

Extremely anisotropic thermally conductive MXene/CNT hybrid film for battery thermal management

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Nano Res., **Just Accepted Manuscript** • <https://doi.org/10.26599/NR.2026.94909210>

<https://www.sciopen.com/journal/1998-0124> on Sep. 22, 2026

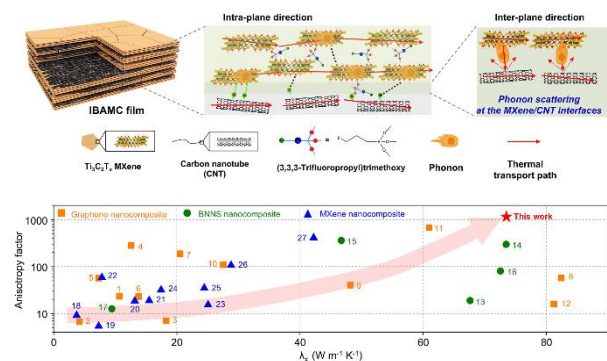
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MXene/CNT films constructed via alternating layer-by-layer blade coating with interfacial covalent bridging achieve a record-high thermal anisotropy factor of 1220 by suppressing inter-plane heat transfer through abundant interfaces while maintaining high intra-plane thermal conductivity via a highly aligned layered structure, thereby demonstrating excellent battery thermal management performance.

Extremely anisotropic thermally conductive MXene/CNT hybrid film for battery thermal management

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Received: 25 May 2026; **Revised:** 20 September 2026; **Accepted:** 22 September 2026

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 **Cite this article:** Nano Research, 2026, 19, 94909210 <https://doi.org/10.26599/NR.2026.94909210>

ABSTRACT: The higher power density of energy storage systems necessitates advanced thermal management strategies and materials, especially for the anisotropic thermally conductive films (TCFs). However, it remains great challenging for assembling high-performance anisotropic TCFs. Here, we demonstrate an extremely anisotropic TCFs with carbon nanotubes (CNT) and MXene nanosheets through alternating layer-by-layer blade coating technique with interfacial covalent bridging. The resultant films exhibit a high in-plane thermal conductivity of $71.9 \pm 1.3 \text{ W m}^{-1} \text{ K}^{-1}$ and an extremely low inter-plane thermal conductivity of $0.06 \pm 0.01 \text{ W m}^{-1} \text{ K}^{-1}$, yielding a record-high anisotropy factor of 1220 ± 24 . This is enabled by the substantial interfaces between MXene nanosheets and CNT that impede the inter-plane thermal transport while the highly aligned layered structure maintains high intra-plane thermal conductivity. And the resultant films show strong mechanical properties and efficient thermal management performance for battery. Our work offers a new pathway for assembling high-performance TCFs used in the battery thermal management.

KEYWORDS: extremely anisotropic, thermally conductive, MXene film, battery thermal management

1 Introduction

Due to the increasing power density of energy storage systems, highly efficient thermal management strategies and materials are essential to address the problem of excessive heat accumulation [1-3]. Recent innovations have led to the emergence of anisotropic thermally conductive films (TCFs) that can disperse heat in the in-plane direction while insulating against heat in the inter-plane direction [4]. The thermal anisotropy factor, defined as the ratio of in-plane (λ_x) to inter-plane (λ_z) thermal conductivities, is often used to characterize the anisotropic thermal properties of TCFs [5, 6]. Two-dimensional (2D) nanomaterials, such as graphene [7], boron nitride (BN) [8], and black phosphorus (BP) [9], have become ideal candidates for fabricating anisotropic TCFs due to their inherent lamellar anisotropic performance. Numerous assembly approaches have been reported, including layer-by-layer assembly [10, 11], vacuum filtration [12, 13], and blade coating [14]. For instance, the layer-by-layer (LbL) assembled graphene/poly(vinyl alcohol) (PVA)

nanocomposite film exhibits λ_x and λ_z values of 13.8 and 0.6 $\text{W m}^{-1} \text{ K}^{-1}$, respectively, yielding an anisotropy factor of 23 [11]. The functional BN nanosheet (BNNS)/BP/cellulose nanofiber (CNF) nanocomposite film produced by vacuum filtration shows λ_x and λ_z values of 72.5 and 0.9 $\text{W m}^{-1} \text{ K}^{-1}$, respectively, leading to a high anisotropy factor of 81 [13]. However, the reported anisotropic TCFs demonstrate subpar thermal performance, exhibiting high λ_z values and small anisotropy factors that limit their practical application.

Titanium carbide (TiC) MXene nanosheets, a novel 2D material with abundant termination groups [15-20], exhibit tremendous potential for thermal management due to their theoretically high thermal conductivity of 108 $\text{W m}^{-1} \text{ K}^{-1}$ [21]. For example, the MXene/liquid metal (LM) nanocomposite film, fabricated through sol-gel film conversion, demonstrates a λ_x of 13.2 $\text{W m}^{-1} \text{ K}^{-1}$, coupled with a low λ_z of 0.71 $\text{W m}^{-1} \text{ K}^{-1}$, yielding a high anisotropy factor of 19 [22]. However, the anisotropic thermal conductivity of the resulting MXene film is significantly lower than the theoretical values. To design

MXene films with ultrahigh anisotropic thermal conductivities, a key feature is aligning MXene nanosheets along the intra-plane direction to create an efficient horizontal thermal transport channel for phonons and maximize the λ_x , while substantially reducing the λ_z along the inter-plane direction [23]. Carbon nanotubes (CNTs) have been widely regarded as ideal fillers for fabricating anisotropic TCFs due to their unique nanostructure, as well as their ultrahigh thermal conductivity ($1950\text{--}5000\text{ W m}^{-1}\text{ K}^{-1}$) and strength in the axial direction [24, 25]. The well-aligned CNTs in the film enhance the degree of orientation along the intra-plane direction, resulting in outstanding anisotropic thermal phonon transport properties [26].

Here, we demonstrate an ultrahigh anisotropic thermally conductive MXene-CNT film produced using an alternating LbL blade coating process with interfacial covalent bridging. The resulting interfacially bridged alternating MXene-CNT (IBAMC) film demonstrates an anisotropy factor as high as 1220 ± 24 , significantly surpassing previous nanocomposite films. This outstanding performance stems from the highly oriented MXene and CNT within the layered structure, created by the shear force during the blade coating process. This optimizes intra-plane thermal transport while minimizing inter-plane thermal conductivity through periodic interface phonon scattering. The IBAMC films exhibit remarkable mechanical properties, with a tensile strength of 302.2 MPa. Furthermore, potential applications for cooling energy storage devices highlight the excellent thermal management capabilities of the IBAMC film.

2 Experimental section

2.1 Materials

Hydrochloric acid (HCl, 36-38%) was obtained from Sinopharm Chemical Reagents Co., Ltd. Lithium fluoride (LiF, $\geq 99.99\%$) was sourced from Aladdin. Ti_3AlC_2 powder was acquired from Jilin 11 Technology Co., Ltd. The carboxylic carbon nanotubes (CNTs) were obtained from Nanjing XFANO Materials Tech Co., Ltd. (3,3,3-Trifluoropropyl)trimethoxy silane was acquired from Adamas. All materials were utilized without further purification.

2.2 Characterization

Atomic force microscopy (AFM) images of MXene and CNT nanosheets were acquired using a Bruker Multimode 8 in tapping mode. The microstructure and morphology of MXene, alternating MXene-CNT (AMC), blending MXene-CNT (BMC), and IBAMC films were characterized using a Hitachi SU8010 scanning electron microscope (SEM) under an accelerating voltage of 10 kV and a current of 10 μA . Transmission electron microscopy (TEM) was employed to examine the microscopic morphology of CNTs using an FEI Titan Themis G3 microscope at an accelerating voltage of 200 kV. X-ray diffraction (XRD) profiles were collected on a Shimadzu XRD-6000 with $\text{Cu-K}\alpha$ radiation, a scanning speed of 5 min^{-1} , and a scanning range of 5° to 70° . Attenuated total reflection (ATR) Fourier transform infrared (FTIR) spectra were recorded using a Thermo Nicolet NEXUS-470 FTIR instrument. X-ray photoelectron spectroscopy (XPS)

spectra were obtained on an ESCALABXI+ (Thermo Scientific) system equipped with a monochromatic $\text{Al-K}\alpha$ X-ray source. Cross-sectional samples of the IBAMC films were prepared by focused ion beam (FIB) milling using an FEI Helios NanoLab 600i, operating at an acceleration voltage of 30 kV with a current ramp from 230 pA to 40 pA.

