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REVIEW

Research Progress on High Thermal Conductivity Graphene/Copper-Based Composite Heat Sinks for Electronic Equipment

Li Hongzhao[1], **Jiang Haojie**[1], **Pan Jiabao**[1], **Jia Hongsheng**[1], **Chen Minghe**[2], **Chen Yang**[2]

[1] School of Mechanical and Automotive Engineering, Anhui Polytechnic University, Wuhu 241000, China; [2] National Key Laboratory of Science and Technology on Helicopter Transmission, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

Abstract: Graphene/copper-based composite heat sinks demonstrate extensive application potential in military equipment thermal management, high-power electronic packaging, new energy vehicles, and 5G communication systems, due to their outstanding properties, including high thermal conductivity, tunable thermal expansion coefficients, excellent mechanical strength, and low density. However, the industrial-scale application of these composites faces critical challenges during the fabrication of components with complex structures, such as inhomogeneous dispersion of graphene within the copper matrix and poor interfacial bonding between the two phases, which substantially undermine the overall performance of graphene/copper-based composites. To address these issues, the preparation methods for graphene/copper-based composite heat sinks were reviewed. For each method, a rigorous analysis was presented to clarify its inherent advantages and unavoidable restrictions. Furthermore, the latest research progress in addressing three core scientific challenges was synthesized, including uniform dispersion of graphene, interfacial optimization mechanisms, and molecular dynamics simulations for elucidating the structure-property relationships. Finally, the future development directions of graphene/copper-based composite heat sinks in engineering applications were prospected.

Key words: graphene/copper-based composite heat sinks; preparation methods; interfacial bonding; mechanism; molecular dynamics

1 Introduction

Copper is widely used in the electronics and aerospace industries due to its excellent electrical conductivity, exceptional thermal conductivity, and superior corrosion resistance[1–4]. However, its relatively low mechanical properties and poor tribological performance have constrained its applications in harsh or high-demand service environments[5–8]. To meet the diverse operational requirements, composite processing of copper is essential to enhance its comprehensive performance. Graphene, as a new two-dimensional carbon nanomaterial, possesses extraordinary intrinsic properties: Young's modulus reaching 1100 GPa, breaking strength of 125 GPa[9–11], and thermal conductivity as high as 5300 W/(m·K)[12–14]. These outstanding characteristics render graphene an ideal reinforcing phase for metal-based composite heat sinks, making it highly suitable for addressing the performance shortcomings of traditional metal materials in traditional industrial applications[15–16]. Graphene-reinforced copper matrix composites have demonstrated superior mechanical, electrical, and thermal properties compared to pure copper[17–18], leading to increasingly widespread applications and higher integration levels in high-tech sectors, such as military technology, electronics manufacturing, new energy vehicles, and aerospace.

Graphene/Cu composite heat sinks, endowed with ultrahigh thermal conductivity, lightweight characteristics, and resistance to extreme environment, have achieved engineering validation in military, aerospace, and 5G application

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Corresponding author: Li Hongzhao, Ph. D., School of Mechanical and Automotive Engineering, Anhui Polytechnic University, Wuhu 241000, P. R. China, Tel: 0086-553-2215051, E-mail: lhz@mail.ahpu.edu.cn

scenarios. In military systems, under a heat flux of 300 W/cm^2, the airborne active phased-array radar equipped with 0.8wt% graphene/Cu heat sinks achieved a significant reduction in junction temperature of transmit-receive modules from 156 ℃ to 107 ℃ (31% reduction in thermal resistance). For high-energy laser weapons, the integration of these composites led to a 37% improvement in thermal diffusivity (930 W·m^{-1}·K^{-1}), which prolongated the continuous operation time from 120 s to 195 s. In the aerospace sector, graphene/Cu composites applied in low-orbit satellites exhibited exceptional thermal cycle stability: after 500 thermal cycles between –180 and 150 ℃, the contact thermal resistance of growth interface was only 8.2%, far outperforming that of traditional Cu-Mo materials which showed a 23% increase under the same conditions. Additionally, the use of graphene/Cu composites enabled an 18.6% mass reduction in thermal modules. In 5G infrastructure, GaN power amplifier modules integrated with graphene/Cu heat sinks achieved a junction-to-case thermal resistance of 0.28 ℃/W (34% lower than that of modules using pure Cu), sustaining chip temperatures below 85 ℃ at an operating frequency of 28 GHz. For data center liquid cooling systems, these composites reduced the coolant flow rate by 22% while improving the coefficient of performance by 19.3% for 300 W processors. Huawei's 2023 white paper further confirmed a thermal conductivity of 680 W/(m·K) (a 42% enhancement) in 5G Massive MIMO antennas with proven long-term reliability through 3000 h of aging tests under harsh conditions (85 ℃/85% relative humidity). Despite these promising engineering outcomes, three critical barriers persist, impeding the large-scale deployment of graphene/Cu composite heat sinks: high material costs (exceeding $300 per gram of graphene), manufacturing challenges for complex components (graphene nanosheets tend to agglomerate due to strong van der Waals forces, hindering their uniform dispersion in the copper matrix[19–22]), and insufficient interfacial stability (poor graphene-copper wettability results in weak mechanical bonding at the interface[23–27]). To address these issues, this review analyzed four core aspects of graphene/Cu-based composite heat sinks: the preparation methods, strategies for achieving uniform graphene dispersion, interfacial bonding mechanisms, and molecular dynamics (MD) models for performance prediction. Furthermore, it outlined future research directions, aiming to provide theoretical insights to transcend material performance boundaries for their engineering applications.

2 Preparation Methods of Graphene/Copper-Based Composite Heat Sinks

With the rapid advancement of electronic devices toward higher performance, miniaturization, and greater integration, efficient thermal management has become a critical challenge. Graphene/copper-based composite heat sinks, which leverage the exceptional thermal properties, have emerged as a promising solution to this challenge[28–29]. The methods for preparing graphene/copper composite heat sinks primarily include powder metallurgy (PM), spark plasma sintering (SPS), chemical vapor deposition (CVD), electrochemical deposition, and hybrid manufacturing techniques[30–33]. A notable trend is the synergistic integration of multiple techniques into hybrid preparation processes. For instance, CVD is firstly employed to from high-quality, continuous graphene layers on copper substrates, capitalizing on its ability to achieve structurally intact graphene with strong interfacial bonding to the copper matrix. Subsequent processing step is tailored to address specific performance or structural requirements, such as mechanical alloying to refine grains and enhance density/homogeneity. PM (involving compaction and sintering) enables the fabrication of heat sink components with tailored geometries and performance. This hybrid approach effectively solves the shortcomings of single methods, such as poor graphene dispersion uniformity, insufficient interfacial bonding strength, and structural integrity, thereby pioneering a new pathway for the fabrication of high-performance graphene/copper-based heat sinks. Looking ahead, ongoing research and technological innovations are expected to further optimize these hybrid processes, accelerating their widespread adoption in advanced electronic thermal management systems[34–35].

2.1 PM

The preparation of graphene/copper composite heat sinks via PM is a process centered on powder compaction and high-temperature sintering. In this method, copper powder and graphene raw materials are mechanically mixed (e.g., by ball milling) to form a homogeneous composite powder. The powder is then cold-pressed under a high pressure (100–500 MPa) in a mold to create a green compact. Subsequent high-temperature sintering (800–950 ℃) in a protective atmosphere (e.g., high-purity argon) facilitates atomic diffusion and interfacial reactions, enabling metallurgical bonding between copper particles while embedding graphene into the copper matrix. This results in a dense graphene/copper composite heat sink material with excellent thermal conductivity. By precisely controlling compaction parameters and sintering temperatures, the process achieves controlled alignment of graphene and interfacial strengthening, significantly enhancing the thermal conductivity and mechanical properties of the heat sink[36–38].

Singh et al[39] fabricated graphene nanoplatelet (GNP)-reinforced copper matrix composites (Cu-GNPs) using PM, focusing on the effect of GNP content (0.1wt%–1.5wt%) on the mechanical properties of composite. In their method, pure copper powder was blended with GNPs via ball milling with strictly controlled parameters: a rotation speed of 100 r/min, a milling duration of 4 h, and a ball-to-powder ratio of 1:10. The mixed powder was compacted under a pressure of 100 MPa and sintered at 850 ℃ for 4 h to form a dense composite heat sink. Results demonstrated that composites with a GNP content of 1.0wt% achieved compressive strength and hardness of 194 MPa and 167 HV, representing increments of 27% and 53% compared to those of pure copper (153 MPa and 108.9 HV), respectively. This study highlights the potential of PM in preparing GNPs-reinforced copper composite heat sinks and provides valuable references for designing high-performance metal matrix composite heat sinks. Fig. 1 shows the preparation of Cu-GNP heat sink by PM method.

The advantages of preparing graphene/copper composite heat sinks via PM lie in its mature process, low cost, scalability for mass production, tunable thermal conductivity and mechanical properties by adjusting graphene content and dispersion methods, and adaptability to complex shapes[40–41]. However, the drawbacks include the tendency of graphene agglomeration, weak interfacial bonding between graphene and the copper matrix, restricted sintering densification, and the densification inhibition by excessive graphene, leading to increased porosity. This ultimately results in the actual thermal conductivity significantly lower than the theoretical predictions[42–43]. Future improvements should focus on achieving uniform graphene dispersion, enhancing interfacial bonding, and adopting advanced sintering techniques to elevate overall performance[44].

2.2 SPS

SPS is an advanced technique for fabricating graphene/copper composite heat sinks, that combines direct pulsed current heating with pressure-assisted densification[45]. The procedure involves uniform mixing of copper powder and graphene via ball milling or an in-situ chemical method, followed by loading the resulting mixture into a graphite die. Under the simultaneous application of pulsed current and axial pressure, the Joule heating effect induces instantaneous surface activation of copper particles. Concurrently, the discharge plasma removes surface oxides and promotes atomic diffusion between particles, achieving rapid sintering densification within 5 – 20 min. This results in uniform dispersion of graphene within the copper matrix and strong interfacial bonding, ultimately yielding composites with high density (>98%) and superior thermal conductivity (500 – 800 $\mathrm{W \cdot m^{-1} \cdot K^{-1}}$)[46]. By precisely regulating current parameters, heating rates, and pressure, SPS effectively suppresses structural damage to graphene while optimizing interfacial heat transfer performance, thus demonstrating both high processing efficiency and precise microstructural controllability[47].

