



Scalable graphene fiber arrays for superior thermal interface materials with high reliability

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ABSTRACT

In response to the thermal dissipation challenges faced by high-power-consumption chips under the trend of artificial intelligence, developing thermal interface materials (TIMs) based on highly thermally conductive graphene fibers (GFs) with high flexibility presents a promising strategy. Here, a flexible GF-based TIM (GF-TIM) was fabricated by an efficient stacking-cutting strategy based on large-area aligned GF arrays. The resulting material features high orientation degree (0.89) with a filler content of 39.0 wt%, and an array density of 0.44 g cm⁻³ (labeled as GF-TIM-40). Benefiting from continuous phonon transport pathways constructed by the highly oriented and thermally conductive GFs, the GF-TIM-40 achieves a through-plane thermal conductivity ranging of 190.22 W m⁻¹ K⁻¹, which is 2.33 times that of metallic indium foil (81.8 W m⁻¹ K⁻¹). By comprehensively evaluating thermal conductivity and contact thermal resistance, the GF-TIM-40 exhibits an excellent interfacial heat dissipation performance. Under a typical packaging pressure of 50 psi, the GF-TIM-40 (0.5 mm thickness) shows an effective thermal resistance of 0.11 K cm² W⁻¹ and a contact thermal resistance of 0.055 K cm² W⁻¹. Compared to commercial pitch-based carbon fiber thermal pad, GF-TIM-40 achieves an additional temperature reduction of 14 °C at the heat source under a power density of 25 W cm⁻². This work promotes the large-scale manufacturing and practical applications of GF-TIM for high-power electronic devices.

1. Introduction

The escalating power density of artificial intelligence chips has positioned thermal management as a critical bottleneck, driving the urgent requirements for high-performance thermal interface materials (TIMs) [1–6]. The efficacy of TIMs is governed by total interfacial thermal resistance, which comprises the intrinsic bulk resistance and the

contact resistance at interfaces [7–9]. Traditional composite TIMs, reinforced with isotropic spherical or 2D fillers [10–14], face an inherent limitation. While high filler contents are requisite to establish thermally conductive percolation networks, they inevitably compromise mechanical properties by drastically increasing viscosity and compressive modulus [15–18]. The resulting stiffness impairs the mechanical compliance to conform to microscopic surface roughness, leading to a

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surge in contact thermal resistance that negates the benefits of enhanced intrinsic thermal conductivity [19–22]. Consequently, resolving this fundamental trade-off between thermal conductivity and mechanical compliance remains a formidable challenge in TIM design.

Constructing vertically aligned architectures using one-dimensional (1D) carbon fillers offers a potential pathway to decouple this trade-off by maximizing thermal enhancement efficiency. By aligning carbon fibers (CFs) along the heat flux direction via electric/magnetic fields [19,23] or directional freeze-casting [24], significant enhancements in through-plane thermal conductivity have been achieved. For instance, Yu et al. reported a record-high conductivity of $141.57 \text{ W m}^{-1} \text{ K}^{-1}$ by magnetically aligning CFs at a high loading of 60 wt% [19]. Despite these advances in intrinsic thermal conductivity, critical limitations persist. Conventional CFs with high thermal conduction are inherently rigid and brittle, and their chemically inert surfaces often lead to weak interfacial adhesion with the matrix. Therefore, these rigid vertical arrays exhibit poor mechanical compliance, yielding high contact thermal resistance under low packaging pressures. Furthermore, under high compressive strains, such rigid arrays are prone to structural collapse or brittle fracture rather than elastic deformation, permanently severing the effective thermal pathways [25–27]. Therefore, developing a high-performance TIM that combines metal-level thermal conductivity with excellent compressibility is an urgent yet unresolved priority.

Graphene fibers (GFs), assembled from 2D graphene sheets, emerge as an ideal candidate to bridge this gap. GFs exhibit outstanding axial thermal conductivity ($1290\text{--}1480 \text{ W m}^{-1} \text{ K}^{-1}$) and significantly lower Young's modulus ($135\text{--}341 \text{ GPa}$), as well as superior flexibility compared to conventional pitch-based CFs [28,29]. These features theoretically allow for efficient phonon transport without sacrificing interfacial compliance. However, translating the single-fiber properties into macroscale TIMs has proven to be difficult. Our group previously attempted a dual-field synergistic alignment strategy, but the achievable filler content was capped at $\sim 17 \text{ wt\%}$ with a low array density (0.335 g cm^{-3}), limiting the thermal conductivity to $82.4 \text{ W m}^{-1} \text{ K}^{-1}$ [30]. The crux of the challenge lies in densifying GF arrays to maximize phonon transport capacity, while simultaneously preserving a low compressive modulus and ensuring the structural integrity and stability of the aligned architecture under compression.

By employing a stacking-cutting strategy using aligned graphene fiber mats (AGFMs), we fabricated a GF-TIM featuring high array density, high vertical alignment, and continuity of the GFs. The material exhibits a through-plane thermal conductivity of $190.22 \text{ W m}^{-1} \text{ K}^{-1}$ at a filler content of 39.0 wt% and an array density of 0.44 g cm^{-3} . Compared with the GF-TIM previously reported by our group, it demonstrates a 1.31-fold higher array density and a 2.31-fold higher thermal conductivity. Benefiting from the low modulus of GFs, the TIM exhibits good thixotropy even at 39.0 wt% filler loading, achieving 50% compression under 71.6 psi stress, along with a low contact thermal resistance of $0.055 \text{ K cm}^2 \text{ W}^{-1}$. This work presents a high-performance TIM that combines excellent thermal conductivity, mechanical properties, and processability, promising scalable fabrication for thermal management in next-generation high-power electronic devices.

2. Experimental section

2.1. Materials

Graphene fiber (GF) filaments with an average diameter of $\sim 15 \mu\text{m}$ were supplied by Hangzhou Gaoxi Technology Co., Ltd. (www.gaotech.com), corresponding parameters are displayed in Table S1.

Organic silicon sealant (195T, transparent, two-component) was purchased from Ausbond. Vinyl silicone oil (viscosity 1500 mpa.s) was purchased from Runhe High-tech Materials.