3 Results and discussion

3.1 Preparation and characterization of the IBAMC film

The exfoliated MXene nanosheets have an average lateral size of 11.5 μm (Figure S1). Atomic force microscope (AFM) images reveal that the thickness of the monolayer MXene nanosheet is approximately 1.5 nm (Figure S2). The X-ray diffraction (XRD) curve (Figure S3) shows that the 104 peak of the Ti_3AlC_2 MAX phase near 39° is absent, indicating the successful exfoliation of the MXene nanosheets [27, 28]. Additionally, the d -spacings of the MXene film (1.20 nm) exceed those of Ti_3AlC_2 MAX (0.93 nm) due to the introduction of surface functional groups [29]. Transmission electron microscopy (TEM) and AFM images of carboxylic CNT reveal a clean morphology with no damage (Figure S4). Figure 1a illustrates the fabrication process of IBAMC films. The MXene sol was initially blade coating onto a polyvinylidene fluoride (PVDF) substrate moving at 50 cm min^{-1} . Subsequently, the CNT dispersion was blade coating onto the dried MXene layer. The AMC films were produced through repeated blade coating cycles. Five types of AMC films (AMC-I to AMC-V) were created by varying the relative concentration of the CNT dispersion. The optimal AMC film is AMC-III, which has MXene and CNT concentrations of 20 and 2 mg ml^{-1} , respectively, and was further processed into IBAMC films. The AMC-III film was immersed in a silane coupling agent, (3,3,3-Trifluoropropyl)trimethoxy silane, and rinsed with ethanol solution.

Five variations of IBAMC films (IBAMC-I to IBAMC-V) were developed by modifying the relative concentrations of the silane coupling agent solution. An IBAMC film (5 $\text{cm} \times 20\text{ cm}$; Figure 1b) was created by drying. For comparison, a BMC film was produced using alternating LbL blade coating with a MXene/CNT sol mixture. The actual compositions of MXene, CNT, and (3,3,3-Trifluoropropyl)trimethoxy silane in the AMC and IBAMC films were analyzed through thermogravimetric analysis (TGA; Figure S5 and Table S1). The cross-sectional SEM images of MXene, BMC, AMC, and IBAMC films are shown in Figure 1c and Figures S6 and S7. The alternating structure of MXene and CNT layers is observed in the IBAMC film (Figure 1c). Additionally, the high-resolution TEM images of the IBAMC film (Figure 1d) further illustrate the alternating layered structure of MXene and CNT. The interlayer spacing of CNT (0.34 nm) corresponds to the interplanar distance of (002) planes of graphite [30].

The IBAMC film demonstrates exceptional anisotropic thermal conductivity, featuring a high λ_x of 71.9 and a low λ_z of $0.06\text{ W m}^{-1}\text{ K}^{-1}$, respectively. Consequently, the anisotropy factor of the IBAMC film reaches 1220, exceeding all anisotropic TCFs based on 2D nanomaterials like graphene, BNNS, and MXene (Figure 1e and Table S2). The anisotropic thermal performance of the IBAMC film was demonstrated

by cooling the commercial 18650 lithium-ion battery (1200 mAh capacity). The vertically lithium-ion battery, wrapped with MXene and IBAMC films with thermal silicon grease, was attached to a heat sink (Figure S8). The temperature evolution of the battery was recorded during the charging/discharging process through infrared camera. When the discharge rate was set 3 capacity (3C) [31], the maximum temperatures of the bare battery and the battery wrapped with IBAMC films are 48.4°C and 40.8 °C, respectively, yielding a temperature reduction of 7.6°C. The cooling efficiency (η) is used to quantified assessed thermal management performance, defined as the percentage ratio of the temperature reduction to the bare battery's maximum temperature. Accordingly, the IBAMC film achieved an η of 15.7%. To highlight the superior thermal management

capability of the IBAMC film, traditional phase change materials (PCMs) are used for comparison. The IBAMC film achieved comparable heat dissipation to PCMs at the discharge rate of 3C for 18650 lithium-ion battery (Table S3). Notably, the thickness of IBAMC film (~ 0.014 mm) is significantly lower than that of typical PCMs (> 1 mm). We further emphasize the outstanding thermal management performance of IBAMC film through the specific cooling efficiency, which is defined as the η divided by the thickness of the composite materials. As shown in Figure S9, the specific cooling efficiency of IBAMC film reaches to the 1121 % mm⁻¹, which is substantially higher than that of PCMs. This enables significant weight reduction and densification in energy storage devices especially for high energy density battery packs.

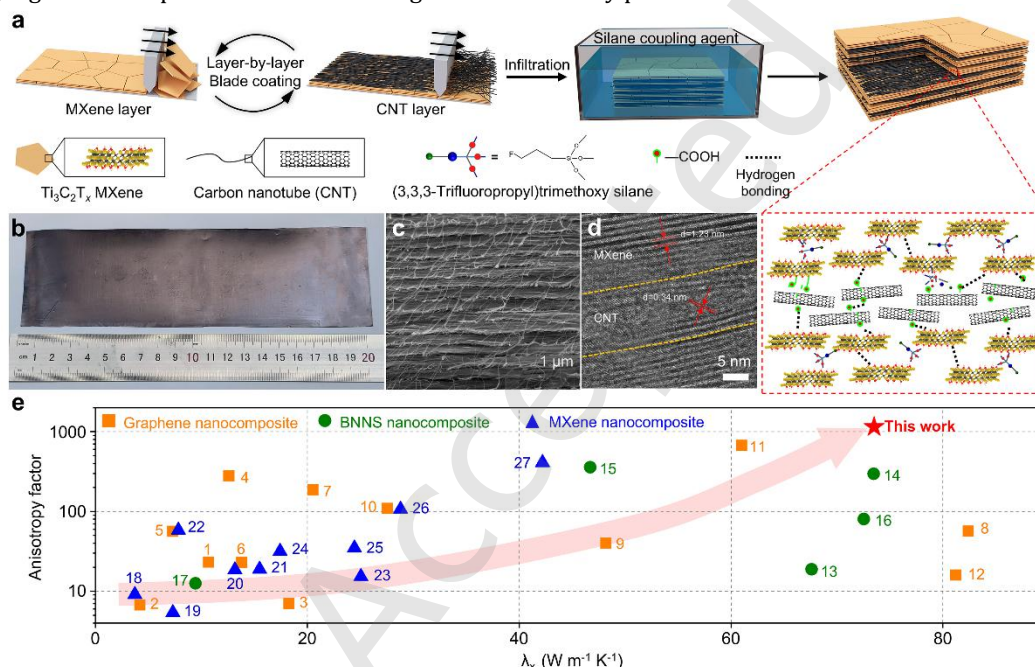


Figure 1 Fabrication process, structural model, and anisotropic thermal performance of the IBAMC film. (a) Fabrication process of the IBAMC film. (b) Photograph of a scalable IBAMC film with an area of 100 cm². (c) Cross-sectional SEM images of the IBAMC film, showing the alternating multilayered distribution of the Ti and Si elements. (d) HR-TEM images of the IBAMC film cross-section. (e) Comparison of λ_x and the anisotropy factor of the IBAMC film with those of the other two-dimensional anisotropic thermally conductive films.

3.2 Structural characterization of the IBAMC film

The XRD results of the MXene, AMC, BMC, and IBAMC films are shown in Figure S10 and Table S4. The d -spacing of the AMC-III film (1.22 nm) is larger than that of the MXene film (1.2 nm) due to the introduction of CNT. Additionally, the d -spacing of the IBAMC-II film (1.23 nm) increases after introducing (3,3,3-Trifluoropropyl)trimethoxy silane. Furthermore, the alignment of the MXene, BMC, AMC, and IBAMC films is investigated using Herman's orientation factor (Figure 2a to d and Figure S11). The Herman's orientation factor of the BMC film (0.888) is lower than that of the MXene film (0.968), indicating that the intercalated CNT disrupts the stacking order of MXene nanosheets [32]. However, the AMC film shows a relatively high Herman's orientation factor of 0.963 compared to the BMC film, demonstrating that the alternating LbL blade coating process almost does not affect the alignment of MXene nanosheets. Furthermore, the IBAMC film exhibits a higher degree of alignment with Herman's orientation factor of 0.976, demonstrating that the interfacial covalent bridging

process improves the stacking order of MXene nanosheets.

Compared to the MXene film, the X-ray photoelectron spectroscopy (XPS) spectra of the IBAMC film exhibit a new Si 2p peak, confirming the modification of (3,3,3-Trifluoropropyl)trimethoxy silane (Figure S12). The IBAMC film presents two new characteristic peaks at 460 eV and 456.5 eV binding energies, respectively, corresponding to the Ti-O-Si and Ti-O-C covalent bonding (Figure 2e) [33-35]. Additionally, the new peak at 925 cm⁻¹ further verifies the formation of Ti-O-C bonding [34]. The -OH peak of the BMC, AMC, and IBAMC films shifts to 3460 cm⁻¹, 3439 cm⁻¹, and 3450 cm⁻¹ when compared to the MXene film [36], respectively, indicating the presence of hydrogen bonding between MXene nanosheets and carboxylic CNT (Figure 2f).

The MXene films exhibit large voids between MXene nanosheets and have a high bulk porosity of $11.3 \pm 0.3\%$ (Figure 2g). After introducing the CNT into the MXene interlayer, the bulk porosity of the BMC film is reduced to $10.6 \pm 1.0\%$. Furthermore, the IBAMC films demonstrate a dense and highly aligned structure with a low porosity of 3.6

$\pm 0.8\%$ due to the alternating LbL blade coating method, followed by hydrogen and covalent bonding. The cross-sectional SEM images of MXene, BMC, and IBAMC films treated with focused ion beam (FIB) confirm the changes in

bulk porosity (Figure S13). Moreover, HR-TEM images reveal that the CNT can fill the voids between the MXene nanosheets (Figure S14).