He et al[48] fabricated copper matrix composites with a 3D graphene-like carbon network (3D-G/Cu) using SPS. Through optimizing copper powder pretreatment, carbon coating/graphitization, and rapid sintering at 800 – 900 °C under a pressure of 50 – 100 MPa, they successfully prepared dense composite heat sinks. Fig. 2 shows the schematic diagram of the key steps. Results demonstrated that the optimized composite (0.65wt% carbon) exhibits an electrical conductivity of 92.3%IACS, approaching that of high-purity copper (101.5%IACS). The introduction of the carbon network significantly enhances hardness: the composite with 2.00wt% carbon reaches a hardness of 78.2 HV, representing a 48% improvement over pure copper (52.8 HV). This study highlights SPS ' s capability to produce graphene/copper composite heat sinks with both high conductivity and high strength, thereby providing critical insights for developing advanced thermal management and power transmission materials.

SPS technique, as an advanced process for fabricating graphene/copper composite heat sink materials, demonstrates significant technical advantages and unique value. First, its distinctive discharge plasma effect effectively activates the surfaces of powder particles, promoting the formation of robust bonding between the copper matrix and graphene. Second, the rapid sintering suppresses abnormal grain growth in copper and structural damage to graphene, preserving their intrinsic properties of both components. Third, through precise control of current parameters, heating rates, and pressure, SPS enables directional graphene distribution and interfacial optimization, achieving a thermal conductivity of 500–800 W/(m·K). However, challenges persist in both material preparation and practical application: high-energy pulsed currents may induce sp^2 bond breakage in graphene, and poor wettability at the copper/graphene interface facilitates the formation of thermal resistance. Additionally, constrained by equipment performance and mold size, large-scale batch

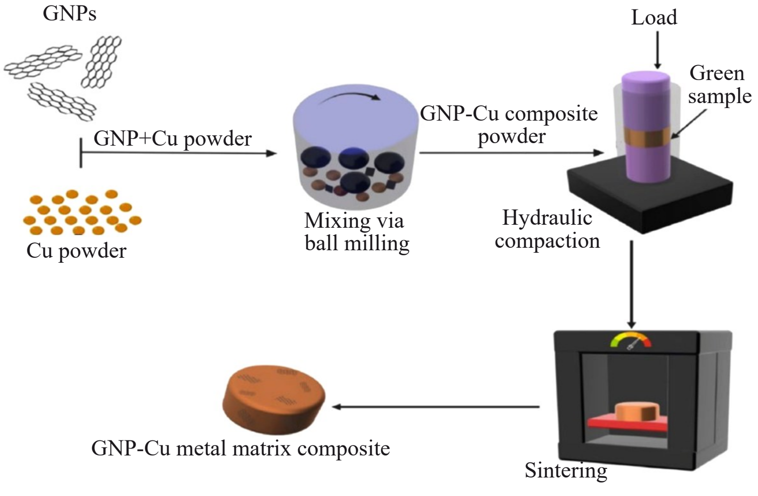

Fig.1 Preparation process flowchart of Cu-GNP heat sink via PM[39]

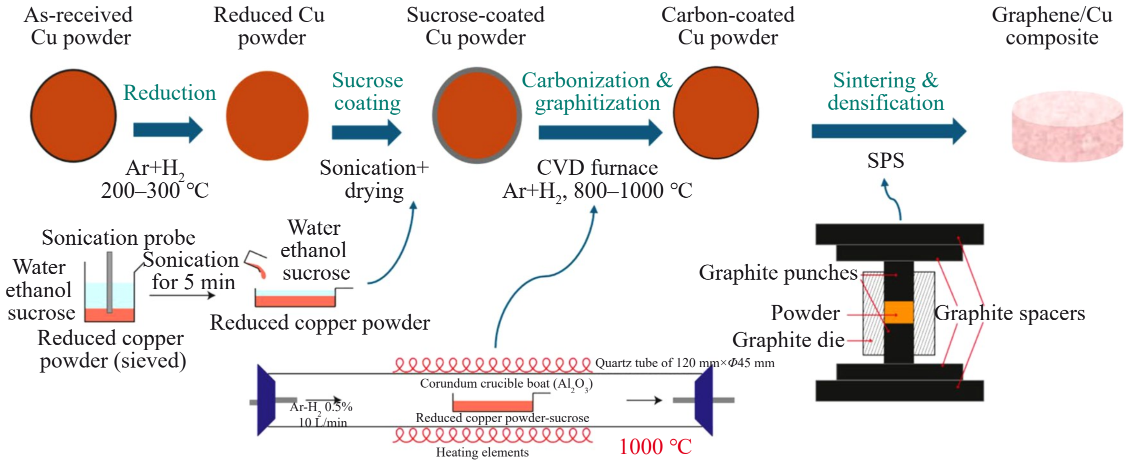

Fig.2 Schematic diagram of key steps of praparation from initial as-received Cu powder to final graphene/Cu composite[48]

production remains not yet feasible, compounded by high production costs. Future research should focus on the surface modification of graphene, interfacial engineering optimization, and multi-field coupled sintering mechanisms to advance its scalable application in electronic thermal management[49].

2.3 CVD

CVD is a synthesis method for fabricating graphene/copper-based composite heat sinks, which employs carbon-containing gases as the carbon source and copper substrates as catalytic growth templates[50]. Under high-temperature conditions and catalytic activation (with the copper substrate serving as both catalyst and growth medium), the carbon precursors decompose into atomic carbon. These carbon atoms subsequently nucleate and grow into graphene layers on the copper surface, thereby forming graphene/copper-based composite heat sinks with robust interfacial adhesion. The CVD furnace used in this approach enables the growth of high-quality and continuous graphene films that exhibit excellent bonding with the copper substrate. Such composites demonstrate superior electrical conductivity, thermal management performance, and structural uniformity, making them promising candidates for advanced thermal dissipation applications.

Guan et al[51] fabricated graphene/copper-based composite heat sinks through a multi-step process, as illustrated in Fig.3 Initially, a Cu-Mn alloy ingot was synthesized by arc-melting electrolytic Cu and Mn at an atomic ratio of Cu:Mn=33:67. The ingot underwent homogenization, quenching, and rolling, followed by dealloying in an HCl solution to yield nanoporous Cu. Subsequently, graphene was grown on the nanoporous Cu substrate via CVD, with final densification of the composite achieved through roll-pressing and sintering. The study revealed three key findings: (1) the nanoporous Cu structure provides abundant nucleation sites, effectively mitigating graphene agglomeration; (2) the CVD-derived graphene exhibits high crystallinity and strong interfacial bonding with the Cu matrix; (3) the formed 3D interpenetrating graphene network enhances electron transport pathways and load-bearing capacity of the composite, resulting in exceptional mechanical properties, specifically a hardness of 55.2 HV and a tensile strength of 330 MPa. These structural advantages position the composite as a high-performance thermal management material.

CVD enables the growth of high-crystallinity graphene, forming continuous films on copper substrates with strong interfacial adhesion. The resulting graphene/copper-based composite heat sinks fabricated via CVD exhibit exceptional electrical conductivity and thermal management performance. However, this method faces challenges in scalability due to its

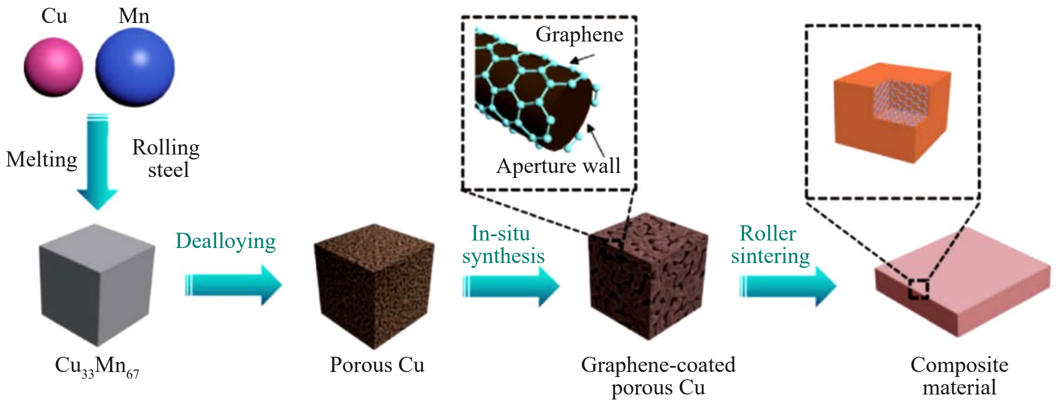

Fig.3 Fabrication process of graphene/copper-based composite heat sink with a 3D interpenetrating network structure[51]

reliance on high-temperature and vacuum conditions, coupled with the high capital costs of specialized equipment. Additionally, its relatively low production yield and high energy consumption further restrict its practical feasibility for large-scale industrial applications[52–54].

2.4 Electrochemical deposition

Electrochemical deposition is a practical method for fabricating graphene/copper-based composite heat sinks, which involves employing copper as the cathode[55] to be immersed in an electrolyte containing graphene dispersions or graphene precursor components. Under electrochemical polarization, copper ions migrate toward the cathode and undergo reductive deposition, while simultaneous co-deposition of graphene with copper occurs through either direct incorporation of graphene sheets or in-situ generation of graphene via electrochemical reduction of carbon precursors at the cathode interface. Subsequent post-treatment processes, including rinsing and drying, yield the final composite structure. This technique demonstrates exceptional adaptability for coating on geometrically complex copper substrates. Moreover, it enables precise control over the composition and microstructure of composite through systematic adjustment of electrochemical parameters such as current density, deposition potential, and electrolyte composition[56–57].

Zhao et al[58] prepared graphene oxide (GO) via a modified Hummers method, as illustrated in Fig.4. The electrochemical deposition was conducted in a system which used a copper foil as the cathode and a copper rod as the anode; the electrolyte for this process contained copper sulfate, nickel sulfate, and GO. By controlling temperature, current, pH value, and deposition time, the team obtained electrodeposited products. Subsequent treatments included centrifugal washing with deionized water, vacuum drying, hydrogen reduction, and removal of large particles, yielding GNP/Cu powder. Finally, the powder was compacted and sintered in atmospheric conditions to fabricate GNP/Cu composite heat sinks. The results demonstrated that copper particles uniformly dispersed on the surface of GNPs, with specific crystallographic orientation relationships between Cu and graphene. The formation of Cu-O-C bonds during electrochemical deposition is confirmed, and the presence of NiO is detected, indicating the existence of Ni. Ni nanoparticles are effectively embedded in graphene to form Ni-GNP hybrid structures, which in turn facilitates the tight connection between Cu and GNPs.