The commercial CF-TIM (WT 5935C-250, $80 \text{ mm} \times 80 \text{ mm} \times 0.5 \text{ mm}$) was purchased from Tianjin Waermtime New Material Technology Co., Ltd. (www.waermtime.com), corresponding through-plane thermal

conductivity is $22.5\text{--}27.5 \text{ W m}^{-1} \text{ K}^{-1}$. The commercial Si-TIM (TS-6, $85 \text{ mm} \times 45 \text{ mm} \times 0.5 \text{ mm}$) was purchased from Zhongshan COXBYTE Electronics Co., Ltd., corresponding through-plane thermal conductivity is $16 \text{ W m}^{-1} \text{ K}^{-1}$.

2.2. Preparation of GF-TIM

Continuous graphene fibers were aligned onto a $15 \text{ cm} \times 15 \text{ cm} \times 0.5 \text{ cm}$ graphite substrate using an automated filament winding system with a fixed winding angle of 85° and winding speed of 10 cm s^{-1} to fabricate an aligned graphene fiber mat (AGFM). The silicone matrix precursor was prepared by mixing 195T(A), 195T(B), and vinyl silicone oil at a mass ratio of 40:1:60. The mixture was homogenized and degassed in a planetary centrifugal vacuum mixer for 10 min to eliminate entrapped air. Subsequently, the AGFM was immersed in the silicone precursor and subjected to vacuum degassing for 10 min. This step ensured the thorough removal of air voids and promoted the complete infiltration of the matrix into the fiber array. Following impregnation, the composite was densified via a pre-rolling process and peeled from the substrate to yield the silicone-based GF prepreg. The prepregs were stacked unidirectionally layer-by-layer in a square mold and hot-pressed to cure the matrix under defined high of 4 cm and 80°C , forming a highly-oriented composite. The GF content of GF-TIM was designed by adjusting the number of prepreg layers and the final compression height. Complete infiltration of silicone by AGFM prior to pressing is essential to prevent cracking in the resulting composites. Despite a pre-rolling process, the inherently loose structure of the graphene fiber tow necessitates relatively high pressure during prepreg pressing, particularly at high filler contents, which tends to expel a substantial amount of silicone. As a result, insufficient infiltration can readily lead to cracking in the final material. Finally, the cured composite block was sliced perpendicular to the fiber alignment direction using a diamond wire saw (DX-2318, DongheScience, Co., Ltd), exposing the fiber cross-sections to form the final GF-TIMs (Unless specified, GF-TIM-x contains $x \text{ wt\%}$ GFs.).

2.3. Characterizations

The morphology and structure of the samples were characterized by SEM (EVO 10 SEM, 5.0 kV). A thin layer of gold was sputtered on the surface of the sample before observation. The GF azimuthal angle distributions were analyzed with ImageJ (NIH). The IR photos were captured by using a calibrated infrared camera (T630sc, FLIR, USA). All samples were coated by a thin graphite layer ($\sim 5 \mu\text{m}$) to ensure the same infrared emissivity for accurately recording surface temperature.

2.3.1. Measuring thermal conductivity of TIMs

The thermal conductivity of TIMs was calculated using equation $K = \alpha \times C_p \times \rho$, where α is the thermal diffusivity, C_p is the specific heat capacity, and ρ is the density of the sample. Therein, the thermal diffusivity (α) was measured by laser flash method on a LFA467 HyperFlash instrument (Netzsch, Germany) at 25°C under atmospheric conditions. Before thermal conductivity characterization, the sample was cut into $10 \times 10 \text{ mm}^2$ square and coated with a thin layer of graphite spray (Graphit 33, KONTAKT CHEMIE, Germany) on both the top and bottom surfaces to prevent permeation of the heating light and to allow detection of the radiation temperature. The density ρ (g.cm^{-3}) was measured according to the Archimedeian method. The specific heat capacity C_p ($\text{J.g}^{-1} \text{ K}^{-1}$) was evaluated using a DSC300 (Netzsch, Germany) in an N_2 atmosphere at the rate of $10^\circ \text{C.min}^{-1}$. All the thermal conductivity results were obtained by averaging five separate measurements, with error bars representing the standard deviation ($n = 5$ independent replicates).