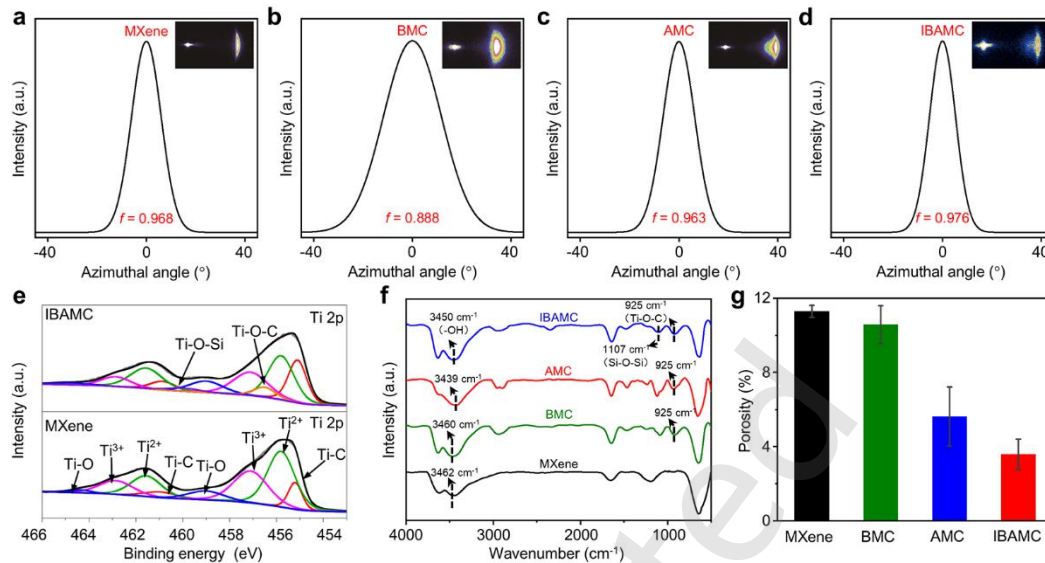


Figure 2 Structural characterization of IBAMC film. (a-d) Wide-angle X-ray scattering (WAXS) patterns for an incident Cu-K α X-ray beam parallel to the film plane, along with the corresponding azimuthal scan profiles for the 002 peak of MXene, BMC, AMC, and IBAMC films. (e) XPS spectra in the Ti 2p region for the MXene and IBAMC films. (f) Fourier-transform infrared spectroscopy (FTIR) spectra of MXene, BMC, AMC, and IBAMC films. (g) Porosities of MXene, BMC, AMC, and IBAMC films derived from density measurements.

3.3 Mechanical properties of the IBAMC film

The typical stress-strain curves for the MXene, BMC, AMC, and IBAMC films are shown in Figure 3a and Figure S15. The associated mechanical properties are outlined in Table S5. The IBAMC films demonstrate the highest mechanical

properties, featuring a tensile strength of 302.2 ± 19.6 MPa and a toughness of 3.5 ± 0.7 MJ m⁻³, which are 1.78 and 3.43 times greater than those of the MXene film; 2.07 and 1.20 times greater than those of the BMC film; and 1.12 and 1.25 times greater than those of the AMC film.

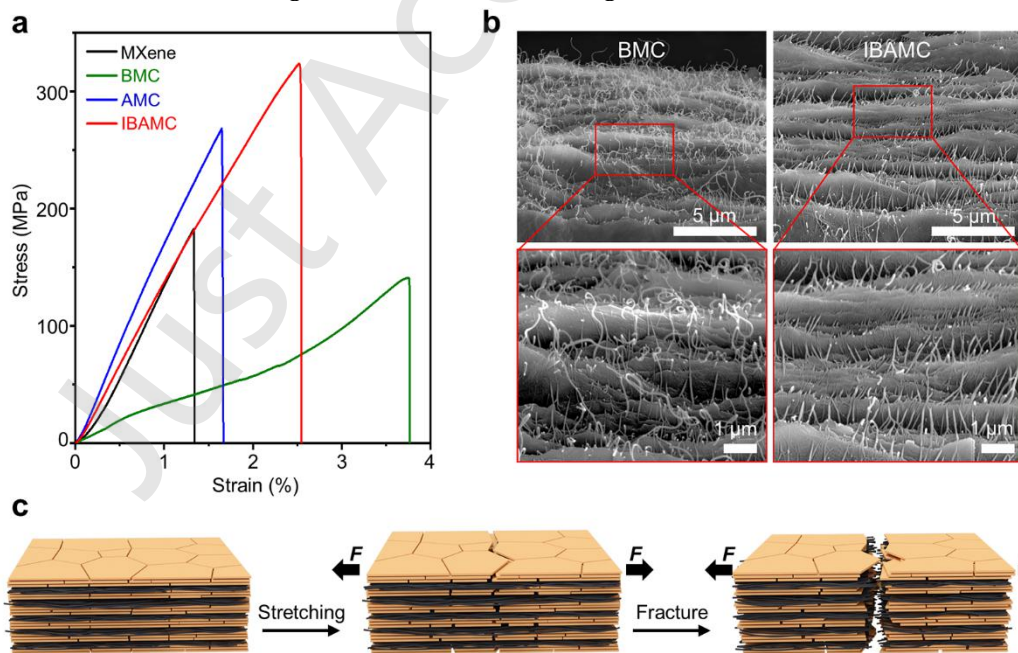


Figure 3 Mechanical properties and fracture mechanism of IBAMC film. (a) Stress-strain curves for the MXene, BMC, AMC, and IBAMC films. (b) Fracture morphology of the BMC and IBAMC films. (c) Proposed fracture mechanism of the IBAMC

The fracture morphologies of the MXene, BMC, AMC, and IBAMC films are presented in Figure 3b and Figure S14. The MXene films exhibit flat fracture edges when the MXene nanosheets are pulled out. For the BMC film, the MXene nanosheets and CNTs are extracted, revealing a close-packed lamellar morphology with hydrogen bonding and Ti-O-C

covalent bonding between the MXene nanosheets and CNTs, leading to typical “zigzag” step-type cracks, and the CNTs [32]. Additionally, the AMC film displays a close-packed lamellar structure and curled fracture edges due to the alternating MXene and CNT layers and enhanced interfacial interactions. In comparison to the AMC film, the IBAMC film

demonstrates the most close-packed and overall stepped lamellar structure with serrated and curled fracture edges, as the CNTs are neatly pulled out along the stress direction, indicating high stress-transfer efficiency between the MXene nanosheets and CNTs. The proposed fracture mechanism of the IBAMC film is illustrated in Figure 3c. Briefly, the adjacent MXene nanosheets first slide relative to one another during the stretching. Then, these adjacent MXene nanosheets slip further due to the hydrogen and covalent bonding of Ti-O-Si and Ti-O-C with the increasing tensile load, which dissipates external energy. Following further stretching, the CNTs gradually straighten along the tensile direction and are ultimately fully stretched out, facilitating greater energy dissipation and high stress-transfer efficiency. [37].

3.4 Anisotropic thermal performance of the IBAMC film

The anisotropic thermal performance of MXene, BMC, AMC, and IBAMC films is presented in Figures 4a and 4b, Figures S17 and S18, and Table S6 and S7, which include density, specific heat capacity, thermal diffusion coefficient, thermal conductivity, and anisotropy factor. The MXene film exhibits λ_x and λ_z values of $37.1 \text{ W m}^{-1} \text{ K}^{-1}$ and $0.12 \text{ W m}^{-1} \text{ K}^{-1}$, respectively, corresponding to an anisotropy factor of 302 (Figure 4a). The BMC film displays λ_x and λ_z values of $55.1 \text{ W m}^{-1} \text{ K}^{-1}$ and $0.14 \text{ W m}^{-1} \text{ K}^{-1}$, with an anisotropy factor of only 388. For comparison, as the concentration of CNT dispersion increases, the AMC films with alternating MXene and CNT layers show a gradual increase in λ_x , while λ_z initially decreases before increasing. The optimized CNT dispersion is 2 mg mL^{-1} , demonstrating λ_x , λ_z , and an anisotropy factor of $64.6 \text{ W m}^{-1} \text{ K}^{-1}$, $0.06 \text{ W m}^{-1} \text{ K}^{-1}$, and 1030, respectively. This phenomenon is due to the alternately aligned lamellar structure and the covalent bonding of Ti-O-C, which facilitates efficient thermal transport along the intra-plane direction while reducing inter-plane thermal transport [38]. Furthermore, the IBAMC film shows the highest λ_x of $71.9 \text{ W m}^{-1} \text{ K}^{-1}$, which is 1.94, 1.30, and 1.11 times higher than that of the MXene, BMC, and AMC films, respectively. This improvement is attributed to enhanced interfacial interactions from Ti-O-C and Ti-O-Si covalent bonding, as well as a high degree of alignment of MXene nanosheets along the intra-plane direction. The λ_z of the IBAMC film ($0.06 \text{ W m}^{-1} \text{ K}^{-1}$) is consistent with that of the AMC film. Thus, the anisotropy factor of the IBAMC film reaches a record high of 1220 (Figure 4b). Furthermore, unlike thermal conductive MXene-based films, the IBAMC film exhibits the highest λ_x across all filler content levels (Figure S19 and Table S8). The proposed mechanism for enhanced thermal conductivity anisotropy is illustrated in Figure 4c. The highly alignment degree alternately layered structure provides an efficient continuous thermal transport path along the intra-plane direction. Furthermore, the strong covalent intra-sheet bridging within the aligned layered structure effectively minimizes interfacial phonon scattering between adjacent MXene nanosheets, facilitating high-speed phonon transmission [39]. In contrast, the substantial interfaces between MXene layers and CNT layers along the inter-plane direction, which results in the abundant phonon scattering