The electrochemical deposition method enables the fabrication of composite heat sinks on complex-shaped copper substrates, as the deposition process conforms precisely to the substrate geometry. Precise control over the composition and microstructure of the composite heat sinks can be achieved through systematic modulation of electrochemical parameters, such as current density and deposition time[59–60]. However, conventional electrolytes often contain cyanide, heavy metal salts (nickel and cobalt salts), and strong acids/alkalis. The toxicity of these components may cause water pollution and occupational health risks. The metal-containing waste liquids generated during electrolyte maintenance require specialized treatment, which significantly increases environmental management costs. In addition, hydrogen precipitation during the deposition process not only affects the plating quality, but also leads to energy waste and explosion safety investment[61–62]. Improvement directions include the following aspects: developing a cyanide-free weak acid electrolyte system, for instance, using degradable ligands (such as citric acid) to replace the traditional cyanides; constructing a closed-loop recycling system for electrolytes, which enable solution reuse through online monitoring and replenishment of metal ions. However, the deposition efficiency of the newly

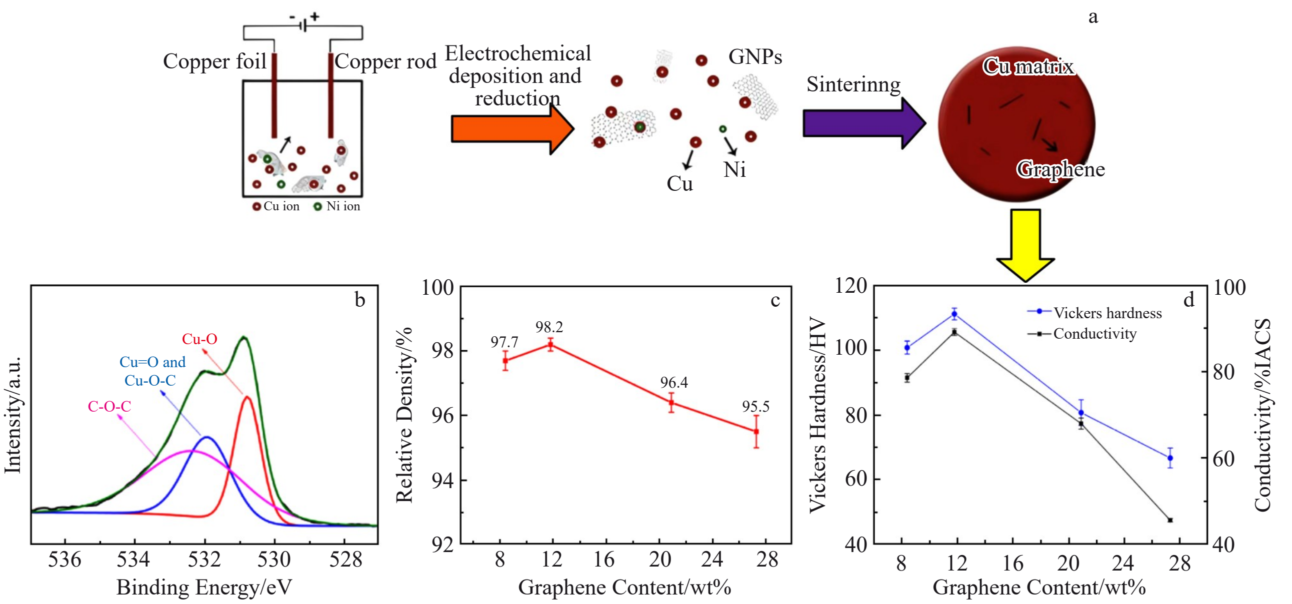

Fig.4 Schematic diagram of fabrication process (a) and property analyses (b–d) of GNP/Cu heat sinks[58]: (b) XPS spectra; (c) relative density; (d) Vickers hardness and conductivity

developed low-toxicity electrolyte systems tends to be reduced by 15%−20% compared to that of the conventional systems. This efficiency gap can be compensated by process optimization, such as pulse deposition. At the policy level, it is recommended to establish a dual-mechanism system combining heavy metal emission taxes and green process subsidies to promote the industry transition toward environmentally friendly manufacturing direction.

2.5 Other preparation methods

In addition to the aforementioned preparation methods, techniques such as electrostatic self-assembly, pulse electrodeposition, and in-situ synthesis are also widely employed for fabricating graphene/copper composite heat sinks. Table 1 shows the performance of graphene/copper composite heat sinks fabricated via different preparation methods. Gorhe et al[63] investigated the effect of GNP content on the compressive properties and thermal conductivity of copper composite foams fabricated via the space-holder method. By employing surfactant-assisted electrostatic self-assembly, they achieved homogeneous dispersion of GNPs within the copper matrix. The results demonstrated that adding 0.25wt% GNPs yields optimal performance: the compressive plateau stress reaches 42±2.7 MPa (a 100% increase compared to that of pure copper foam), the energy absorption capacity attains 21±1.0 MJ/m^3 (114% enhancement), and the thermal conductivity improves to 96.06 W/(m·K) (12% elevation). Changzhou University disclosed a graphene/copper composite heat sink and its preparation method in a patent[64]. This method employs pulse electrodeposition, using alternating deposition baths with two specialized formulations, combined with asynchronous rolling and annealing treatments. The resulting composite exhibits a tensile strength of 483 MPa and a thermal conductivity of 850 W/(m·K), effectively meeting the thermal management requirements for electronic component cooling. Zeng et al[65] developed a copper matrix composite reinforced by a continuously in-situ grown graphene network. At a graphene content of 0.100wt%, the composite demonstrates exceptional mechanical properties: a hardness of 103.7 HV (a 47.8% increase compared to that of pure copper), a yield strength of 181 MPa (a 154.9% improvement over that of pure copper), and a tensile strength of 244 MPa (a 19.0% enhancement relative to that of pure copper).

3 Core Issues of Graphene/Copper-Based Composite Heat Sinks

3.1 Dispersion uniformity of graphene/copper-based composite heat sinks

The fabrication of graphene/copper composite heat sinks inherently faces challenges in achieving uniform graphene dispersion, due to the strong van der Waals forces between graphene sheets and the significant density difference between graphene and copper[66–67]. These factors promote graphene agglomeration within the copper matrix[68–69]. During sintering, agglomerated graphene acts as a barrier that impedes the copper matrix material, resulting in formation of internal pores that severely degrade the mechanical properties[70–71]. Therefore, achieving effective graphene dispersion remains a critical technical challenge for enhancing the performance of graphene-reinforced copper composite heat sinks. Current dispersion strategies are categorized by their medium environments: solid-phase dispersion, liquid-phase dispersion, and solid-liquid interface-assisted dispersion[72].

Solid-phase dispersion predominantly relies on PM: high-energy ball milling is used to ensure uniform dispersion of graphene, followed by sintering to achieve microstructural densification of the composite heat sinks. Yue et al[73] fabricated GO via a modified Hummers method, and subsequently produced graphene nanosheet/copper composites through high-energy ball milling combined with hot-press sintering. The effect of ball milling duration on the mechanical properties of composite heat sinks was investigated. Results revealed an optimal ball milling window: prolonged durations severely degrade the structural integrity of graphene, while insufficient milling durations fail to achieve effective dispersion of graphene within the copper matrix.

Liquid-phase dispersion for graphene/copper composite fabrication primarily employs aqueous or ethanol-based media[74], relying on physical methods such as ultrasonic and electromagnetic stirring to achieve homogeneous graphene dispersion. This graphene dispersion is subsequently combined with copper precursors to form a molecular-level hybrid composite system. Sufficient interactions between copper atoms/ions and graphene sheets ensure highly uniform copper dispersion, either on graphene surfaces or within its interlayer spaces, forming tightly integrated composite heat

Table 1 Performance of graphene/copper composite heat sinks fabricated via different preparation methods

Preparation method	Tensile strength/MPa	Hardness/HV	Thermal conductivity/W·m^{-1}·K^{-1}	Ref.
PM	194	167	380	[30,39]
SPS	-	78.2	332	[31,48]
CVD	330	55.2	503.2	[32,51]
Electrochemical deposition	-	111.2	556.6	[33,58]
Electrostatic self-assembly	-	-	96.1	[63]
Pulse electrodeposition	524	-	1180	[64]
In-situ synthesis	244	103.7	-	[65]
Hybrid fabrication	-	-	400	[34]

sinks. Hu et al[75] synthesized GO via an optimized Hummers method, then prepared graphene/GO hybrid dispersions using molecular-level mixing and ultrasound-assisted dispersion techniques. This hybrid-dispersion product was uniformly coated onto copper foil and vacuum-dried. Due to the catalytic effect of the copper substrate, the GO in the coating was thermally reduced to reduced graphene oxide (rGO). Final densification was accomplished through rolling compression, yielding both rGO/graphene-Cu composites and densified rGO/graphene coatings. Notably, GO acts as a dual-function component, serving as both a surfactant and a binder, effectively addressing graphene agglomeration while enhancing interfacial bonding between rGO and copper. Fig.5 illustrates the fabrication protocol for rGO/graphene-Cu composite heat sinks.

Solid-liquid interface-assisted dispersion combines the principles of solid-phase and liquid-phase dispersion[76]. This method initially involves mixing graphene with copper powder in a solution under appropriate agitation, followed by drying and ball milling. Effective graphene dispersion is achieved through controlling stirring and ball milling durations. By integrating the core advantages of both solid and liquid dispersion techniques, this hybrid approach has been widely employed in fabricating graphene-reinforced copper composite heat sinks.

Despite recent progress in achieving uniform dispersion of graphene in copper-based composite heat sinks, critical challenges persist. The lamellar structure of graphene, combined with its ultrahigh specific surface area[77], strong van der Waals forces[78], and poor interfacial compatibility with copper matrices, promotes agglomeration and results in weak interfacial bonding, ultimately degrading the structural stability of the composite coating. Current mitigation strategies focus on three key aspects: (1) process optimization through mechanical dispersion, ultrasonic dispersion, and in-situ growth techniques[79–80] to enhance dispersion efficiency; (2) interface engineering via additives or surface modifications[81], where rGO and phthalocyanine materials act as both dispersants and bonding enhancers; (3) development of specialized equipment, such as the Graphene Shear Dispersion Machine developed by Harbin Xike Intelligent Equipment Co., Ltd, which significantly improves the graphene dispersion uniformity.

