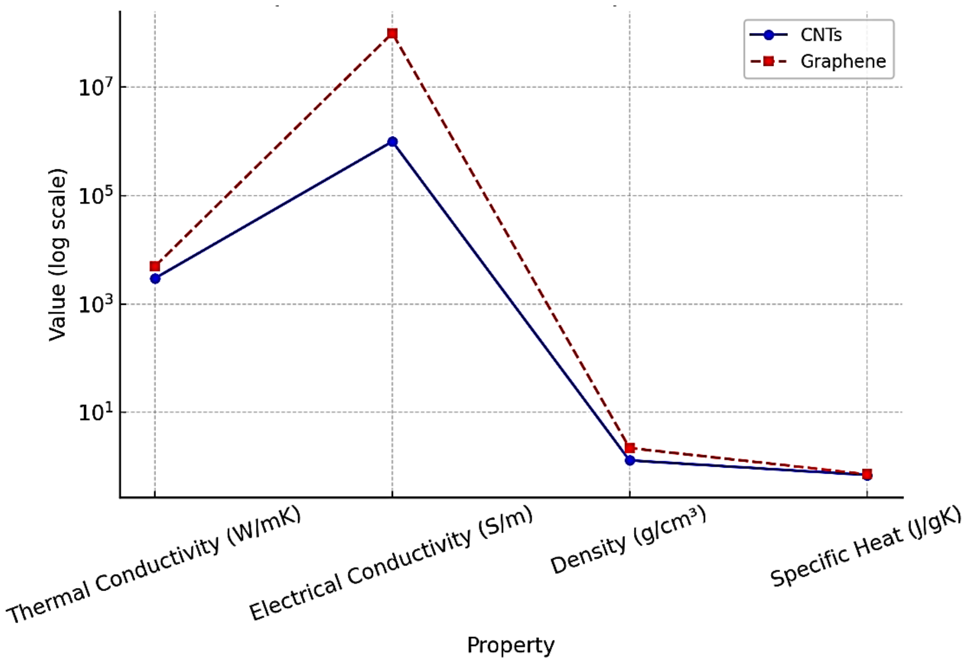

Fig. 7. Comparison of different properties of CNT and graphene.

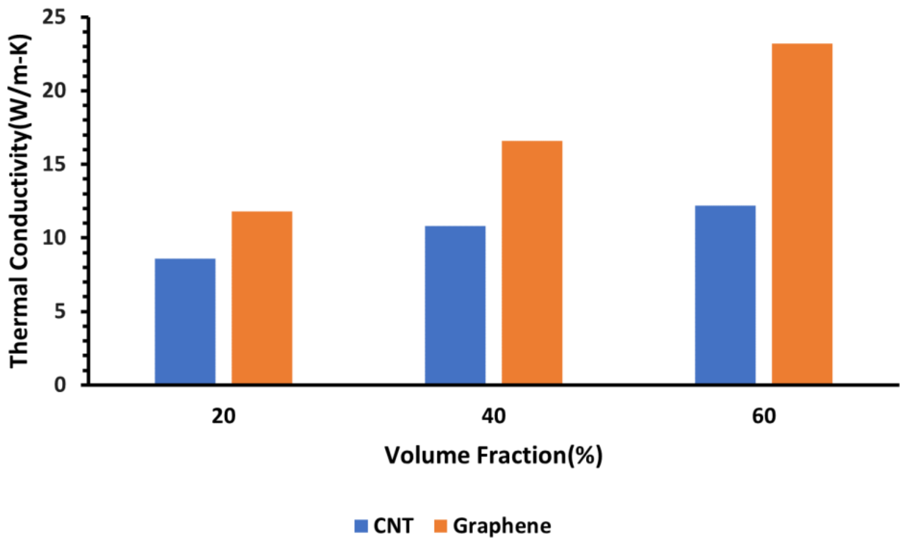

Fig. 8. Comparison of Thermal Conductivity of CNT and Graphene.

in high-power and high-performance electronic devices.