and interfacial thermal resistance, minimizing thermal conductivity along the inter-plane direction. Moreover, the finite element simulation models of IBAMC film and MXene film was developed to elucidate the anisotropic thermal mechanism (Figure 4d). The detail parameters used in the simulation are shown in the characterization of Supplement materials. Briefly, the initial temperature of the model system was set as 25°C , and a continuous temperature load of 100°C (linear heat source) was applied to the top surface of the model, leading to a heat flow along the vertical direction. The IBAMC model shows the lower temperature of the bottom surface and causes a larger decrease of temperature (66.4°C) in the vertical direction compared to MXene model (53.8°C) after heating 4 nanoseconds (Figure 4d). Moreover, the IBAMC model shows the better thermal insulation along the vertical direction during the whole heating process (Figure S20). The reason is that the alternating multilayered nanostructured has substantial interfaces between MXene nanosheets and CNTs that can generate more phonon scattering along the inter-plane direction while the intra-plane direction heat dissipation is unaffected. In addition, the anisotropic thermal performance of the IBAMC film demonstrates exceptional thermal stability up to 100°C (Figure S21). This stability persists despite significant temperature-dependent shifts in the individual conductivities: the intra-plane conductivity increases from 71.9 to $123.5 \text{ W m}^{-1} \text{ K}^{-1}$, while the inter-plane conductivity rises from 0.06 to $0.10 \text{ W m}^{-1} \text{ K}^{-1}$ over the same temperature range.

The practical thermal management application of the IBAMC film was examined by cooling the commercial 18650 lithium-ion battery (1200 mAh capacity), and the schematic illustrating the use of the IBAMC films integrated into the 18650 lithium-ion battery as shown in Figure S8. The bare 18650 lithium-ion battery exhibits peak temperatures of 31.5°C and 48.4°C at charge and discharge rate of 2C and 3C, respectively. The 18650 lithium-ion battery wrapped with MXene film slightly reduces the peak temperatures to 31.1°C and 46.4°C , respectively. Then, the peak temperatures of 18650 lithium-ion battery wrapped with IBAMC film significantly decrease to 29.8°C and 40.8°C , which is 1.7°C and 7.6°C lower than that of the bare battery (Figure 4e), resulting in a cooling efficiency (η) of 15.7%. Furthermore, the specific cooling efficiency ($\eta/\text{thickness}$) was calculated for comparison with traditional thick PCMs for lithium-ion battery. The ultrathin IBAMC film demonstrates an exceptionally high specific η of $1121 \text{ \% mm}^{-1}$, significantly higher than that of reported PCMs (Figure S9 and Table S3). The outstanding thermal management performance of IBAMC film primarily stems the highly alignment arrangement of MXene nanosheets and CNTs within the IBAMC, which results in an ultrahigh λ_x values of $71.9 \text{ W m}^{-1} \text{ K}^{-1}$. This high λ_x value enables efficient and fast conduction heat generated by the battery along the intra-plane direction to the heat sink and further transfer to the environment. In comparison, PCMs typically exhibit the relative low TC ($< 10 \text{ W m}^{-1} \text{ K}^{-1}$) due to the high content level of ultralow TC polymers such as paraffin and polyethylene glycol and the disordered distribution of nanofillers including expand graphite, BN, and CNTs within the polymer matrix [40, 41].

Although the PCMs can absorb a large amount of latent heat during the phase transition, their low TC impedes the efficient dissipation of excess heat to the environment. Moreover, the IBAMC film shows the unique anisotropy thermal properties that are well-suited for lithium-ion battery. It provides excellent heat dissipation while simultaneously offering substantial thermal insulation in the inter-plane direction, thereby minimizing thermal influence

on adjacent lithium-ion batteries. Additionally, the IBAMC film demonstrates good cyclic stability, maintaining the most of initial cooling efficiency through three consecutive charge/discharge cycles (Figure 4f). Moreover, the IBAMC film exhibits slight oxidation under the 50°C environment 24 h. As shown in Figure S22, the peak area ratio of Ti-O bond of the IBAMC-50°C film (8.1%) is similar to the initial IBAMC film (7.1%).

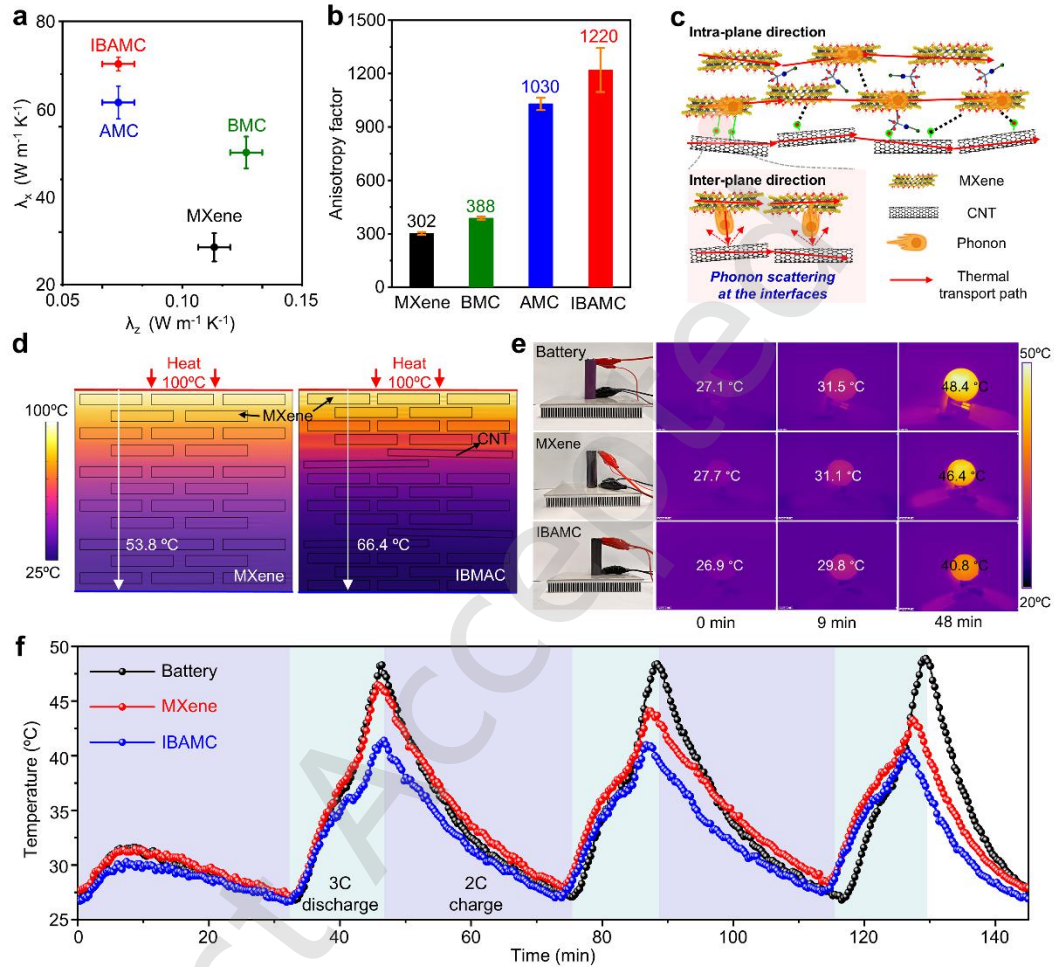


Figure 4 Anisotropic thermal performance and thermal management applications of IBAMC film. (a) λ_x and λ_z , and (b) the anisotropy factors of MXene, BMC, AMC, and IBAMC films. (c) Proposed anisotropic thermal mechanism for the IBAMC film. Finite element simulated temperature distribution through plane for (d) MXene and IBAMC model. (e) Infrared thermal images displaying the maximum charging and discharging temperatures of the bare battery, MXene, and IBAMC films. (f) Surface temperatures of the bare battery and those wrapped with MXene and IBAMC films during three charging and discharging cycles.