3.2 Interfacial bonding of graphene/copper-based composite heat sinks

The poor wettability between graphene and copper[82], coupled with their restricted chemical reactivity[83], restricts interfacial bonding primarily to mechanical interlocking, often resulting in inferior mechanical properties of composite heat sinks. Nowadays, research on optimizing the interfacial structure between these two components focuses mainly on several key directions, such as matrix alloying, graphene modification, and theoretical simulation and design. Matrix alloying involves introducing carbide-forming elements, such as tungsten, chromium, and titanium, into graphene/copper-based composite heat sinks. After high-temperature sintering, corresponding carbide nanolayers or nanoparticles are generated at the interface between graphene and copper. These sub-interfaces act as bridges, building a solid connection between graphene and copper and effectively enhancing the mechanical and thermal properties of graphene/copper-based composite heat sinks[84–86]. Graphene modification is realized by attaching metal nanoparticles, carbide nanoparticles, or rare earth elements to the surface of graphene, or by graphene derivatives. Graphene modified by nanoparticle loading, on the one hand, can reduce its own surface energy, effectively reducing the phenomenon of agglomeration and thereby achieving good dispersion; on the other hand, it can also significantly improve the interfacial bonding state with copper, thus enhancing the comprehensive performance of the material. Theoretical simulation and design include MD simulation and first-principles calculations. MD simulation mainly studies atomic-level interactions at the interface and predicts the optimal doping elements or functional groups; first-principles calculations can analyze the interfacial bonding energies of different interlayer materials, which can provide a guidance for experimental design. These research directions complement one another. In the future, they may tend to synergizing multiple strategies and combining with artificial intelligence to accelerate material screening, ultimately achieving breakthroughs in the engineering

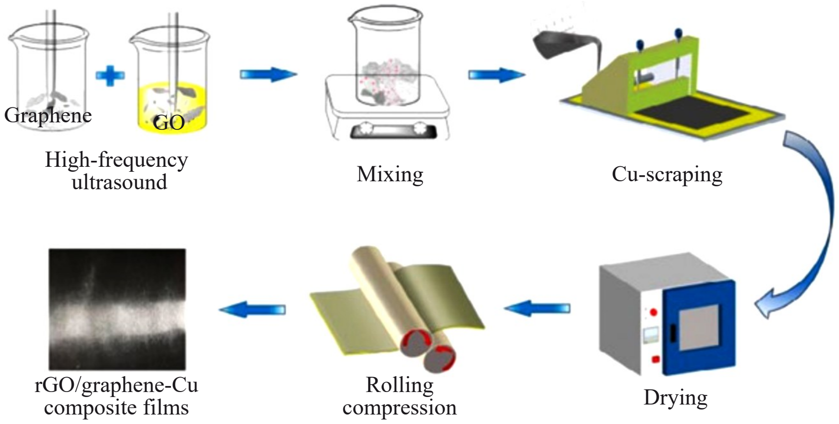

Fig.5 Fabrication process of rGO/graphene-Cu composite heat sinks[75]

applications of graphene/copper-based composite heat sinks in fields such as national defense science and technology as well as thermal management[87–90].

Liu et al[91] fabricated a hierarchical GO-TiC-Cu interfacial structure by synthesizing nano-sized TiC through the reaction between fragmented GO and Ti powder on surfaces of intact GO, achieved through sequential ball milling and SPS, as shown in Fig.6. Multiscale experimental characterizations and first-principles calculations revealed the interfacial strengthening mechanism of the TiC@GO/Cu composite. The semi-coherent TiC-Cu interface (with a 15% lattice mismatch) reduces interfacial energy to – 0.78 eV through strong covalent-metallic bonding, while enhancing the strain hardening rate via Orowan strengthening (by hindering dislocation motion). Concurrently, the coherent GO-TiC interface (with a 4.5% lattice mismatch) repairs defects in GO (evidenced by the Raman spectroscopy analysis of I_D/I_G ratio decreasing from 1.06 to 1.04) and improves load transfer efficiency. These synergistic effects increase the yield strength and ultimate tensile strength of composite by 148% and 26%, respectively, compared to those of pure copper, providing an atomic-scale theoretical foundation for understanding interfacial bonding mechanisms in metal matrix composite heat sinks.

Saad et al[92] achieved uniform dispersion of gold nanoparticles on GNP surfaces through ultrasonication-induced surface defects and functional groups, leveraging the dual effects of C-O-Au chemical bonding and the geometric anchoring of gold nanoparticles. A metallurgically bonded interface was formed via Au-Cu lattice distortion and interdiffusion, reducing interfacial energy and enhancing bonding strength. Experimental results demonstrated that the 1vol% Au-GNPs/Cu composite exhibits a reduced porosity (6.5%), a tensile strength of 214.7 MPa (44% higher than that of pure Cu), and a hardness of 65.5 HV, which is attributed to the synergistic mechanisms of gold nanoparticle pinning, load transfer, and dislocation strengthening. Compared to conventional oxide-modified interfaces, inert nature of Au avoids the formation of brittle interphases while balancing mechanical and functional properties, offering a novel strategy for interfacial design in metal matrix composite heat sinks.

Guo et al[93] developed a continuously confined interface in graphene/copper composite heat sinks through biomimetic design, which employed in-situ synthesized carbon nanotubes (CNTs) to modify GNPs and constructed a 3D interlocking structure. The interface structure is shown in Fig. 7. This interface enhanced bonding strength via mechanical interlocking rather than chemical reactions, achieving an interfacial shear strength six times higher than that of the matrix while preserving the structural integrity of GNP. The resulting composite heat sink demonstrated a yield strength of 280 MPa (50.5% improvement over that of pure Cu) and retained an electrical conductivity of 96.5%IACS. MD simulations and experimental analyses revealed that the confined interface optimizes the strength-toughness-conductivity trade-offs through synergistic mechanisms: efficient load transfer, controlled dislocation accumulation, and crack deflection/bridging (21.4% enhancement in fracture toughness). This strategy addresses the issues of structural damage and performance compromises associated with conventional interface modification, offering a scalable solution for fabricating multifunctional composite heat sinks and advancing interface engineering in advanced thermal management materials.

Shi et al[94] investigated the effects of Sc, Y, and La rare-earth doping on the interfacial bonding strength and mechanical properties of copper/graphene composite heat sinks using first-principles calculations, revealing the atomic-scale strengthening mechanisms. Rare-earth elements induce significant charge redistribution due to their low electronegativity ($\Delta\chi$=0.54–0.8), reversing the electronic state of interfacial Cu atoms from electron-deficient to electron-rich state, accompanied by localized charge accumulation. Concurrently, strong hybridization occurs between rare earth element-d and C-2p orbitals near the Fermi level. Compared to the clean interface, the La-5d/C-2p hybridization peaks are 37% stronger, establishing a covalent bond-dominated interfacial network. This electronic reconstruction reduces the interfacial spacing by 34% (0.22 nm for La doping), enhances the separation work to 1.59 J/m^2 (297% improvement), and increases the tensile strength to 6.22 GPa (226%

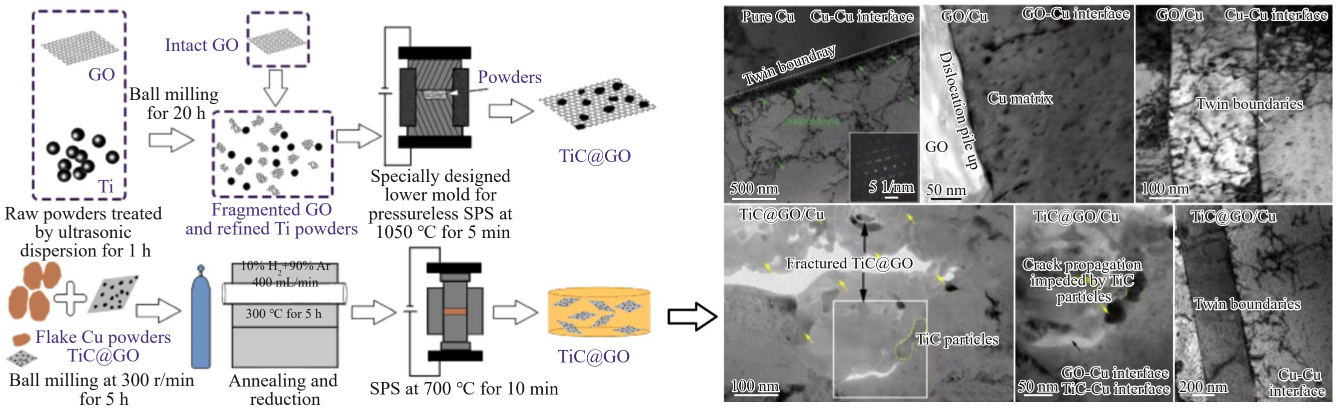

Fig.6 Preparation process of TiC@GO/Cu composite heat sink and its corresponding characterization map[91]

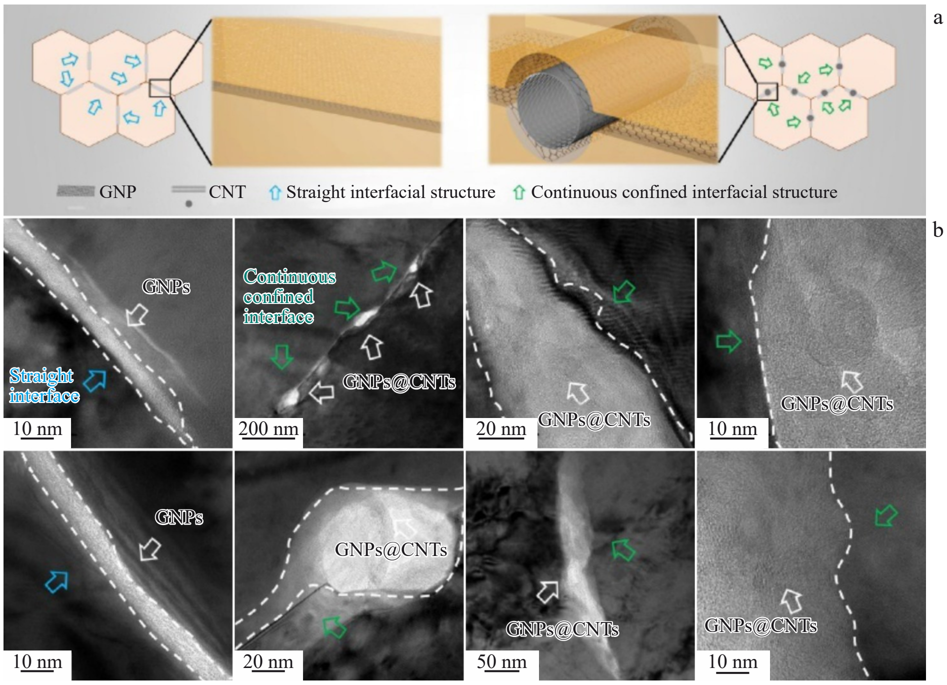

Fig.7 Schematic diagram (a) and microstructures (b) of a composite interfacial structure perpendicular to the normal direction[93]

improvement). Differential charge density profiles of various interfaces calculated by the generalized gradient approximation and van der Waals (GGA+vdw) method are shown in Fig. 8. This study established a quantitative relationship between electronegativity gradients and interfacial reinforcement, offering an atomic-level design strategy for high-strength and high-conductivity Cu-based composite heat sinks.