2.3.2. Measuring thermal resistance of TIMs

The Longwin 9389 MD steady-state heat-flow method, in accordance

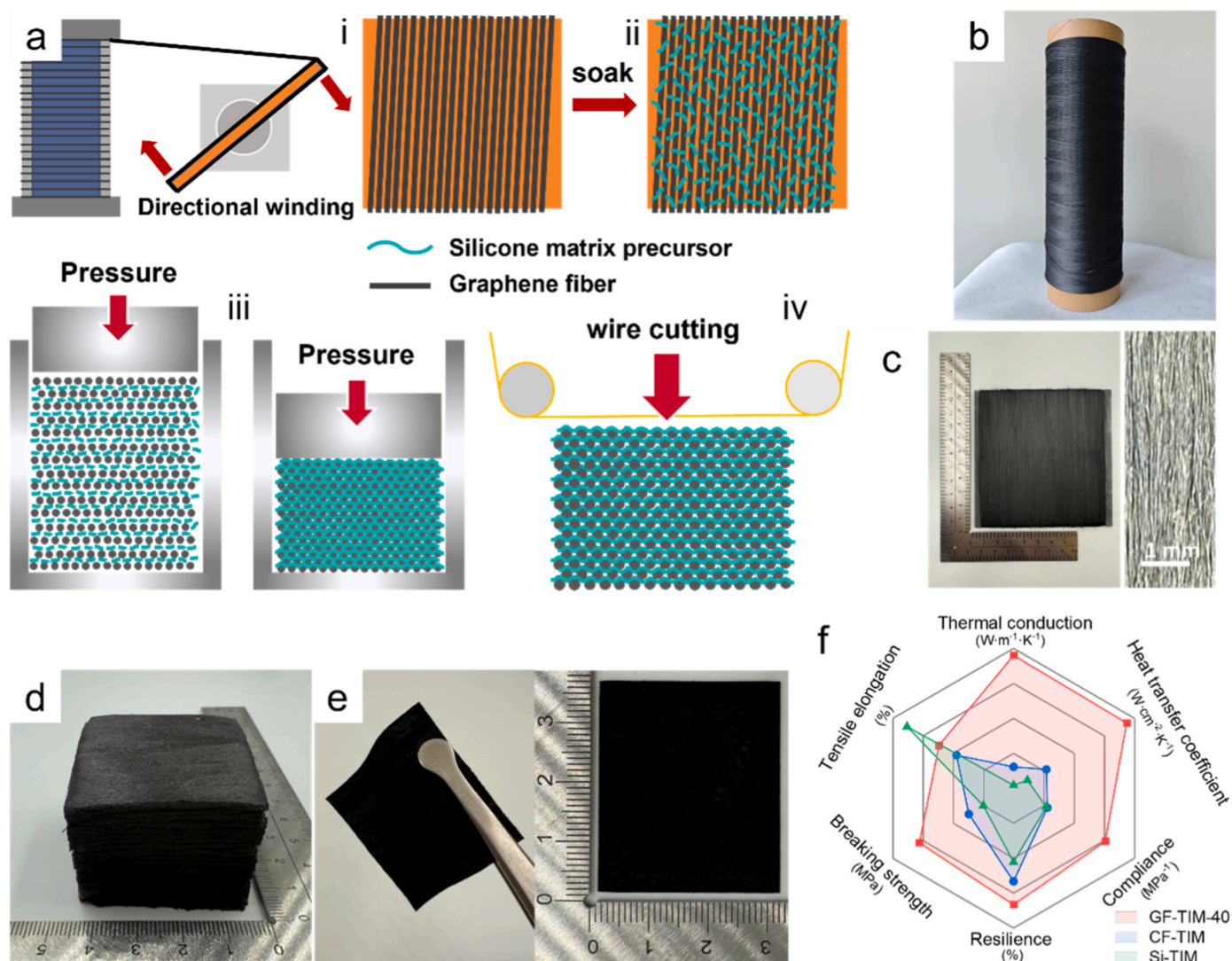


Fig. 1. Fabrication of GF-TIMs. (a) A schematic diagram of the GF-TIM prepared by the stacking-cutting strategy: (i) winding GFs onto a substrate to form an AGFM; (ii) impregnating the AGFM with silicone gel to obtain a prepreg; (iii) stacking and cutting the prepreg of AGFM to obtain GF-TIMs. The black line/circle represents graphene fibers, the blue line denotes the silicone matrix precursor, the orange plate indicates the graphite substrate, and the yellow line corresponds to diamond wire. (b and c) Photographs of the GF filaments and an AGFM. (d and e) Photographs of the AGFM/silicone composite before and after cutting. (f) Comparison of overall performances, including thermal conductivity, compressive compliance, and tensile properties of the GF-TIM, commercial carbon fiber-based TIM (CF-TIM, Wearthmo, promote $25 \text{ W m}^{-1} \text{ K}^{-1}$), and alumina-based TIM (Si-TIM, COXBYTE, TS-6, promote $16 \text{ W m}^{-1} \text{ K}^{-1}$). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

with ASTM D5470 standard, was used to analysis the K_{eff} , R_c and R_{eff} of the TIMs over a surface area of $25.4 \times 25.4 \text{ mm}^2$ under varying pressures. The thickness of the TIM under examination at different pressures is denoted as BLT. The contact thermal resistance was calculated by extrapolation based on three samples of different thicknesses.

2.3.3. Measuring mechanical properties of TIMs

The compressive and tensile properties of TIMs were tested using a universal testing machine (UTM, model 5567A, Instron, USA). Compression tests were performed on $20 \text{ mm} \times 20 \text{ mm}$ specimens at a loading rate of 5 mm min^{-1} . Tensile properties were evaluated using $25 \text{ mm} \times 10 \text{ mm}$ strips at a crosshead speed of 100 mm min^{-1} .

3. Results and discussion

3.1. Preparation and characterization of GF-TIMs

Aligning graphene fibers (GFs) along the thickness direction of TIMs

enhances the through-plane thermal conductivity. Using a stacking-cutting strategy based on AGFMs, we fabricated GF-TIMs with high vertical orientation and array density. The typically preparation process of GF-TIMs is illustrated in Fig. 1a. The AGFM was fabricated by winding continuous GFs (Fig. 1b) onto a graphite substrate at a fixed angle by an automatic winding system (Fig. 1a-i). The GFs display a high degree of orientation along the winding direction (Fig. 1c). The AGFM was then impregnated with an adhesive of silicone matrix precursor. After densification via roll-pressing process, it was peeled from the substrate to obtain a silicone-based prepreg of the AGFM (Fig. 1a-ii). The prepreps were stacked layer-by-layer in a square mold along the same direction and cured under defined pressure and temperature (Fig. 1a-iii) to obtain a highly-oriented graphene-fiber/silicone composite (Fig. 1d). The GF content and array density were adjusted by varying the amount of AGFMs and the compression height. Finally, the composite was cut perpendicular to the direction of GFs using a wire-cutting machine (Fig. 1a-iv and S1a), yielding the GF-TIM (Fig. 1e-S1b and c) with excellent comprehensive performance (Fig. 1f and Table S2).