4 Conclusion

In summary, we demonstrated an ultrahigh anisotropic TCFs using MXene nanosheets and CNT through an alternate layer-by-layer blade coating with interfacial covalent bridging. The high degree of alignment of MXene nanosheets and the improved interfacial interactions between MXene and CNT in the TCFs maximize the intra-plane thermal conductivity while reducing the inter-layer thermal conductivity. The resulting TCFs demonstrate a record-high anisotropic factor, high tensile strength, and efficient thermal management for cooling energy storage devices. This is attributed to the substantial interfaces between MXene nanosheets and CNT that impede the inter-plane thermal transport while the highly aligned layered structure maintains high intra-plane thermal conductivity. Our work provides an avenue for assembling high-performance TCFs used in the next-generation electronic devices.

Electronic Supplementary Material: Supplementary material (please provide the brief detail of the ESM) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909210>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant Nos. 52550002, 52522303, 52373066), the National Science Fund for Distinguished Young Scholars (Grant No. 52125302), the National Key Research and Development Program of China (Grant No. 2021YFA0715700), the Open Research Fund of Suzhou Laboratory (Grant No. SZLAB-1108-2024-ZD002), the Suzhou Key Laboratory of Bioinspired Interfacial Science (Grant No. SZ2024004), the Postdoctoral Fellowship Program of CPSF (GZB20250931), and the China Postdoctoral Science Foundation (Grant No. 2025M784263) and the New Cornerstone Science Foundation through the XPLOER PRIZE. We would like to thank the Physical and Chemical Analysis Center at the Suzhou Institute for Advanced Research, University of Science and Technology of China, the High-Performance Computing Platform at Beihang University, and the Analysis and Testing Center at Beihang University.

Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Qunfeng Cheng supervised the project, conceived the idea and designed the experiments. Lei Li, Ruohan Xu, and Jia Yan, performed the experiments and characterizations. Wangwei Lian, and Zejun Zhang, performed SEM, AFM, and TEM characterizations. Lei Li, Ruohan Xu, Jia Yan, Jie Sun, Baohua Jia, and Qunfeng Cheng, co-wrote the text. All the authors have approved the final manuscript.

Use of AI statement

None.

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Extremely anisotropic thermally conductive MXene/CNT hybrid film for battery thermal management

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Supporting information to <https://doi.org/10.26599/NR.2026.94909210>

Supplementary text

Preparation of MXene (Ti₃C₂T_x) nanosheets

Ti₃C₂T_x MXene nanosheets were prepared by selectively etching Ti₃AlC₂ via in situ synthesis of hydrofluoric acid using the minimally intensive layer delamination (MILD) method [27]. Briefly, HCl (60 mL, 9 M) was poured into a PTFE vial, and 4.8 g of LiF was added and stirred for 5 min at room temperature. Subsequently, 3 g of Ti₃AlC₂ was slowly added and stirred for 30 h at 50°C. After the etching process, the resultant solution was washed repeatedly with deionized water (DIW) and centrifuged for 5 min at 3500 rpm. The supernatant was decanted and centrifuged several times until the pH exceeded 5. Next, the swelled sediment was re-dispersed in a specific amount of DIW and vibrated for 5 min. The supernatant was collected by centrifugation at 1500 rpm for 30 min. Finally, the obtained supernatant was centrifuged at 4500 rpm for 20 min to collect the sediment, which was again dispersed in a specific amount of DIW to prepare a uniform dispersion. The concentration of the MXene dispersion was measured by vacuum filtration of specific amounts of the dispersion, followed by drying at room temperature for 24 h.

Preparation of CNTs

Briefly, 100 mg of CNT powder and 20 mg of dispersing agent were added to a beaker, followed by the addition of 100 ml of DIW. The mixture was then ultrasonicated at 10°C for 1 hour. Subsequently, the resulting mixed dispersion was centrifuged at 2500 rpm for 20 minutes to eliminate the non-dispersed CNT. Next, different concentrations of CNT dispersions were prepared using water beads. Specifically, 30 water beads were added to the CNT dispersion and left to sit for 6 hours. Afterward, the concentrated CNT dispersion was strained into a new beaker.

Preparation of IBAMC films

The prepared MXene dispersion was centrifuged at 10000 rpm for 20 minutes to obtain a slurry with a concentration of 20 mg mL⁻¹. After ultrasonication (40 W) for 5 minutes, the sol was degassed for 30 minutes. Subsequently, the alternating MXene-CNT (AMC) films were prepared using alternate LbL blade coating. First, the MXene dispersion was blade-coated onto the PVDF substrate with a scraper length of 5 cm and a height of 100 μm at a speed of 50 mm s⁻¹, and the spread paste was dried at 43°C to form the MXene layer. Then, the CNT dispersion was blade-coated onto the MXene layer to form the CNT layer after drying. This process was repeated for 99 cycles, resulting in an AMC film with 50 layers of MXene and 49 layers of CNT (named AMC-III). Moreover, based on different concentrations of the MXene and CNT dispersions, four types of AMC films were prepared using the same process but varying the concentration of CNT (0.5, 1, 2, 3, and 5 mg mL⁻¹), referred to as AMC-I, AMC-II, AMC-IV, and AMC-V films. For comparison, the BMC film was prepared with LbL blade coating based on a mixed MXene/CNT dispersion. A specified amount of MXene dispersion was mixed with CNT dispersion and stirred for 1 hour; then, the mixed dispersion was centrifuged at 10000 rpm for 20 minutes to obtain a uniform dispersion. Next, the AMC-III film was immersed in the prepared solution (0.5 wt%) for 12 hours, washed with ethanol five times, and dried for 12 hours at room temperature. The resulting film was termed interfacial bridging alternating MXene/CNT-II (IBAMC-II). Based on the concentration of the immersed content, three types of IBAMC films were prepared using the same process, differing only in the content of (3,3,3-(0.2, 1, 2, and 5 wt%), referred to as IBAMC-I, IBAMC-III, IBAMC-IV, and IBAMC-V films, respectively.

Porosity calculation

Thermogravimetric analysis (TGA) curves were conducted under a nitrogen atmosphere using a STA 449 F5 and a temperature scan rate of 10°C min⁻¹ from room temperature to 800 °C. The CNT weight fraction (W_1) in the AMC films is approximately given by equation (1):

$$W_1 = \frac{M_{AMC} - M_{MXene}}{M_{CNT} - M_{MXene}} \quad (1)$$

where M_{CNT} , M_{AMC} , and M_{MXene} are the fraction of weight loss for the CNT, AMC, and MXene films between room temperature and 800°C. Since the MXene and CNT weight ratio in IBAMC should be the same as in AMC-III film, the weight fraction (W_2) of (3,3,3-Trifluoropropyl)trimethoxy silane in IBAMC should be approximately given by equation (2):

$$W_2 = 1 - W_{MXene} - W_{CNT} \quad (2)$$

where W_{MXene} and W_{CNT} are the weight fraction of MXene and CNT nanosheets in the IBAMC films. The volume percent porosity is derived from mass density measurements as follows:

$$V_p = 1 - \frac{\rho}{\rho_0} \quad (3)$$

where ρ and ρ_0 represent the bulk density of films and the theoretical density of films without any voids. According to previous literature¹, the theoretical density of MXene nanosheet is 3.68 g cm⁻³. The theoretical densities of MXene, BMC, AMC, and IBAMC films without any voids are derived from the theoretical density of the MXene nanosheet, CNT (1.7 g cm⁻³), and (1 g cm⁻³), respectively.

Orientation calculation

The orientation of the films was characterized using a wide-angle X-ray scattering (WAXS) structural analyzer (Xeuiss 3.0 HR), employing a Cu-K α X-ray beam incident parallel to the film plane and directed at the cross-section region. The detector-sample distance was maintained at 300 mm, with test specimens as 1.5 × 15 mm² strips. The alignment degree of the resultant films was quantified by using the Herman's orientation factor (f), which is calculated by the following formula:

$$f = \frac{1}{2} \langle 3 \cos^2 \phi \rangle \quad (4)$$

Where $\langle \cos^2 \phi \rangle$ is the average value of the cosine of the azimuthal angle for the 002 peak of the resultant films, which is calculated as follows:

$$\langle \cos^2 \phi \rangle = \frac{\int_0^{\pi/2} I(\phi) \cos^2 \phi \sin \phi d\phi}{\int_0^{\pi/2} I(\phi) \sin \phi d\phi} \quad (5)$$

Where $I(\phi)$ is the intensity at the azimuth angle of ϕ .