The thermal conductivity related to graphene-based TIMs and carbon nanotubes (CNTs) at three distinct volume fractions, 20%, 40%, and 60%, is contrasted in Figure 8. The bar graph illustrates how graphene has better heat conductivity than CNT at all volume fractions. Graphene shows a conductivity of 11.8 W/m-K at 20% volume fraction, which is significantly higher than CNT, which is 8.6 W/m-K. Similarly, graphene continues to outperform CNT at 40% and 60% fractions, with values of 16.6 W/m-K and 23.2 W/m-K, respectively, as opposed to 10.8 W/m-K and 12.2 W/m-K for CNT. This discrepancy highlights the fact that graphene dissipates heat more effectively than other materials, which makes it a more promising material for high-performance TIM applications. Higher loadings appear to improve overall thermal performance, as evidenced by the consistent rise in thermal conductivity for both materials with increasing volume fraction (graphene showing a much steeper rise).

Table 4. Practical Adoption Barriers for CNTs and Graphene in TIM Applications.

Material	Barrier	Impact	Possible Solution
CNT	Cost	The high production cost of CNTs limits commercial viability.	Develop low-cost CNT synthesis methods and hybrid composites.
CNT	Scalability	Difficulty in achieving uniform alignment and dispersion at industrial scale	Optimize alignment techniques (electrospinning, in situ functionalization).
CNT	Real-World Integration	Challenges in meeting packaging constraints (thickness, adhesion)	Surface functionalization and improved bonding strategies
CNT	Reliability	Performance degradation under thermal cycling and oxidation	Protective coatings and accelerated reliability testing
Graphene	Cost	High cost of pristine graphene and CVD processes	Explore scalable graphene production and hybrid fillers.
Graphene	Scalability	Agglomeration and dispersion issues in polymer matrices	Use surfactants and functionalization to improve dispersion.
Graphene	Real-World Integration	Electrical conductivity risks short-circuiting in insulating applications.	Apply insulating coatings and hybrid architectures.
Graphene	Reliability	Structural changes and oxidation reduce long-term stability.	Material stabilization and environmental protection layers

3- 1- Practical Adoption Barriers for Carbon- and Graphene Based TIMs

Table 4 compares key barriers faced by CNTs and graphene in real-world thermal interface material applications. For both materials, challenges include high production cost, scalability issues, integration with device packaging, and long-term reliability. CNTs struggle with alignment and adhesion, while graphene faces agglomeration and electrical insulation concerns. Proposed solutions include low-cost synthesis, surface functionalization, hybrid composites, and protective coatings to enhance performance and enable commercial viability [68-69].

4- Challenges and Future Directions

Despite significant progress in carbon-based and graphene-based TIMs, several critical challenges hinder their widespread adoption:

Figure 8 illustrates the approximate share of each major challenge in hindering the commercialization of carbon-based and graphene-based thermal interface materials (TIMs).

Interfacial resistance (30%) is the largest contributor, as poor contact significantly limits heat transfer efficiency.

Cost (25%) and **manufacturability (25%)** are equally critical, reflecting the high expense of synthesis and difficulties in achieving uniform dispersion and alignment.

Reliability (20%) accounts for long-term performance issues under thermal cycling and environmental stress.

This visualization emphasizes that addressing interfacial resistance and improving manufacturing scalability should be top priorities for future research.

Interfacial Thermal Resistance: Even with high intrinsic thermal conductivity, poor contact at the interface between TIM and device surfaces limits heat transfer efficiency. Surface roughness, voids, and inadequate bonding lead to increased thermal impedance [19, 60]. Advanced surface functionalization and bonding layers are needed to minimize this resistance.

Cost and Scalability: Production methods such as chemical vapor deposition (CVD) for CNTs and graphene are expensive and energy-intensive [38, 47]. Large-scale manufacturing remains a challenge, making these materials less viable for cost-sensitive applications. Research into low-cost synthesis and hybrid composites is essential.