Current developments in graphene/copper-based composite heat sinks present a coexistence of challenges and advancements in interfacial bonding. The inherent two-dimensional structure of graphene, coupled with its high specific surface area, density mismatch with copper, and poor interfacial wettability[95–96], results in weak graphene-copper bonding that restricts reinforcement efficiency[97]. This deficiency induces phonon/electron scattering and interfacial debonding, ultimately leading to material failure. Recent progress in interfacial optimization focuses on four strategies. (1) Graphene defect engineering[98]: chemical modification of GO followed by reduction, or plasma irradiation-induced defect creation, forms transitional interfacial layers that enhance graphene-copper bonding. (2) Carbon-carbon hybrid reinforcements: covalent bonding between copper and carbon hybrid structures (e. g., CNT-graphene hybrids) improves the efficiency of interfacial load transfer. (3) Nanoparticle decoration: metal/ceramic nanoparticles on graphene interact with both copper matrix and graphene's functional groups/defects, strengthening interfacial adhesion. (4) In-situ synthesis: thermal decomposition of metal-organic frameworks enable the in-situ growth of graphene-based hybrid reinforcements, which optimize interfacial stress distribution through chemically bonded structures[99].

3.3 Strengthening mechanism of graphene/copper-based composite heat sinks

The reinforcement mechanisms of graphene in metal matrix composites have recently emerged as both a research focus and a scientific challenge. In-depth investigation of graphene's strengthening mechanisms in metallic matrices will facilitate performance optimization of graphene-reinforced metal

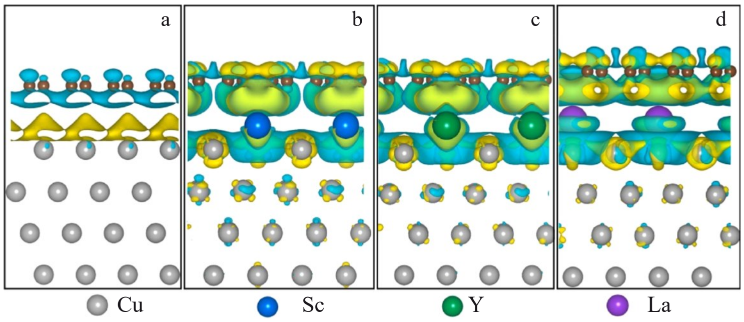

Fig.8 Differential charge densities at different interfaces calculated by GGA+vdw method[94]: (a) clean interface, (b) Sc-doped interface, (c) Y-doped interface, and (d) La-doped interface

matrix composite heat sinks. The unique two-dimensional lamellar structure of graphene enables diverse deformation mechanisms within metal matrices, primarily encompassing load transfer mechanisms, grain refinement strengthening, thermal mismatch-induced strengthening, and Orowan looping mechanisms[100–103].

(1) Load transfer mechanism

The load transfer mechanism in graphene/copper composite heat sinks refers to the process where the matrix material effectively transmits externally applied stresses to the reinforcements, thereby enhancing the overall mechanical performance of the composite. The efficiency of this stress transfer is predominantly governed by the volume fraction of reinforcement and the dimensional ratio of graphene flakes parallel to the loading direction to those perpendicular to loading direction[104]. Higher volume fractions and greater length ratios of reinforcements enable more efficient stress transfer, thereby yielding superior reinforcement effects in composite heat sinks. The predictive equation for reinforcement efficiency is expressed as follows:

$$\sigma_{\mathrm{LT}} = \sigma_{\mathrm{m}}\left[\frac{(4l + t)}{4l}v_{\mathrm{p}} + v_{\mathrm{m}}\right] \tag{1}$$

where σ_{LT} is yield strength of the composite heat sink; σ_{m} is yield strength of the metal matrix; v_{p} is volume fraction of the reinforcement phase; v_{m} is volume fraction of the metal matrix; l is length of the reinforcement phase parallel to the loading direction; t is length of the reinforcement phase perpendicular to the loading direction[105].

The interfacial characteristics between graphene and copper matrices as well as their influence on load transfer mechanisms have been systematically investigated through experimental and computational approaches. For instance, advanced characterization techniques such as high-resolution transmission electron microscope (TEM) have been employed to observe interfacial microstructures and stress distributions, providing critical experimental insights into load transfer processes. Meanwhile, fabrication processes including CVD and PM have been optimized to enhance the interfacial bonding strength of graphene/copper systems, thereby improving load transfer efficiency. Furthermore, significant progress has been made to develop theoretical models that enable more accurate predictions of load transfer behavior in graphene/copper-based composite heat sinks.

(2) Grain refinement strengthening

Grain refinement strengthening enhances the strength of the metallic matrix through grain size reduction. In polycrystalline metals, high-angle grain boundaries impede dislocation motion during plastic deformation. Grains with higher Schmid factors preferentially activate dislocation sources, initiating slip along specific crystallographic slip planes. Dislocations propagating toward grain boundaries become obstructed, preventing direct transmission of plastic deformation to adjacent grains while inducing intragranular dislocation pile-ups. Under external loading, these dislocation pile-ups at grain boundaries generate localized stress concentrations, which provide the critical driving force for activating dislocation sources in neighboring grains[106]. Fig. 9 shows the effect of grain refinement on dislocation slip[107]. Fine-grained materials contain more grain boundaries, leading to dislocation pile-up and resulting in grain refinement strengthening.

Grain refinement strengthening is intrinsically correlated with the Hall-Petch relationship. As described by the Hall-Petch equation, the strength of metallic materials exhibits a positive correlation with grain size reduction within a specific dimensional range. Grain refinement remains one of the most effective strategies for enhancing the mechanical strength of the composite ' s metallic matrix, primarily due to the obstruction of dislocation motion by grain boundaries, which impedes dislocation slip[104]. The Hall-Petch relationship is expressed as follows:

$$\sigma_{\mathrm{LT}} = \frac{k_{\mathrm{y}}}{\sqrt{d}} \tag{2}$$

where k_{y} is the enhancement factor for different metal matrices; d is the average grain size of the grains.

Experimental studies on graphene/copper composite heat sinks reveal that graphene content, flake size, and spatial distribution critically govern the efficacy of copper grain refinement. Precise control of key processing parameters, such as temperature, holding duration, and pressure, enables optimized graphene dispersion within the copper matrix, thereby achieving tailored regulation of grain size. Advanced fabrication techniques such as rapid solidification and hot-press sintering have demonstrated the capability to produce graphene/copper composite heat sinks with refined

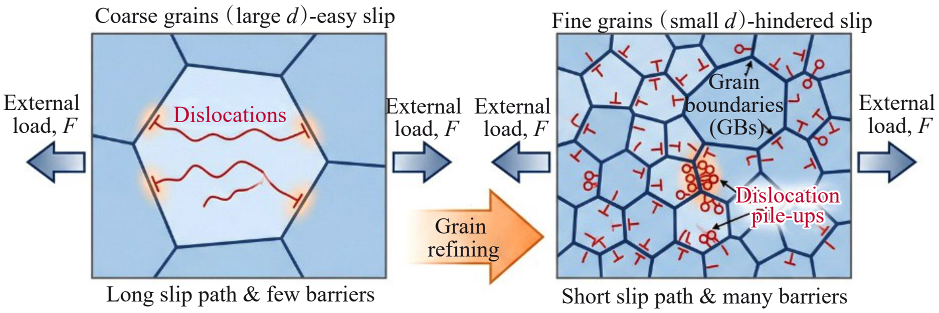

Fig.9 Schematic diagram of effect of grain refinement on dislocation slip[107]

microstructures. Furthermore, investigations into the synergistic effects between graphene and secondary phases or alloying elements provide novel strategies for enhancing grain boundary strengthening mechanisms. These findings thus offer promising pathways for the performance optimization of graphene/copper composite heat sinks.

(3) Thermal mismatch-induced strengthening

Thermal mismatch strengthening in graphene/copper composite heat sinks is induced during sintering or thermal processing, due to the mismatch in coefficient of thermal expansion between the matrix and graphene reinforcements. This mismatch generates localized plastic deformation, creating high-density dislocation regions around graphene, and these regions impede dislocation motion, thereby enhancing the mechanical strength of the composite[108]. The expression for the strengthening effect $\Delta\sigma_c$ due to thermal mismatch can be expressed as follows:

$$\Delta\sigma_c = 1.25\mu b\sqrt{\rho} \tag{3}$$

where μ is the modulus of rigidity of the metal matrix; b is the value of Burgers vector; ρ is dislocation density in metal matrix composite heat sinks. The dislocation density is expressed by the following formula:

$$\rho = \frac{10v\varepsilon_h}{bt(1-v)} \tag{4}$$

where v is volume fraction of graphene in composite heat sinks; ε_h is thermal strain; t is aspect ratio of graphene.

Current research on the thermal mismatch strengthening mechanism in graphene/copper-based composite heat sinks remains restricted, with its focus primarily on theoretical analyses and finite element simulations. Computational models have been developed to quantify the spatial distribution of thermal mismatch stresses and evaluate their impact on mechanical performance. Experimental validation of this mechanism has been conducted through temperature-dependent mechanical property measurements. However, experimental quantification of thermal mismatch stresses remains technically challenging due to measurement complexities and confounding factors, such as interfacial bonding strength, graphene content, and dispersion uniformity. Consequently, comprehensive understanding of the thermal mismatch strengthening mechanism necessitates systematic investigations that integrate multiscale characterization techniques and advanced computational modeling.