Anisotropic thermal conductivity measurement and calculation

Intra-plane and inter-plane thermal conductivity of the resultant films were calculated by the following formula (6):

$$\lambda = \alpha \times C_p \times \rho \quad (6)$$

where α , C_p , ρ is thermal diffusivity, specific heat capacity and density. A LFA 467 (NETZSCH, Germany) was used to measure α of the resultant films with voltage and pulse width of 200 V, 50 μ s (inter-plane direction) and 300 μ s (intra-plane direction). The results for each sample were evaluated from the average value of at least five times. Briefly, the thermal diffusivity α was measured using a laser flash apparatus (LFA 467 NETZSCH). According to the dimensions of the sample tray for the LFA 467 NanoFlash system, the sample to be tested was first cut into the circular disc with the diameters of 25.4 mm (intra-plane direction) and 12.8 mm (inter-plane direction) using standard cutter. Then, the moderate graphite was coated on the sample surface to minimize surface reflection and enhance laser energy absorption. The detailed parameters of laser source were setting with voltage and pulse width (200 V 300 μ s, intra-plane direction) and (200 V 50 μ s, intra-plane direction). The thermal diffusivity values are measured at 5 points on the same sample. Subsequently, the thicknesses (d) of samples were measured by SEM (scanning electron microscope). And the thermal diffusivity α can be calculated by (7):

$$\alpha = \frac{0.1388d^2}{t_{50}} \quad (7)$$

Where d is the sample thickness and t_{50} is the characteristic time for the sample to heat up to the half of the maximum temperature. Furthermore, the C_p (J g⁻¹ K⁻¹) was evaluated by using differential scanning calorimeter (DSC Q2000, TA Instruments) measurements. Density ρ was calculated based on the weight and volume. The C_p was determined according to the following equation (8):

$$C_{composites} = C_{filler} \times \phi + C_{matrix} \times (1 - \phi) \quad (8)$$

Where " ϕ " corresponds to the mass fraction, and $C_{MXene}(25^\circ\text{C})$ and $C_{CNT}(25^\circ\text{C})$ were tested as 1.12 J g⁻¹ °C⁻¹ and 1.54 J g⁻¹ °C⁻¹.

Finite-element analysis

The COMSOL Multiphysics 6.3 software was used to simulate the heat transfer properties of IBAMC and MXene films. In this modeling, the analysis was performed in a 50 × 0.03 μ m rectangular region, applying a time-dependent mode. The bottom boundary adopted a linear heat source with a constant temperature of 100°C, and the upper boundary was set with a natural heat convection of 20 W m⁻² K⁻¹ in a 25°C ambient environment. The left and right boundaries were set as adiabatic to ignore the lateral heat transfer effect. The length and thickness of MXene nanosheets were set 10 μ m and 1.5 nm. The length and diameter of CNTs were set 20 μ m and 1 nm. The intra-plane thermal conductivity, inter-plane thermal conductivity, and specific

heat capacity of MXene nanosheet and CNT was set $108 \text{ W m}^{-1} \text{ K}^{-1}$, $0.38 \text{ W m}^{-1} \text{ K}^{-1}$, $1120 \text{ J kg}^{-1} \text{ }^{\circ}\text{C}^{-1}$ and $1100 \text{ W m}^{-1} \text{ K}^{-1}$, $10 \text{ W m}^{-1} \text{ K}^{-1}$, $1540 \text{ J kg}^{-1} \text{ }^{\circ}\text{C}^{-1}$, respectively [21, 43, 44]. The investigated time region was set from 0 to 10 nanosecond (ns) with a step length of 0.05 ns.

Battery thermal management measurement

The vertically 18650 lithium-ion battery mounted on a heat sink was interfaced with IBAMC films using 1.0 g thermal silicon grease ($3.5 \text{ W m}^{-1} \text{ K}^{-1}$). The charging and discharging tests are performed by connecting the battery to the tester, and the real-time temperature of the battery was record by the infrared camera (Fotric 228S) which vertically placed on top of the battery.

Just Accepted

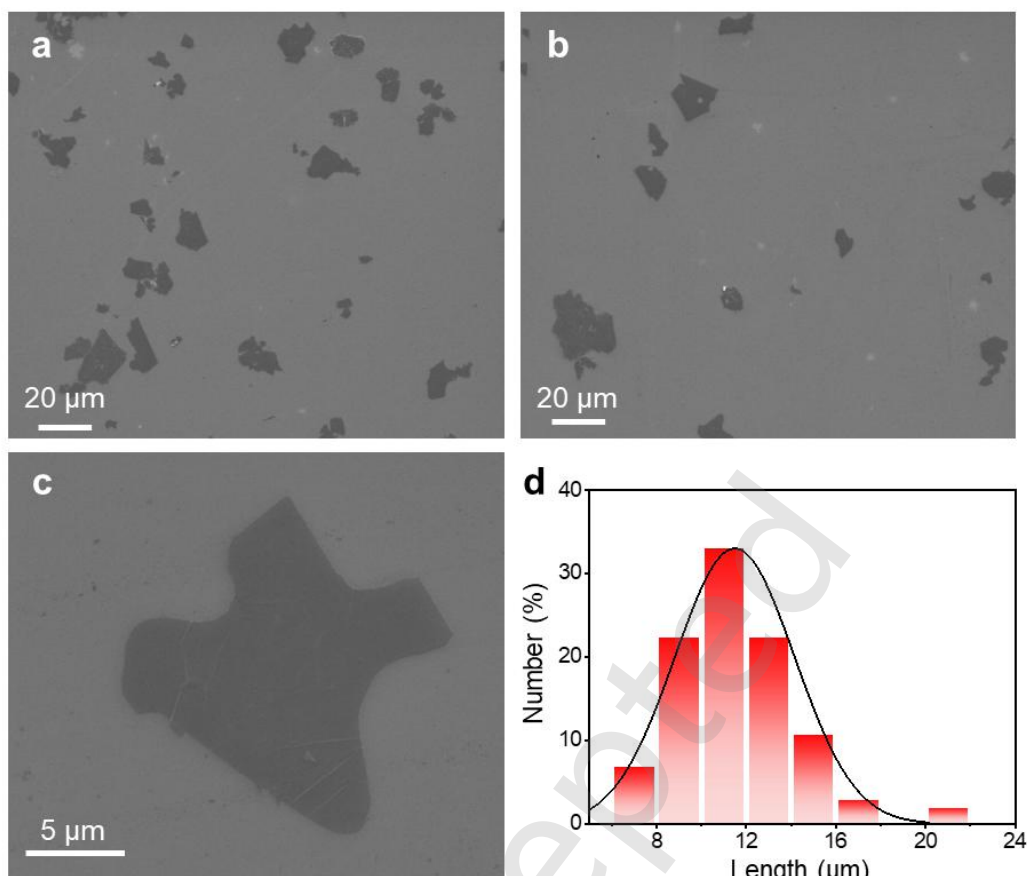


Figure S1. Measuring the average maximum lateral size of MXene nanosheets. SEM images and the distribution of maximum lateral sizes of MXene nanosheets for (a), (b), (c), and (d). These results show that the average maximum lateral platelet size is 11.5 μm.

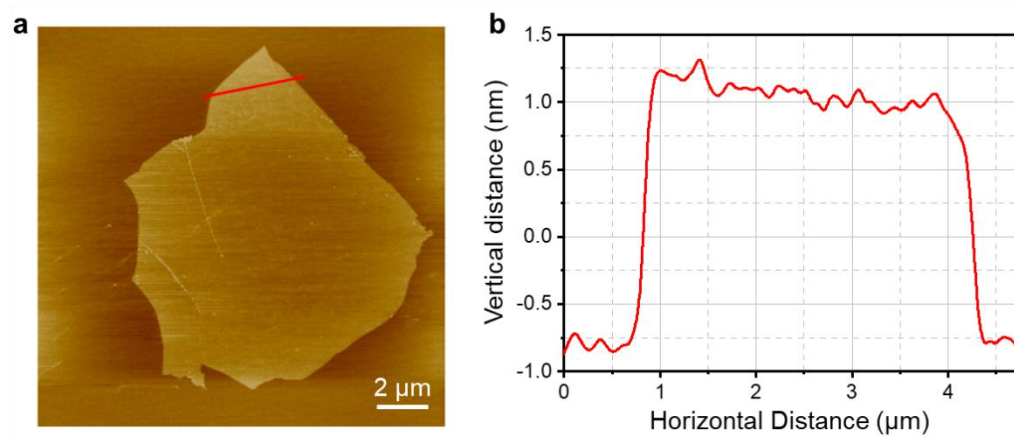


Figure S2. The thickness of a MXene nanosheet. (a) AFM image of a MXene nanosheet. (b) Thickness profile for the depicted MXene nanosheet.

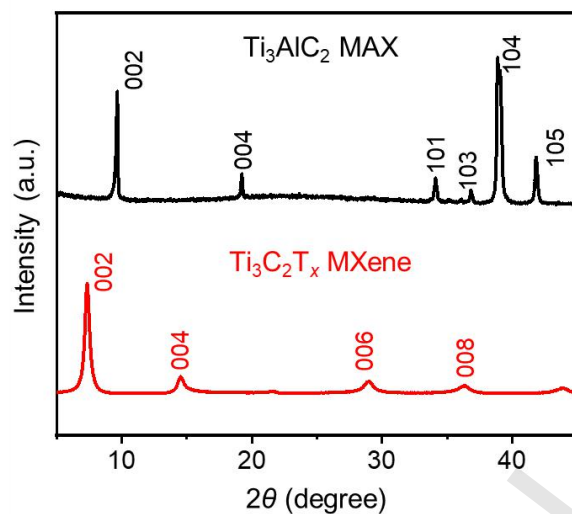


Figure S3. XRD curves were obtained using Cu-K α radiation for Ti_3AlC_2 MAX and $\text{Ti}_3\text{C}_2\text{T}_x$ MXene. The monolayer MXene nanosheets were successfully prepared by exfoliating Ti_3AlC_2 MAX (the top diffraction pattern) with the MILD method, which provided the bottom diffraction pattern.