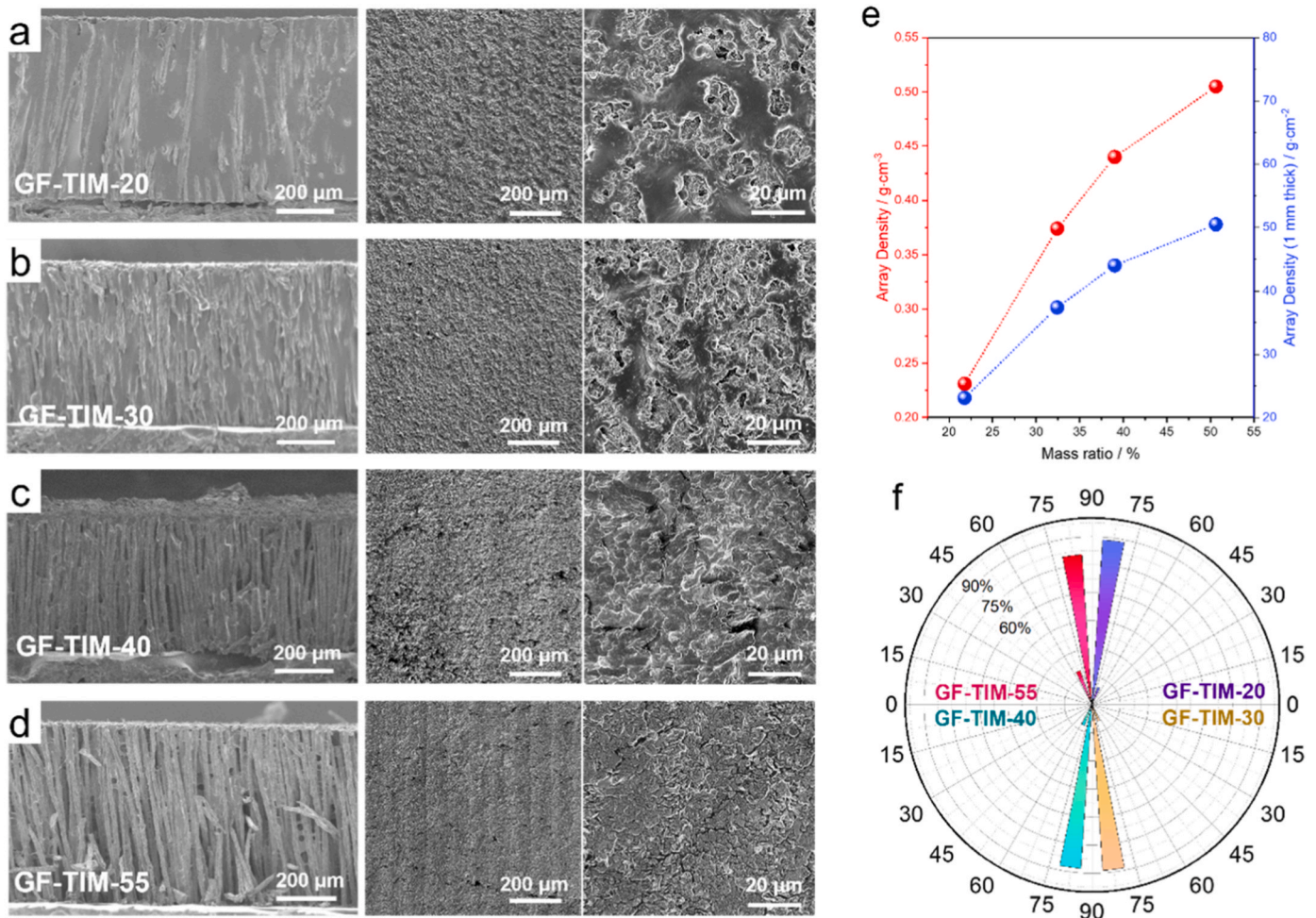


Fig. 2. Structural characterization of GF-TIMs. (a) The cross-section (left panels) and surface morphology (right panels) of GF-TIM-20, (b) GF-TIM-30, (c) GF-TIM-40, and (d) GF-TIM-55. (e) Array density of graphene fibers with various filler contents. (f) Fiber orientation of GF-TIM-20, GF-TIM-30, GF-TIM-40 and GF-TIM-55.

Fig. 2a–d (left panels) present the cross-sectional morphologies of GF-TIMs with filler contents of 21.6, 32.3, 39.0, and 54.2 wt%, labeled as GF-TIM-20, GF-TIM-30, GF-TIM-40, and GF-TIM-55, respectively. Corresponding array densities are 0.231, 0.374, 0.44, and 0.505 g cm⁻³ (Fig. 2e), with the maximum density attains 150% of that for previously reported GF-TIMs [30], and 400% of that for conventional field-oriented pitch-based carbon fiber TIMs [29,31]. Continuous and vertically aligned GFs can be clearly observed in GF-TIMs. The percentage of GFs oriented above 75° reached as high as 88.5%, 87.9%, 89.0%, and 80.4% in GF-TIM-20, GF-TIM-30, GF-TIM-40, and GF-TIM-55, respectively (Fig. 2f), with several exceeding those reported for electric-field-aligned GFs [30]. The right panels of Fig. 2a–d show that surface region covered by graphene sheets increases significantly with higher GF content, rising from 32.8% at 21.6 wt% to nearly 100% at 39.0 and 54.2 wt%. A complete surface coverage by graphene regions is achieved even at a GF content lower than 55 wt%. Such a phenomenon is attributed to the plastic deformation of GFs induced by diamond wire shearing during cutting, which significantly enlarges the effective heat-transfer region of TIM surface, and reduces the contact thermal resistance when it was employed in electronic devices.

3.2. Thermal performance of GF-TIMs

The through-plane thermal ($K_{\perp}$) of GF-TIM was measured using the laser flash method according to the equation [27]:

$$K = \alpha \times \rho \times C_p \quad (1)$$

where α is the thermal diffusivity, ρ is the density, and C_p is the specific heat capacity. The corresponding results are shown in Fig. 3a. Benefiting from highly aligned, continuous, and densified GF arrays which provide effective pathways for phonon conduction, the GF-TIM exhibits outstanding through-plane thermal conductivity. The α of GF-TIM increased with GF content, from 80.36 mm² s⁻¹ at 21.6 wt% to 199.24 mm² s⁻¹ at 54.2 wt%, corresponding $K_{\perp}$ ranged from 126.34 to 190.22 W m⁻¹ K⁻¹ (Fig. 3b). It is 1.54–2.35 times that of indium foil (81.8 W m⁻¹ K⁻¹) and 1.53–2.31 times that of the previously reported GF-TIM [30]. The $K_{\perp}$ of GF-TIM peaks at 39.0 wt% GF content, but decreases slightly as the content further increases to 54.2 wt%. This decline occurs because the linear increase in thermal diffusivity is offset by reductions in both density and specific heat capacity at higher filler loadings. Considering that TIMs are usually subjected to temperature fluctuations under operational conditions, the through-plane thermal conductivity of GF-TIM was further investigated at various temperatures (Fig. 3c). For example, the $K_{\perp}$ of GF-TIM-40 decreased by only 9.25% (to 172.63 W m⁻¹ K⁻¹) and 11.42% (to 168.50 W m⁻¹ K⁻¹) when the temperature was raised from 25 °C to 80 and 100 °C, respectively. This slight decline is attributed to the increased Umklapp phonon scattering in crystalline materials at higher temperatures [32,33]. Nevertheless, GF-TIM-40 retains metal-level thermal conductivity even at 100 °C. Moreover, the fluctuation of $K_{\perp}$ is less than 2.30% after 20 heat loading cycles from 25 to 100 °C (Fig. 3d), demonstrating excellent thermal stability of the GF-TIM-40. Compared with previously reported elastic TIMs, the GF-TIM-40 prepared in this work shows a significant advantage in through-plane thermal conductivity (Fig. 3e and Table S3) [19,