(4) Orowan looping mechanism

The Orowan strengthening mechanism refers to the process where dislocations in crystalline materials bypass the second-phase particles by forming dislocation loops, thereby enhancing the strength and hardness of material[109]. This mechanism operates through dislocation pile-up formation under externally applied stress: newly generated dislocations interact with existing obstacles, impeding further dislocation motion and plastic slip. Fig. 10 schematically illustrates the process of dislocation looping around fine particles[1]. According to the strengthening mechanism in graphene/copper composite heat sinks, the shear yield strength (τ_y) of the composite can be expressed via the Orowan equation, as follows:

$$\tau_y = \alpha\frac{GB}{R} \tag{5}$$

where α is a dimensionless constant typically ranging from –0.5 to 1; G is the shear modulus of the metal matrix; B is the value of Parker vector of the metal matrix; R is the radius of curvature of the dislocation.

Research on the Orowan strengthening mechanism in graphene/copper-based composite heat sinks is progressively advancing. Systematic investigations combining experimental observations and theoretical calculations have elucidated the influence of graphene characteristics, including flake dimensions, morphology, content, and dispersion uniformity, on Orowan strengthening efficacy. For instance, optimized fabrication processes enable uniform graphene distribution with tailored dimensions in the copper matrix, thereby maximizing the Orowan strengthening effects. Concurrent numerical simulations reveal the dynamic processes of dislocation-graphene interaction, providing theoretical guidance for performance optimization.

The aforementioned reinforcement mechanisms of graphene in composite heat sinks have been comprehensively validated through both experimental and computational simulations. For instance, Zhang et al[110] reported that the fabricated graphene/copper-based composite heat sink exhibits significantly enhanced mechanical performance due to its 3D graphene nanosheet/Cu structure. As illustrated in Fig. 11, this composite heat sink achieves a remarkable yield strength of 221 MPa and a fracture elongation of 53.8%, representing 126% and 41% enhancements over those of pure copper, respectively. These superior mechanical properties originate from multifaceted synergistic mechanisms. ① The

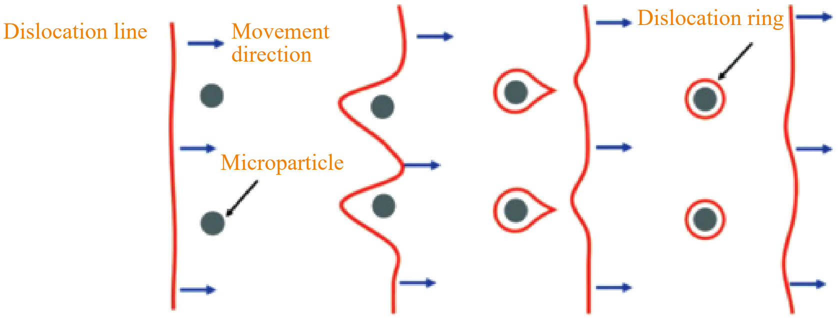

Fig.10 Schematic diagram of dislocation ring formed by dislocation through tiny particles[1]

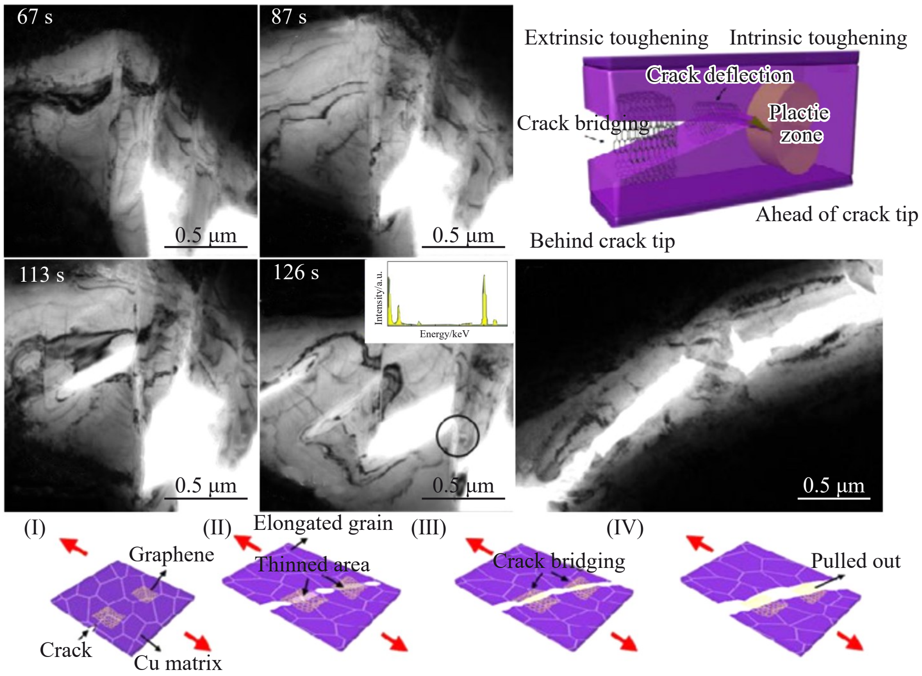

Fig.11 TEM micrographs and schematic illustrations of reinforcement mechanisms in 3DGN/Cu composite heat sink[110]

3D graphene network effectively refines Cu grains following the Hall-Petch relationship, substantially improving yield strength while enabling grain size regulation through controlled volume fraction adjustment. ② The uniformly dispersed 3D graphene nanosheet structure induces dispersion strengthening, with its unique structural interplaying with Cu matrix, which impedes dislocation motion. ③ Strong interfacial bonding facilitates efficient load transfer during tensile deformation, allowing full utilization of the reinforcement effect. ④ Graphene-like nanosheets bridge microcracks strongly obstruct crack propagation, thereby enhancing the fracture resistance and ductility. These coordinated mechanisms collectively endow the composite heat sink with exceptional strength-toughness synergy.

Xu et al[111] achieved notable mechanical performance in graphene/copper composites through interfacial engineering and grain refinement: for a WC-graphene/copper system with 1.0wt% graphene content, the composite exhibits a yield strength of 250 MPa (117.4% increase compared to that of pure Cu) and a tensile strength of 310 MPa (28.1% increase). Whereas, a layered graphene/copper composite with 0.6wt% graphene achieves a yield strength of 317 MPa (78% increase) and a hardness of 121 HV (14% increase). Key innovations include three core techniques: ① a titanium-rich transition layer which improved load transfer efficiency (yield strength of 295.6 MPa, a 239% increase compared to that of pure copper); ② graphene dispersion for the in-situ copper coating; ③ sub-surface embedding behavior of graphene on semi-molten copper, which enabled guided directional growth of graphene. These composites show dual functionality for thermal management and electrical applications, but challenges remain in scalable production and the design of hybrid reinforcement structures. A breakthrough in roll-to-roll synthesis of meter-scale graphene/copper substrates highlights the near-term industrial feasibility for these composites.

Sun et al[112] synthesized a graphene/copper composite heat sink featuring a 3D graphene network loaded with copper nanoparticles. Fig. 12a presents their theoretical model, delineating the quantitative relationships among grain network diameter, graphene network volume fraction, and the respective contribution ratios of Orowan strengthening and load transfer mechanisms. The composite exhibits exceptional mechanical properties, attaining a maximum tensile strength of 290.7 MPa and a yield strength of 231 MPa. The reinforcement mechanisms operate through two distinct pathways: macroscopic pathway and microscopic pathway. Macroscopic pathway: the 3D graphene network assemblies at grain boundaries facilitate coordinated grain deformation, alleviate stress concentration, and enable efficient load transfer and crack bridging. These effects collectively suppress crack propagation effectively, synergistically enhancing both strength and toughness. Microscopic pathway: although Orowan strengthening theory suggests material enhancement through reduced reinforcement diameter or increased volume fraction, experimental observations revealed sintering temperature-dependent complexities in controlling the size and spatial distribution of intragranular 3D graphene network. This discrepancy highlights the critical balance required between theoretical reinforcement predictions and practical microstructural engineering constraints during composite fabrication.

4 MD Investigations of Graphene/Copper-Based Composite Heat Sinks

To fundamentally understand the exceptional performance of graphene/copper composite heat sinks, it is critical to

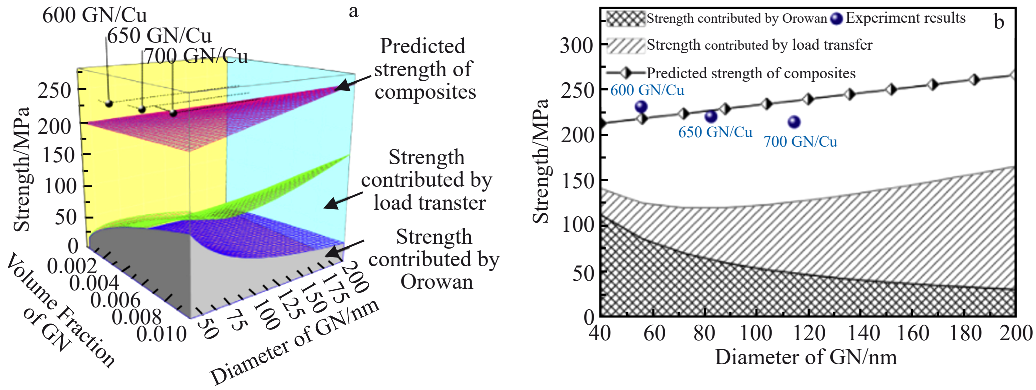

Fig.12 Theoretical model of relationships among grain network diameter, graphene network volume fraction, and contributions ratios of Orowan strengthening and load transfer mechanisms (a); diameter dependence of yield strength contributions in GN/Cu composite heat sink (b) (GN means graphene network)[112]

elucidate their intrinsic microscopic mechanisms. MD, a powerful computational tool in materials science, enables atomic-scale simulations and analyses of the material structures and properties[113]. By constructing atomistic models, MD simulates the material behavior under diverse service conditions, providing atomic-scale insights such as atomic trajectories, interatomic interactions, and defect generation/evolution[114]. These details are indispensable for unraveling the origins of the composite's key performance metrics, including thermal conductivity, mechanical strength, and electrical conductivity. To date, extensive experimental investigations and computational simulations have been conducted to reveal the mechanical performance and reinforcement mechanisms of graphene/metal composite heat sinks[115-116].