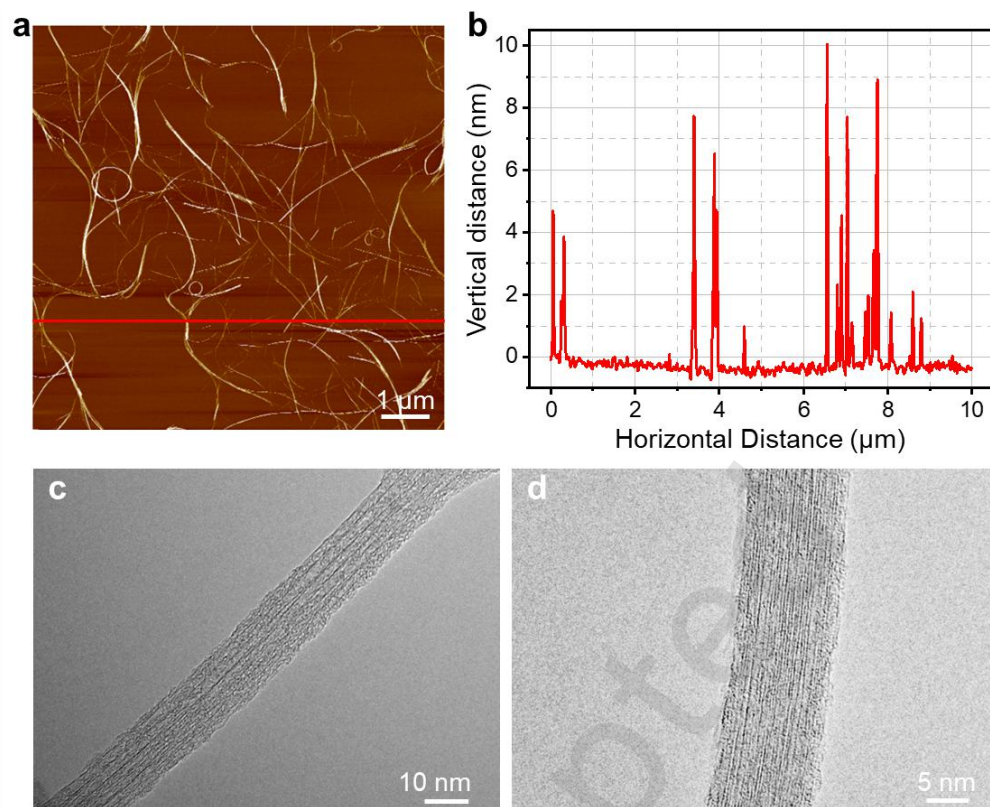


Figure S4. The morphology of CNTs. (a) AFM image of CNTs. (b) The height measurement of CNTs. (c) and (d) TEM images of CNTs at various magnifications.

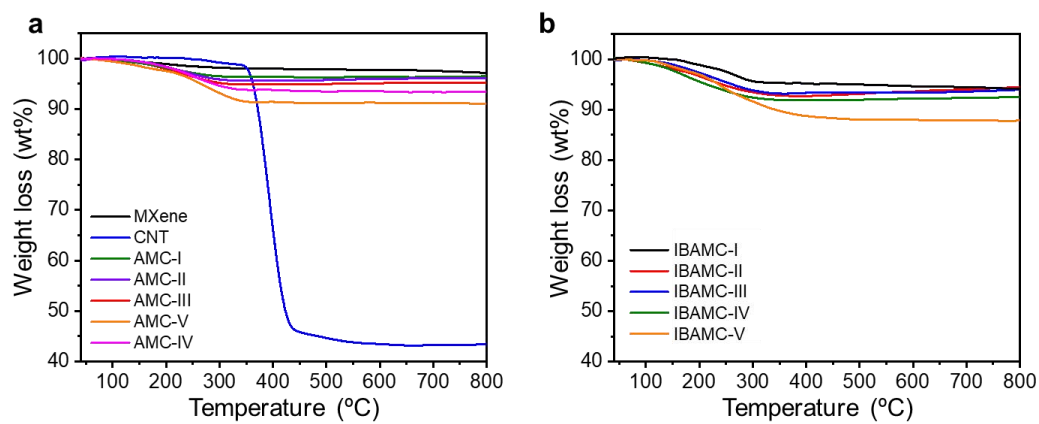


Figure S5. TGA curves for $10^{\circ}\text{C min}^{-1}$ scans of MXene, CNT, AMC, and IBAMC films. (a) Scans for MXene, CNT, and various types of AMC films. (b) Scans for different types of IBAMC films.

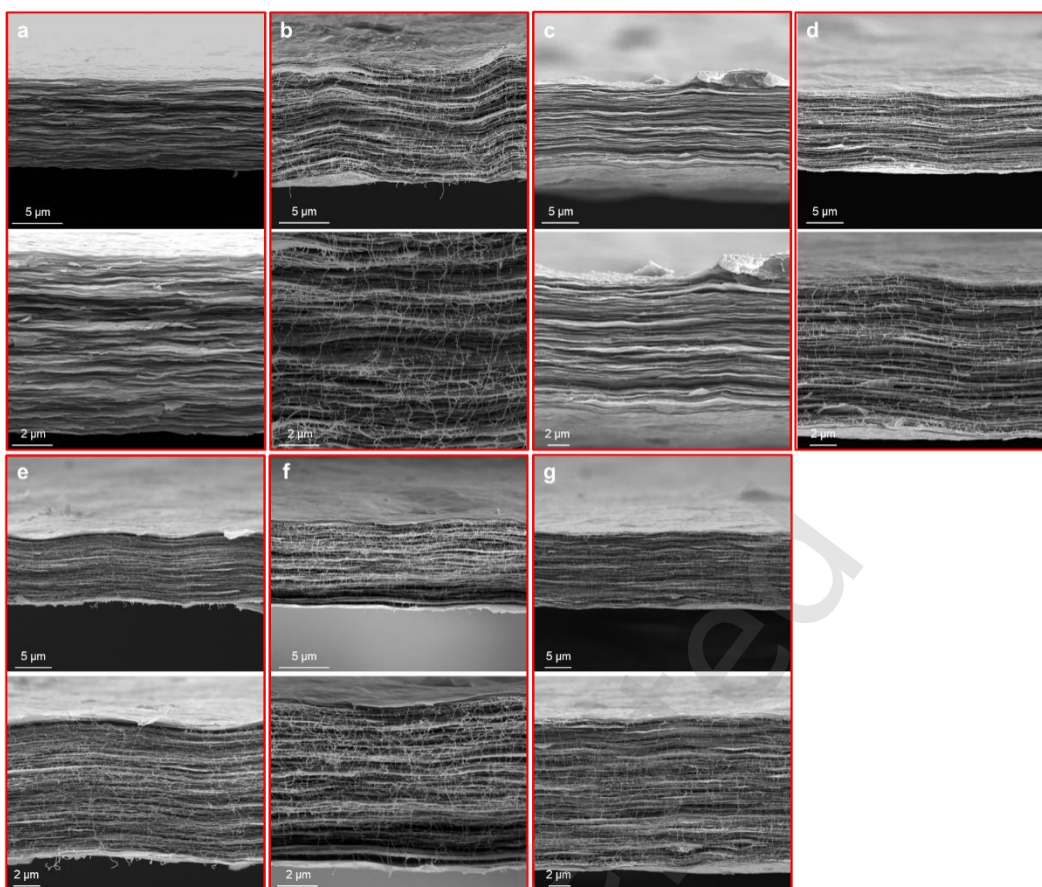


Figure S6. SEM images of the tensile fracture surfaces of the resulting films. (a) MXene, (b) BMC, and (c-g) AMC-I to AMC-V.