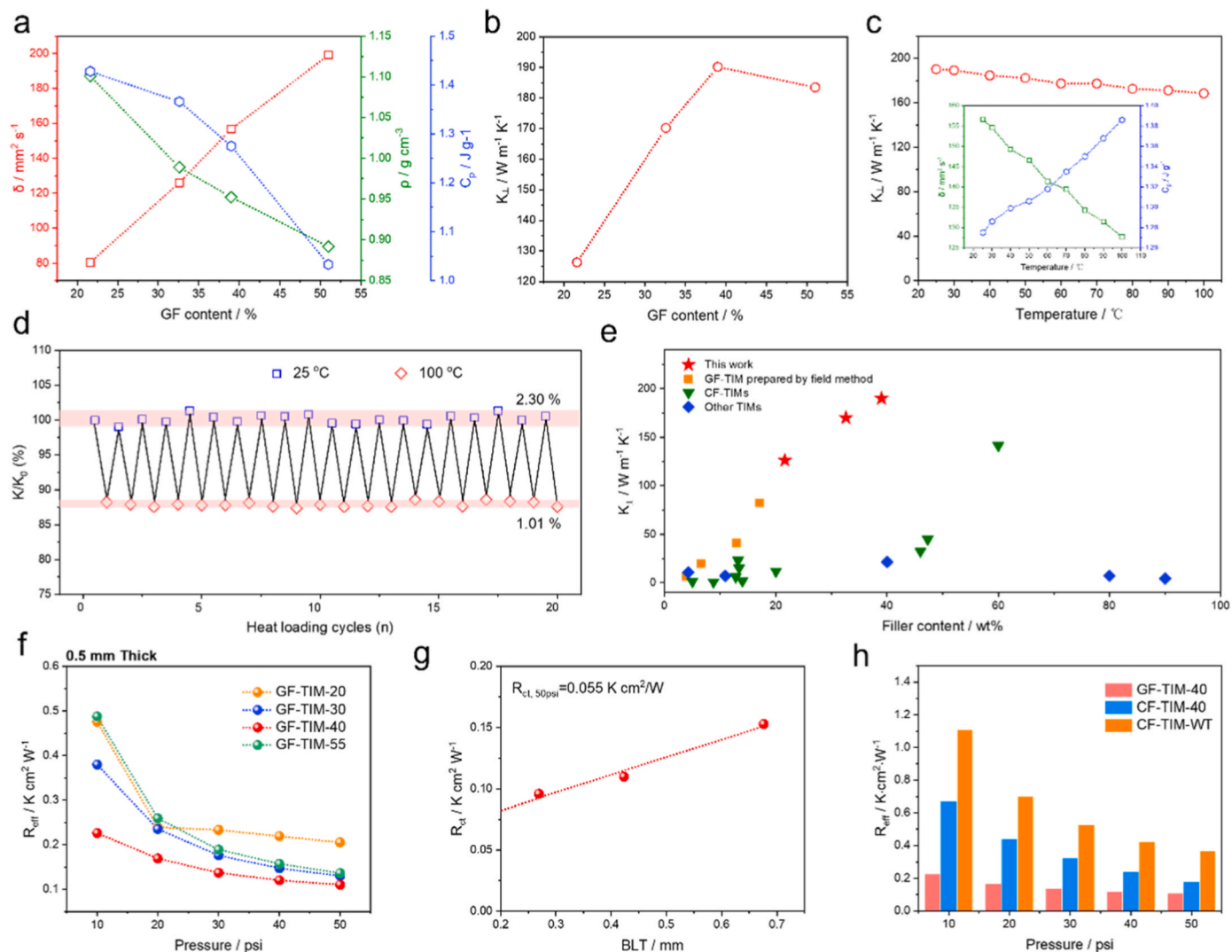


Fig. 3. Thermal-performance of GF-TIM-40. (a) Thermal diffusivity (α), specific heat capacity (C_p), volume density (ρ) and (b) through-plane thermal conductivity ($K_{\perp}$) of GF-TIMs with different filler contents. (c) The effect of temperature on the $K_{\perp}$ of GF-TIM-40. The inset shows the variation of α and C_p of GF-TIM-40 with temperature (25–100 °C). (d) The $K_{\perp}$ retention of GF-TIM-40 under cyclic heating loading from 25 to 100 °C. (e) Comparison of the $K_{\perp}$ versus filler content of GF-TIM-40 and previously reported elastic TIMs. (f) Effective thermal resistance (R_{eff}) of GF-TIM-20, -30, -40, and -55 (0.5 mm thick, 50 psi), respectively. (g) Contact thermal resistance (R_{ct}) of GF-TIM-40 under 50 psi. (h) Effective thermal resistance (R_{eff}) of GF-TIM-40, CF-TIM-40, and CF-TIM-WT under 10–50 pressure.

22,23,30,31,34–45].

In addition to $K_{\perp}$, The thermal conductive performance of a TIM is quantitatively evaluated by its effective thermal resistance (R_{eff}), which is the sum of material impedance (R_{TIM}) and contact thermal resistance (R_c) [15], described as follows:

$$R_{eff} = R_{TIM} + R_c = \frac{BLT}{K_{\perp}} + R_c \quad (2)$$

The R_{eff} of 0.5 mm thick GF-TIMs with varying filler contents was measured using a thermal resistance tester. As a result, the GF-TIM-40 showed the lowest R_{eff} of 0.11 K cm² W⁻¹ and highest effective through-plane thermal conductivity (K_{eff}) of 38.5 W m⁻¹ K⁻¹ under 50 psi (Fig. 3f). According to Equation (2) and previous discussion, the GF-TIM-40 shows the highest $K_{\perp}$, and consequently the lowest R_{TIM} , outperforming GF-TIM-20 and GF-TIM-30. However, further increasing the GF content to 54.2 wt% leads to a decrease in $K_{\perp}$ and a rise in R_{TIM} . Concurrently, the increased compression modulus and worsened thixotropy of the GF-TIM-55 with higher GF content may reduce the effective contact area at interfaces, thus increasing the R_c . The increase in R_{eff} for GF-TIM-55 may be attributed to the concurrent rise in both R_{TIM} and R_c . Consequently, the GF-TIM-40 demonstrates the optimal interfacial

heat dissipation performance. The K_{eff} of GF-TIM with different filler contents at 10–50 psi is further compared in Fig. S2.