(1) Model construction and potential function selection

The accuracy of MD simulations heavily relies on the rationality of atomic models and interatomic potentials. For example, Wang et al[117] constructed four models using MD simulations, including pure copper, graphene/2D-SiC, copper/graphene composite, and copper/graphene/2D-SiC, as shown in Fig. 13. In these models, the copper layers were oriented along the [100], [010], and [001] crystallographic directions. To form the graphene/2D-SiC heterostructures, a 4×4 graphene supercell (lattice constant: 0.246 nm) was matched with a 3×3 2D-SiC supercell (lattice constant: 0.310 nm). This supercell matching strategy resolved the inherent 26% lattice mismatch, thereby ensuring interfacial stability. The selection of interatomic potentials was tailored to balance material characteristics and computational precision. For Cu-Cu interactions, metallic plastic behavior of copper was described by the embedded atom method (EAM) potential. For graphene, C-C bonds in graphene and interlayer interactions were modeled using the adaptive intermolecular reactive bond order (AIREBO) potential combined with the Lennard-Jones potential. For Si-C bonds in 2D-SiC, the Vashishta potential was employed to accurately simulate the mechanical response. This methodology ensures reliable simulations of atomic-scale deformation mechanisms and interfacial stability in composite heat sinks.

Bai et al[118] constructed a 3D MD model of origami-structured graphene/copper nanocomposites by embedding a periodically folded graphene structure, designed based on the Miura-origami pattern (acute angle γ=60°, dihedral angle θ=60°, and side lengths a=1.476 nm and b=2.556 nm) into a face-centered cubic single-crystal copper matrix, as shown in Fig. 14, with an optimized interfacial atomic spacing of 0.28 nm. The interatomic potentials were carefully selected. For graphene, the AIREBO potential was used to accurately describe the C-C covalent bonds within graphene sheets and the torsional behavior; for the copper matrix, the EAM potential was adopted to model metallic bonding; the Lennard-Jones potential (σ=0.328 nm, ε=0.018 eV) was employed to

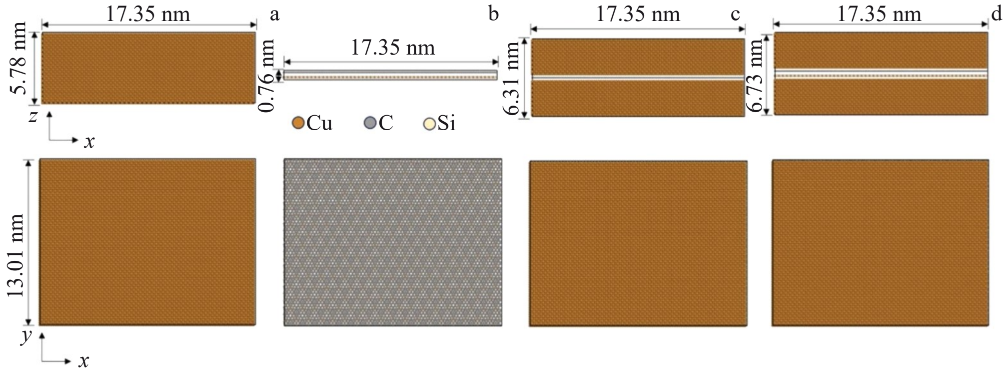

Fig.13 Geometrical models[117]: (a) pure copper, (b) graphene/2D-SiC, (c) copper/graphene, and (d) copper/graphene/2D-SiC

simulate van der Waals interactions at the heterointerface. The stability of model was validated through relaxation under the number-pressure-temperature ensemble, and parametric studies were conducted across temperatures of 4.3–1000 K and mass fractions of 1.72wt%–3.27wt%, which collectively established a multiscale computational framework for unraveling the negative Poisson's ratio mechanism of composite. The combination of tailored potential functions and periodic boundary conditions effectively balanced computational accuracy and efficiency, offering a robust platform for systematic studies of nanoreinforcement-metal matrix interfacial behavior.

(2) Microscopic analysis of interfacial heat transfer mechanisms

Interfacial thermal conductance (ITC) is a critical parameter determining the heat dissipation efficiency of composite heat sinks. Wang et al[119] investigated the ITC enhancement mechanisms at copper-based tri-layer graphene (Cu-TLG) interfaces using non-equilibrium MD simulations, as shown in Fig. 15. The simulation methodology employed the EAM potential for metallic bonding in copper, the AIREBO potential for covalent bonds within TLG and interlayer van der Waals forces, and Lennard-Jones potentials (parameters derived from first-principles calculations, e. g. Ti-C: ε=0.417 eV, σ=0.213 nm) for differentiate chemically bonded (Ti/Co) and physically/mixed bonded (Au/Pd) interfaces. The simulation model adopted free boundaries along the heat flux direction, with a laterally fixed interfacial area of 5 nm×5 nm. Thermal source/sink regions were clamped to maintain a steady-state temperature gradient. After 0.5 ns of relaxation under the NVT ensemble, a 3 ns of heat flux was applied in the NVE ensemble to calculate the steady-state temperature difference (ΔT), from which ITC was derived via Fourier's law: $G=J/(A\cdot\Delta T)$, where J is the heat flux density and A is the interfacial area. Mechanistic analysis revealed that chemically bonded interfaces (Ti/Co) enhance phonon coupling through strong covalent bonds (e. g. Ti-C), exhibiting significantly higher phonon density of states (PDOS) overlap integrals than physically bonded interfaces, thereby facilitating efficient transport of mid-to-high-frequency phonon across the interface. Increasing Ti layer thickness further optimized PDOS overlap and reduced Cu-Ti lattice mismatch, achieving a 32% improvement in ITC. This work elucidates the phonon-mediated synergistic transport mechanism dominated by chemical bonding, providing theoretical guidance for the interfacial thermal management design.

(3) Multi-scale correlation of mechanical strengthening mechanism

MD simulations not only elucidate the thermal transport mechanisms of composite heat sinks but also resolve their mechanical responses. Wang et al[119] revealed the multi-scale mechanical mechanisms of copper/graphene/2D-SiC composite heat sinks under tensile loading through MD simulations. The composite achieves exceptional mechanical performance, with a peak stress of 15.01 GPa and a fracture strain of 0.23, driven by synergistic interactions between the graphene/2D-SiC heterostructure and copper matrix layers. At the microscopic level, plastic deformation in the copper matrix is governed by the crystallographic slip and the nucleation of Shockley partial dislocation. In contrast, the heterostructure delays failure by suppressing the dislocation propagation (65% reduction in dislocation density). Bonding analysis shows that 2D-SiC fractures first due to lower Si-C bond stiffness, whereas graphene dominates load-bearing in the later stages of deformation through bond-angle adjustments and bond-length elongation. Elevated temperatures reduce the initial peak stress by 38.8% via thermal activation-induced softening. High strain rates enhance the strength of composite by 15.6% through dynamic strain hardening. These findings establish a cross-scale correlation between atomic-level defect evolution and macroscopic mechanical behavior, providing theoretical guidance for designing high-strength and high-toughness nanocomposite heat sinks.

(4) Co-validation of simulations and experiments

Current MD research has progressively complemented experimental data. Wang et al[120] combined experimental characterization and MD simulations to elucidate the formation mechanisms of copper/graphene composite nanoparticles in plasma flows. Experimentally, composite nanoparticles were synthesized using a direct current plasma torch in a helium/hydrocarbon gas system. Scanning electron microscope and TEM results revealed a core-shell structure

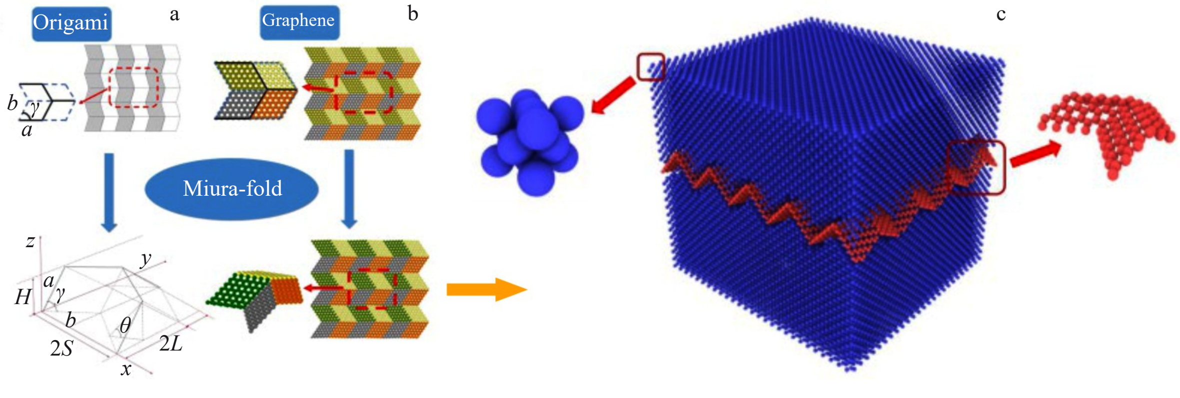

Fig.14 Schematic diagrams of Miura origami (a), Miura origami graphene (b), and origami-structured graphene/copper nanocomposites (c)[118]