Manufacturability and Integration: Achieving uniform dispersion of CNTs or graphene in polymer matrices and controlling their alignment is difficult [32,37]. Techniques like electrospinning and in-situ functionalization show promise but

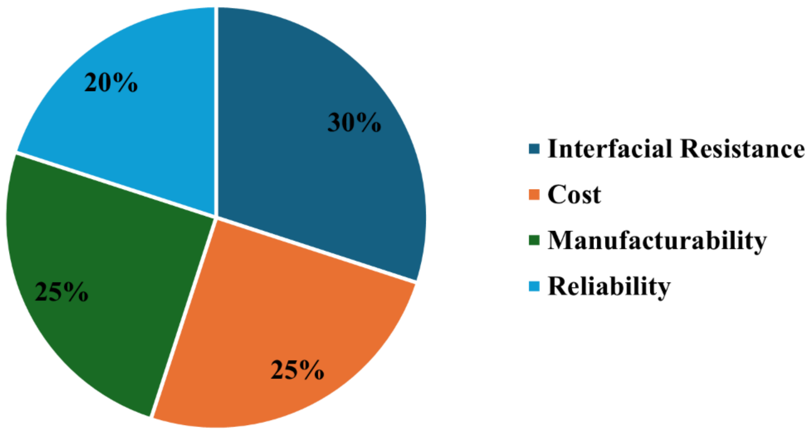

Fig. 9. Relative Contribution of Key Challenges in Carbon-Based TIM Adoption.

require further optimization for industrial-scale processes.

Long-Term Reliability: TIMs must maintain performance under thermal cycling, humidity, and mechanical stress. CNT and graphene composites can degrade over time due to oxidation or structural changes [33, 45]. Reliability testing under accelerated aging conditions and protective coatings are critical for commercialization.

Future research should focus on hybrid architectures (e.g., graphene-CNT composites), improved bonding strategies, and scalable fabrication methods. Additionally, life-cycle cost analysis and environmental impact assessments will be important for sustainable deployment.

5- Conclusion:

Carbon-based and graphene-based TIMs represent a transformative approach to thermal management, outperforming traditional greases and solders in conductivity and mechanical adaptability. Graphene-based composites consistently deliver superior thermal performance, achieving up to 23.2 W/m-K compared to CNT-based TIMs at 12.2 W/m-K under similar loadings. Despite these advances, practical adoption is constrained by interfacial resistance, cost, and scalability. Future research should prioritize hybrid architectures (e.g., graphene-CNT composites) to exploit synergistic properties, eco-friendly and recyclable polymer matrices to reduce environmental impact, and scalable fabrication techniques such as roll-to-roll processing and low-temperature CVD for industrial viability. Additionally, reliability testing under thermal cycling and environmental stress, coupled with surface functionalization strategies, will be essential for commercialization. By addressing these challenges, next-generation TIMs can deliver high thermal conductivity, mechanical flexibility, and sustainability, enabling their integration into high-power electronics, automotive systems, and emerging technologies.

Future Scope:

Future research should focus on developing hybrid architectures that combine graphene, CNTs, and other high-conductivity fillers to overcome limitations in through-plane heat transfer and mechanical flexibility. Eco-friendly and sustainable TIMs, such as bio-based polymers or recyclable composites, are critical for reducing environmental impact in large-scale electronics manufacturing. Additionally, scalable and cost-effective fabrication methods such as roll-to-roll processing, low-temperature chemical vapor deposition, and advanced dispersion techniques must be optimized to enable industrial adoption. Integration of these strategies with surface functionalization and reliability testing will pave the way for next-generation TIMs that meet performance, sustainability, and economic requirements.

List of abbreviations

TIMs: Thermal interface materials, CNTs: carbon nanotubes, CVD: chemical vapor deposition, FLG: few-layer graphene, SWCNTs: Single-Walled Carbon Nanotubes, MWCNTs: Multi-Walled Carbon Nanotubes

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