According to thermal resistance of GF-TIM-40 with different BLT, the R_c under 50 psi was calculated to be 0.055 K cm² W⁻¹ (Fig. 3g). Such a low R_c is attributed to the wire-cutting process, during which the graphene coverage is increased by the shear action of diamond wire as previously discussed, while the surface is simultaneously polished to improve flatness, thereby expanding the effective contact area of GF-TIM-40. As a reference, GF-TIM-20 with only 32.8% graphene surface coverage exhibited an R_c of 0.1083 K cm² W⁻¹ at 50 psi (Fig. S3). Based on the Cooper-Mikic-Yovanovich model [46], the contact thermal resistance is proportional to hardness and surface roughness, while inversely proportional to the packaging pressure. Consequently, further reductions in contact thermal resistance may be achieved in the future by decreasing the surface roughness through polishing, or decreasing hardness via employing GFs and silicone matrix with lower modulus. Moreover, the R_{eff} of a 0.5 mm thick, ultrasonically cut GF-TIM_U-40 with a surface comprising isolated graphene and silicone regions (Fig. S4a–c) was also measured for further comparison. The resultant R_{eff} of GF-TIM_U-40 was displayed in Fig. S4d, that was 1.4 times higher than GF-TIM-40 at 50 psi. Therefore, the GF-TIM-40 fabricated by the

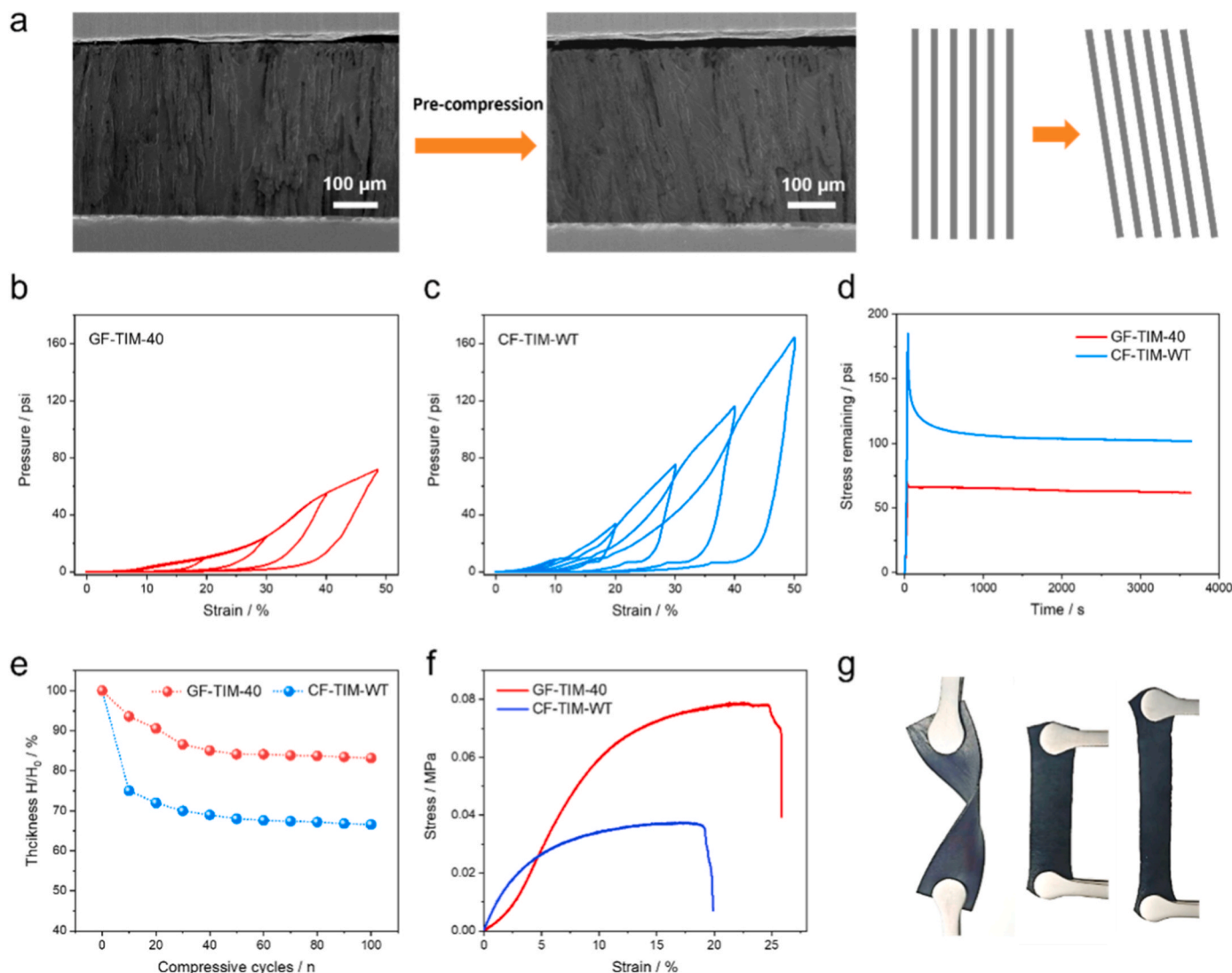


Fig. 4. Mechanical properties of the GF-TIM-40. (a) Illustration of compression mode changed by a pre-compression process of 20% strain after spring back. (b) The compressive stress-strain curve of the GF-TIM-40 and (c) CF-TIM-WT. (d) Stress relaxation behaviors of the GF-TIM-40 and CF-TIM-WT at 50 % strain. (e) Thickness evolution of the GF-TIM-40 and CF-TIM-WT after compressive cycles under 50 % strain. (f) Comparison of the tensile properties between GF-TIM-40 and CF-TIM-WT. (g) Photographs of GF-TIM-40 under different tensile and twisted states.

stacking-cutting strategy using AFGMs combines high intrinsic thermal conductivity with low contact resistance, demonstrating excellent interfacial heat dissipation performance.

To further verify the thermal performance of GF-TIM-40, R_{eff} under 10–50 psi was compared between a commercial pitch-based carbon-fiber TIM (CF-TIM-WT, WearnTIMo, $25 \text{ W m}^{-1} \text{ K}^{-1}$) and a 39.2 wt% pitch-based carbon-fiber TIM (CF-TIM-40) prepared using the same stacking-cutting strategy, both with a thickness of 0.5 mm (Fig. 3h). The results indicate that GF-TIM-40 exhibits the lowest R_{eff} over the pressure range of 10–50 psi. The CF-TIM-WT shows a R_{eff} 3.36 to 4.91 times higher, while the CF-TIM-40 with the same filler content presents a R_{eff} 1.65 to 2.98 times higher than that of GF-TIM-40. Additionally, the effective thermal conductivity (K_{eff}) of GF-TIM-40 is also compared with commercial and previously reported materials in Fig. S5, revealing a significant advantage for the former [11,47–51].