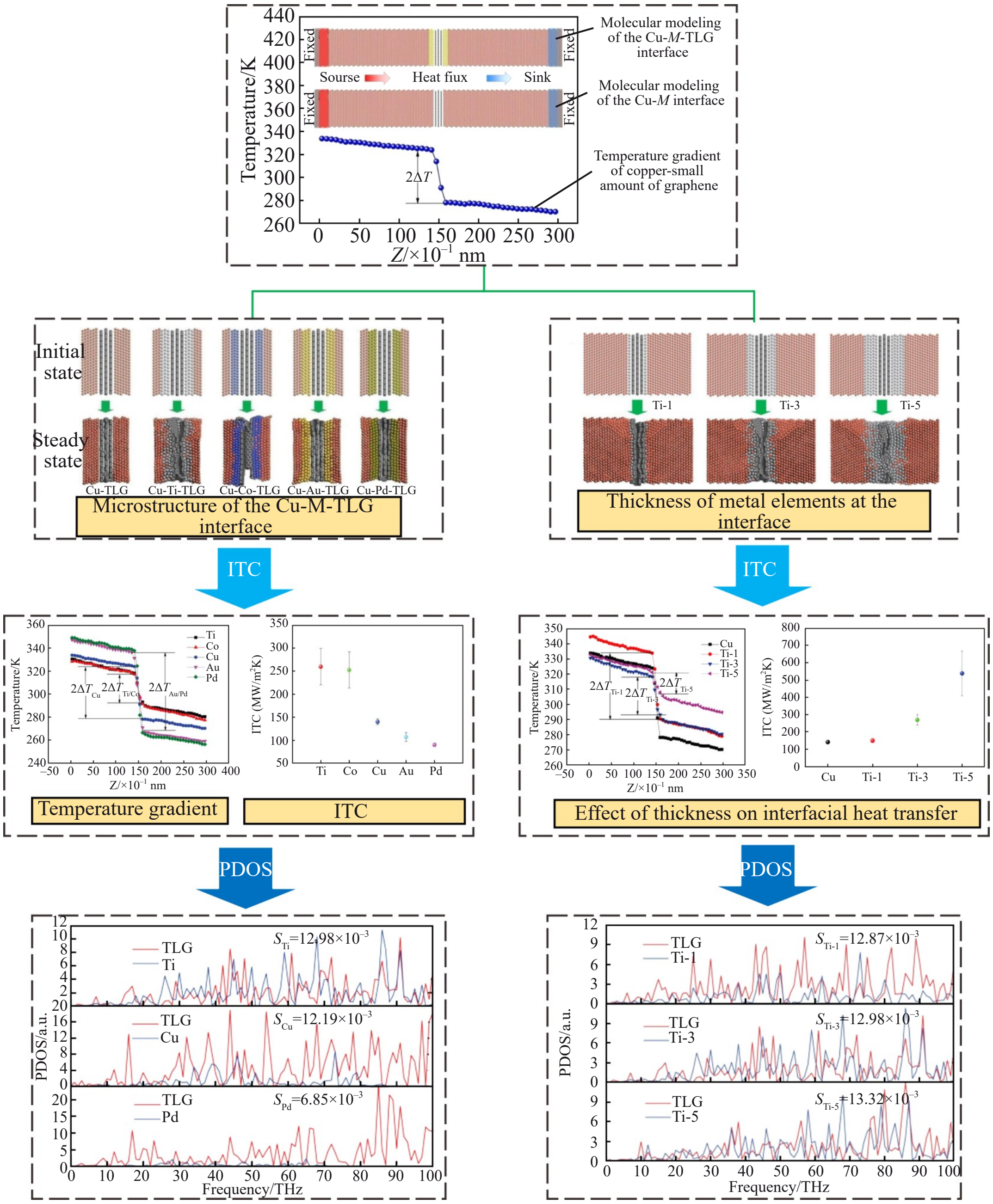

Fig.15 MD modeling of the enhancement mechanism of interfacial metal elements on heat transfer at the Cu/graphene interface[119]

with a copper core of approximately 30 nm in size encapsulated by a graphene shell. X-ray photoelectron spectroscopy analysis confirmed that the copper in the composite existed primarily in the Cu^{2+} oxidation state, while Raman spectroscopy analysis indicated a prominent D-band in the graphene shell (a key signature of defects in graphene). Plasma emission spectra identified a atomic emission line of Cu at 318.7 nm, verifying successful copper incorporation. In simulations, collision models between curved graphene and copper nanoparticles (0.7 and 5 nm in diameters) showed three regimes: low-speed collisions (<1 km/s) led to graphene anchoring and encapsulation, medium-speed interactions (1–5 km/s) caused partial rejection, and high-speed collisions (≥7 km/s) induced penetration or fragmentation. The MD simulation results aligned with experimental observations: low-speed collision mechanisms explained the core-shell structures observed via TEM, simulated velocity distributions matched the plasma flow velocity measurements from experiments, and variations in Raman D-band intensity correlated well with the graphene deformation defects predicted by MD simulations. This experimental-MD multi-scale synergy not only deciphered the kinetic pathways of plasma-driven self-assembly of copper/graphene composite nanoparticles, but also established a theoretical-experimental framework, enabling precise control of composite nanostructures.

MD simulations, endowed with atomic-level spatial and temporal resolution and multi-physics field coupling capability, have unique advantages in revealing the interfacial microscopic mechanisms of materials. They have become a low-cost trial-and-error tool for optimizing composite structures and properties. However, the application of MD simulations is currently constrained by the bottleneck of nanosecond time scales and micrometer spatial scales. Traditional empirical potential functions are insufficiently accurate in describing interfacial chemical reactions, and the cross-scale interface between microscopic mechanisms and macroscopic properties has not been fully resolved. In the future, it is necessary to integrate machine learning potential function, multi-scale simulation algorithms, and supercomputing technologies to break through the balance bottleneck of precision-efficiency-scale of MD simulations, promoting MD from a tool for phenomenal interpretation to one for engineering design. Ultimately, it will accelerate the large-scale application of graphene/copper-based composite heat sink in fields such as power transmission and electronic devices, bringing new opportunities and transformative changes for the development of related industries[121–123].

5 Conclusions and Outlook

Common preparation methods for graphene/copper-based composite heat sink include PM, SPS, CVD, and electrochemical deposition. The function of graphene in these composites primarily depends on its dispersion uniformity within the copper matrix and the interfacial bonding strength between graphene and copper. Uniform graphene dispersion is critical for maximizing its reinforcing effects, while robust interfacial bonding ensures efficient load transfer and mechanical enhancement. Although notable advancements have been achieved in optimizing these preparation methods of graphene/copper composites, several critical challenges remain. For example, the initial investment in SPS equipment exceeds \$500 000, and the production cycle for a single batch is 6–8 h, resulting in a 3.2-fold higher unit production cost compared with that of the traditional PM method. In continuous production mode, the electrochemical deposition method exhibits an interlayer stripping rate exceeding 15%. To promote the practical application of graphene/copper-based composite heat sinks, future research should focus on addressing the following aspects.

1) Development of new equipment and preparation technique. Current preparation methodologies, including CVD, exhibit distinct advantages but face restrictions in scalable production. These constraints stem from intricate processing requirements and high equipment/raw material demands for specific applications. For instance, the CVD method relies on ultra-high-purity copper foil and high-cost graphene precursors, resulting in a single-batch production cost of \$480/m^2. Additionally, CVD is difficult to integrate into continuous production lines. Future research should prioritize developing innovative fabrication methodologies to address the core challenges of graphene dispersion uniformity and agglomeration. A promising example is plasma-assisted ball milling technique, which has achieved 92% graphene dispersion uniformity in copper matrices. To advance this direction, research should focus on three key areas: investigating high-efficiency surface modification strategies or standardized processing protocols, exploring novel eco-friendly dispersants or hybrid processing techniques, and optimizing composite structures to enhance both dispersion uniformity and interfacial compatibility between graphene and the copper matrix. Systematic optimization of these parameters will enable scalable manufacturing of high-performance graphene/copper-based composite heat sinks.

2) Research on dispersion mechanism and interface regulation technique. Graphene exhibits poor dispersibility in the copper matrix, with agglomeration being a common issue. This agglomeration generates internal defects. For instance, during thermal cycling tests, the interfacial thermal resistance increases by up to 28%, weakening the reinforcement effect of graphene, and ultimately degrading the comprehensive performance of graphene/copper-based composite heat sinks. Although existing methods such as surface modification and ultrasonic dispersion are widely used to improve dispersibility, their effectiveness remains restricted. In addition, good interfacial bonding is another key factor for effective load/phonon transfer from graphene to the copper matrix, but the vastly different physical and chemical properties of the graphene and copper result in weak interfacial forces. Currently, the interfacial bonding strength between graphene and copper is only 58–75 MPa. Current strategies to address this issue, such as optimizing processing parameters or introducing intermediate interlayers, have shown restricted success in enhancing interfacial adhesion. Structural stability and operational reliability under complex working conditions require further enhancement to fully exploit reinforcement potential of graphene. Future research should prioritize two fundamental directions: fundamental investigations into dispersion control mechanisms for achieving atomic-scale graphene uniformity and innovative interface structures for strengthening interfacial bonding strength and stability.

3) Multi-scale modeling and experimental validation. Prioritize the deep integration of MD simulations and experimental research. First, use multi-scale simulations to predict the performance of graphene/copper-based composite heat sinks. Second, leverage simulation results to guide experimental design (e. g., machine learning-assisted interfacial energy databases containing 2000+ typical interfacial configurations). Third, experimentally validate and correct the simulation results (combined with the strength prediction error of <8%), and establish an accurate multi-scale model. Combining artificial intelligence and machine learning algorithms can accelerate material property prediction and optimization to achieve efficient research and development and design of graphene/copper-based composite heat sinks.

4) Military and civil industrialized applications. Graphene/copper-based composite heat sinks with high thermal conductivity show dual industrialization potential in both military and civil fields. In military scenarios, this composite can significantly improve the thermal management performance of high-power electronic equipment. Key

applications include airborne phased array radar (the composite heat sink reduced the temperature difference of transceiver components from 32 ℃ to 9 ℃), high-power laser weapons, and military electronic countermeasures systems (the lightweight characteristics and electromagnetic interference shielding ability of composite meet the strict requirements of military equipment in complex battlefield conditions). However, a critical bottleneck for military application lies in interlayer interface stability in batch production. In civil sectors, typical application scenarios include 5G base station chips (the anisotropic thermal conductivity ratio of composite of 8: 1 supports a 26% reduction in chip size), new energy vehicle IGBT modules (a 0.3 mm-thick composite heat sink reduces the junction temperature by 19 ℃), and ultra-thin folding terminals (the anisotropy and high thermal conductivity of composite support the miniaturization of the device and the development of high-power density). However, it is necessary to further reduce the raw material and process costs. To promote large-scale application, future industrialization needs to focus on three core tasks: synergistic breakthrough between military and civil technologies, cost optimization of civil processes, and gradient structure design for multi-scenario adaptation to match the diverse heat flow density requirements.

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电子装备高导热石墨烯/铜基复合热沉研究进展

李宏钊[1]，姜豪杰[1]，潘家保[1]，贾红升[1]，陈明和[2]，陈　阳[2]
(1. 安徽工程大学 机械与汽车工程学院，安徽 芜湖 241000)
(2. 南京航空航天大学 直升机传动技术国家重点实验室，江苏 南京 210016)

摘　要：石墨烯/铜复合热沉凭借高导热、热膨胀系数可调控、高强度及低密度等特性，在军工装备热管理、高功率电子封装、新能源汽车及5G通信等领域具有广泛的应用前景。然而，复杂结构件制备工程中石墨烯与铜分散不均匀、界面结合不良等问题会严重影响石墨烯/铜基复合热沉的综合性能，限制其工程化应用。基于此类问题，总结了石墨烯/铜基复合热沉的制备方法，并对其优缺点进行了分析；综述了石墨烯/铜基复合热沉的均匀分散性、界面结合、作用机制及分子动力学模拟等关键问题，最后展望了石墨烯/铜基复合热沉在工程应用中的发展前景。

关键词：石墨烯/铜基复合热沉；制备方法；界面结合；作用机制；分子动力学

作者简介：李宏钊，男，1989年生，博士，安徽工程大学机械与汽车工程学院，安徽 芜湖 241000，电话：0553-2215051，E-mail：lhz@mail.ahpu.edu.cn