3.3. Mechanical properties of GF-TIM

In practical assembly, both thermal conductive performance and compressibility are required for the TIM to accommodate varying component heights and absorb tolerances in electronic devices [52]. The

packaging pressure for TIMs in electronics is usually limited to 25–200 psi to avoid component damage [53]. Pre-compression of 20% strain was applied to introduce a slight inclination to the GF array, which altered its compression mode from complete vertical collapse to inclined buckling (Fig. 4a) and consequently reduced the compressive modulus without compromising its high through-plane conductivity. Fig. S6 shows the $K_{\perp}$ and thickness retention of GF-TIM-40 after pre-compression. As shown in Fig. 4b and c, GF-TIM-40 exhibited compressive stresses of 71.6 psi under 50% compressive strain, which are only 43.6% of those for the CF-TIM-WT (164.3 psi), thus well meeting the requirements for TIM encapsulation. The good compressibility of GF-TIM-40 is also ascribed to the high flexibility and low modulus of GFs, which have a modulus approximately 65% to 86% lower than that of pitch-based CFs [28–30].

To further evaluate the structural stability, the residual stress of GF-TIM-40 and CF-TIM-WT was measured under compression. After stress relaxation for 3600 s, the GF-TIM-40 and CF-TIM-WT held 87.0% and 54.9% residual stress at 50% strain, respectively (Fig. 4d). Moreover, after 100 cycles of compression-rebound tests under high strains of 50%, the thickness retentions of GF-TIM-40 and CF-TIM-WT were measured to be 83.2% and 66.6% (Fig. 4e). To elucidate the different compressive

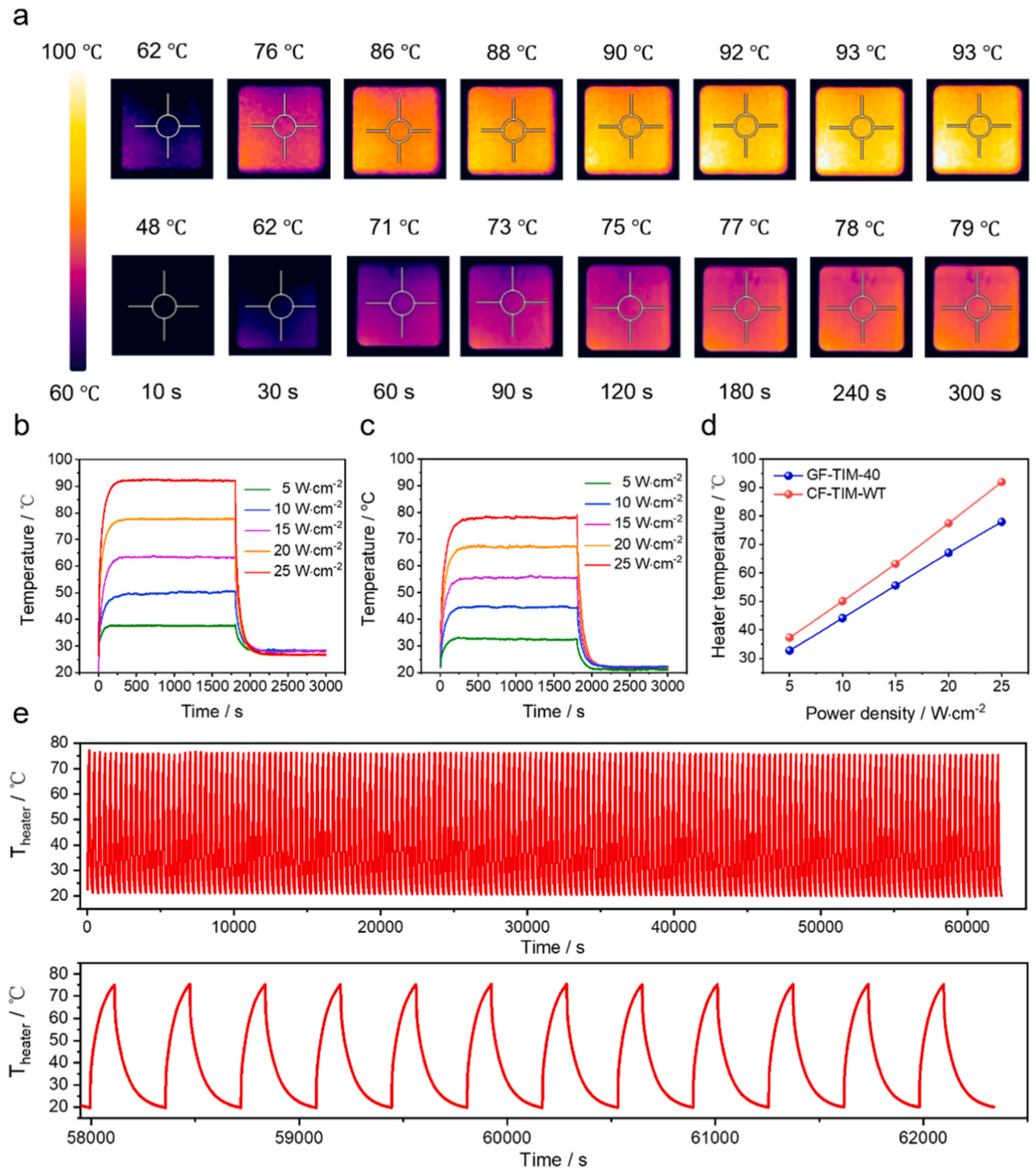


Fig. 5. Practical performance of GF-TIM-40 for cooling electronic devices. (a) Infrared thermal images of ceramic heaters thermally managed by GF-TIM-40 versus CF-TIM-WT under a heating power of 25 W cm^{-2} . (b) Temperature-time curves of ceramic heaters thermally managed by CF-TIM-WT and (c) GF-TIM-40 under a heating power range of $5\text{--}25 \text{ W cm}^{-2}$. (d) Steady-state temperature of ceramic heaters thermally managed by GF-TIM-40 and CF-TIM-WT with increasing input power. (e) Temperature evolution of the ceramic heater in cyclic heat loading tests.

behaviors of GF-TIM and CF-TIM, in-situ microscopic observations during compression were performed. Under compressive loading, CF-TIM exhibited fiber fracture, interfacial slip, debonding, and the generation of irreversible voids (Fig. S7a). These structural defects cannot recover upon unloading, resulting in significant permanent deformation (Fig. S7b). This failure mode is primarily attributed to the high modulus, smooth surface, and chemical inertness of CFs, which result in weak interfacial adhesion with the polymer matrix. In contrast, GFs exhibit a fundamentally different deformation mechanism. The abundant surface wrinkles on GFs facilitate robust physical entanglement and mechanical interlocking with the matrix. Owing to their lower modulus and superior flexibility, GFs undergo cooperative tilting and bending deformation in concert with the elastic polymer matrix during compression, rather than sliding or fracturing (Fig. S7c). This structural synergy preserves the stability of TIMs, promoting near-complete elastic recovery (Fig. S7d) to the original state and thereby minimizing permanent deformation. Furthermore, the cross-sectional diagrams of GF-TIMs before and after cyclic compression are shown in Fig. S8. We analyzed the GFs orientation distribution before and after 100 compression cycles at high strain of 30% and 50% in Fig. S9a to validate the structural stability of the GF-TIM. The orientation angle remained nearly unchanged after 100 compression cycles compared to the original state. After 100 cycles of 50% strain compression, the orientation degree of the GF array only decreased slightly from 0.867 to 0.794 (Fig. S9b and c). After 100 cycles of 30%-strain and 50%-strain compression, the resilience reached as high as 97.7% and 83.2%, respectively. These results illustrate that GF-TIM-40 exhibits not only ease of compression but also excellent elasticity and structural stability. Thus, it can effectively cushion impact, protect components, and maintain performance under vibrational or shock conditions.

Another important mechanical property for TIMs during operation is the in-plane tensile strength in addition to compression. For GF-TIM-40, the in-plane tensile fracture strength was measured as 0.0783 MPa with an elongation at break of 24.68%, which are 2.13 and 1.31 times those of CF-TIM-WT, respectively (Fig. 4f). The better tensile properties of GF-TIM-40 are primarily attributed to the micro-wrinkled surface of graphene fibers. And the interfacial contact area between fillers and polymer matrix is significantly larger than CFs with the smooth surface, leading to enhanced interactions and strength. Fig. 4g displays GF-TIMs under different tensile and torsional states, illustrating its favorable operability.

3.4. Practical application of GF-TIM in thermal management

To elucidate the practical thermal management performance of GF-TIM-40 in electronic components, a setup as shown in Fig. S10 was constructed to simulate the heat dissipation process of a central processing unit (CPU). The TIM was placed on an air-cooled aluminum plate, with a 2 cm × 3 cm ceramic plate positioned above it as a heater. A packaging pressure of 0.1 MPa was applied, and temperature was monitored through an observation window on the top plate. Infrared thermal images shown in Fig. 5a indicate that the steady-state temperature of ceramic heater is 93 °C at a heating power of 25 W cm⁻² when a 0.5 mm thick CF-TIM-WT is used. Compared with the cases employing a 0.5 mm thick GF-TIM-40, the ceramic heater temperature has dropped 14 °C at the same condition. Based on the steady heater temperatures of ceramic heater versus varies power densities (Fig. 5b and c), the equivalent heat transfer coefficient (equal to the reciprocal of the slope in Fig. 5d) for the GF-TIM-40 and CF-TIM-WT is calculated, respectively. As a result, the GF-TIM-40 exhibited an equivalent heat transfer coefficient that was 1.21 times of the CF-TIM-W. Furthermore, temperature fluctuations of the ceramic heater during a temperature cycling test (heating for 120 s at 25 W cm⁻² and cooling for 240 s) is only 2.5% (Fig. 5e), demonstrating the excellent thermal shock stability of GF-TIM-40 in practical applications.

While state-of-the-art thermal interface materials (TIMs), including

liquid metals, rigid carbon/graphite pads, and phase-change materials have advanced specific performance metrics, they inherently struggle to decouple the critical trade-offs between high thermal conductivity, mechanical compliance, and long-term reliability [54–57]. By leveraging the unique combination of excellent thermal conductivity and low modulus inherent to graphene fibers, our GF-TIMs successfully bridge these gaps. This new material system and preparation strategy offer a promising direction for the development of novel TIMs with well-rounded comprehensive performance.

4. Conclusions

In summary, a stacking-cutting strategy using aligned graphene fiber mats was proposed, through which the GF-TIM-40 was fabricated from graphene fibers, featuring a thermally conductive skeleton with high continuity, vertical orientation (>0.89), and array density (0.44 g cm⁻³). Consequently, a through-plane thermal conductivity of 190.22 W m⁻¹ K⁻¹ along with a contact thermal resistance of 0.55 K cm² W⁻¹ (under 50 psi) were achieved. As for mechanical properties, excellent compressibility (50% strain at 71.61 psi), structural stability (87.0% residual stress at 50% strain after relaxation for 3600 s), and resilience (H/H₀>80% at 50% strain) were still exhibited by the material. Concurrently, a tensile fracture strength of 0.0738 MPa and a corresponding fracture elongation of 24.68% were measured, suggesting its excellent operability. It also demonstrated excellent heat dissipation capability and thermal stability under thermal shocks in practical applications. This work paves a new way for developing GF-TIMs that exhibit outstanding thermal conductivity, mechanical compliance, and ease of handling.

CRediT authorship contribution statement

Jiahao Lu: Writing – original draft, Formal analysis, Data curation, Conceptualization. **Bo Zhao:** Investigation, Data curation. **Manman Li:** Formal analysis, Data curation. **Shengying Cai:** Writing – review & editing, Writing – original draft, Methodology, Funding acquisition, Formal analysis, Conceptualization. **Chenghua Cui:** Investigation, Data curation. **Ying Zhang:** Investigation. **Yelong Tong:** Resources. **Tongliang Wang:** Software. **Jie Li:** Formal analysis. **Kai Pang:** Methodology. **Peng Li:** Investigation, Formal analysis. **Zhen Xu:** Methodology. **Zhijian Wang:** Writing – original draft, Methodology. **Wenjun Li:** Resources. **Yingjun Liu:** Project administration, Funding acquisition, Conceptualization. **Chao Gao:** Validation, Supervision, Resources, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.carbon.2026.121466>.

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