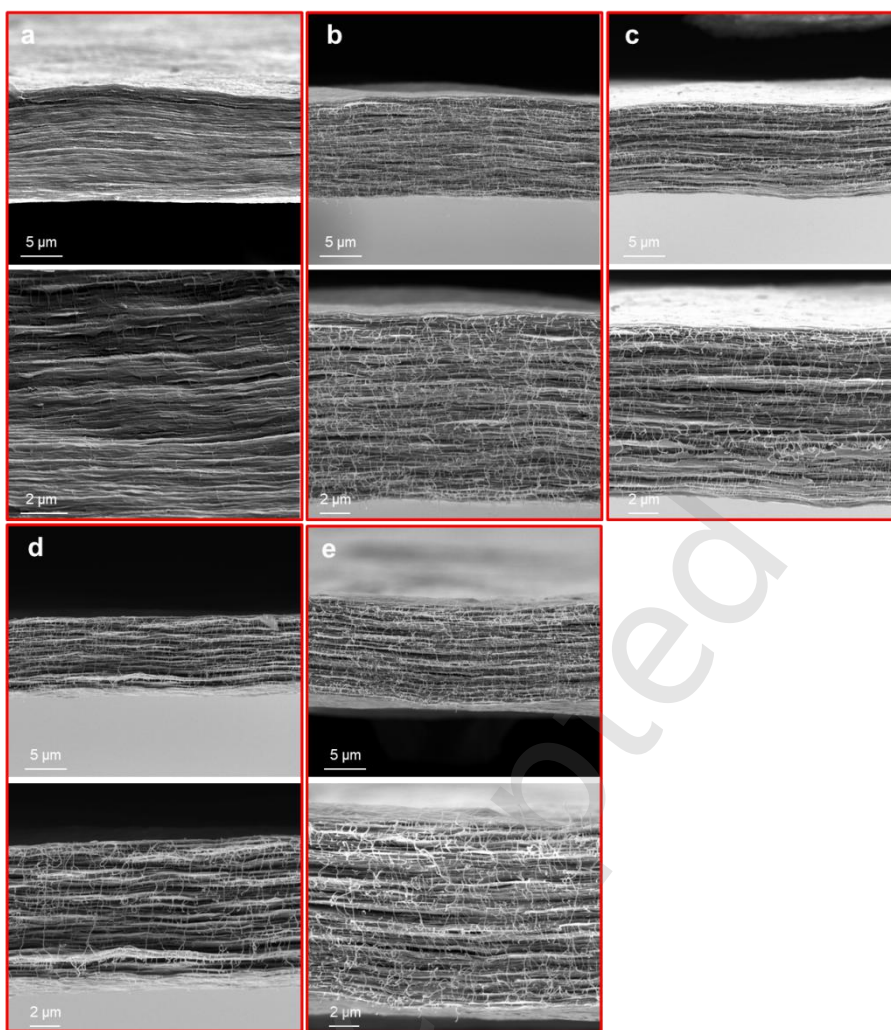


Figure S7. SEM images of the tensile fracture surfaces of the resulting films. (a to e) IBAMC-I to IBAMC-V.

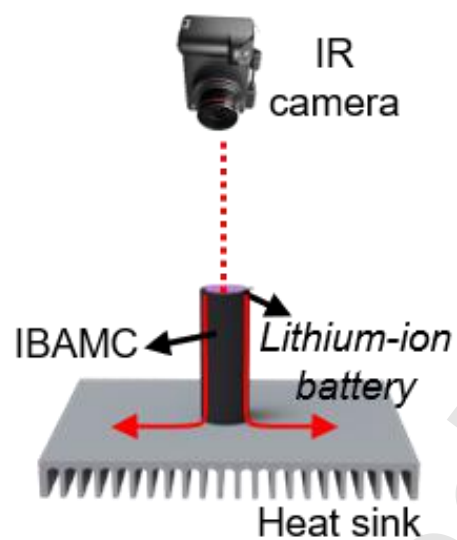


Figure S8. Schematic illustrating the use of the IBAMC films integrated into the 18650 lithium-ion battery.

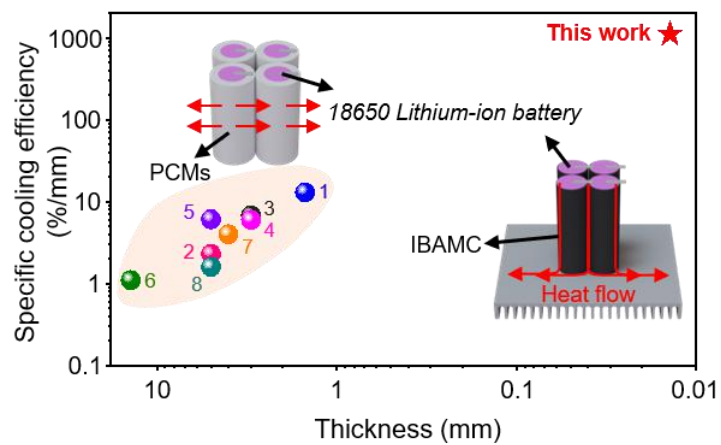


Figure S9. Comparison of thickness and the specific cooling efficiency of the IBAMC film with those of PCMs for thermal management of commercial 18650 lithium-ion battery.

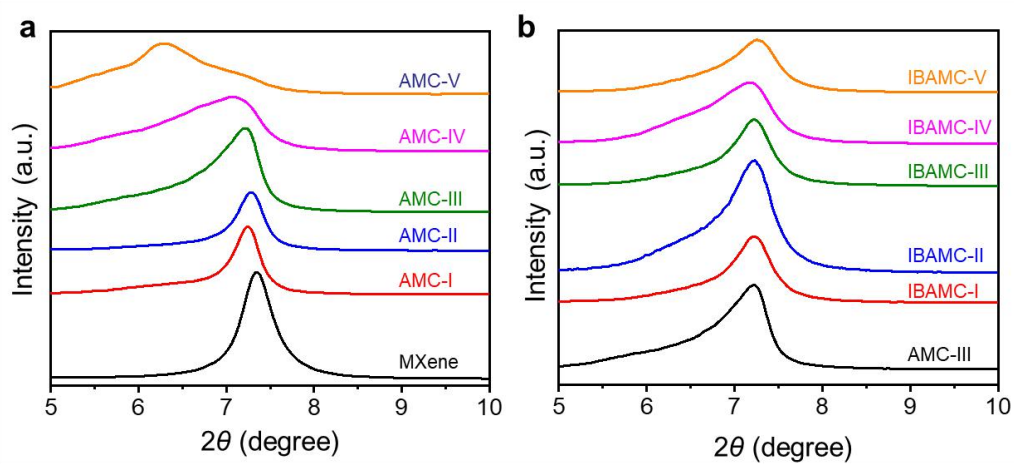


Figure S10. XRD curves obtained using Cu-K α radiation. (a) MXene and AMC films. (b) AMC-III and IBAMC films.

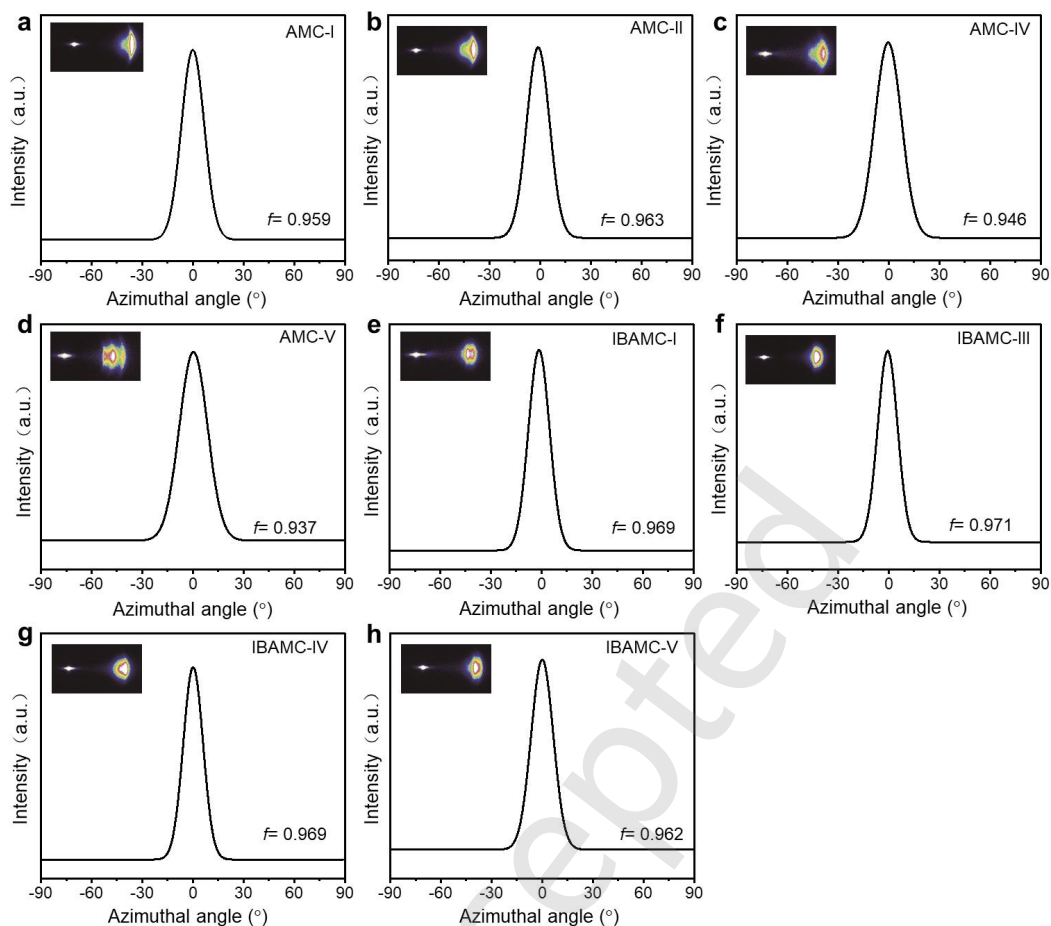


Figure S11. WAXS patterns were obtained for an incident Cu-K α X-ray beam parallel to the film plane, along with the corresponding azimuthal scan profiles for the (002) peak. AMC-I, AMC-II, AMC-IV, and AMC-V films (a-d), and the IBAMC-I, IBAMC-III, IBAMC-IV, and IBAMC-V films (e-h).

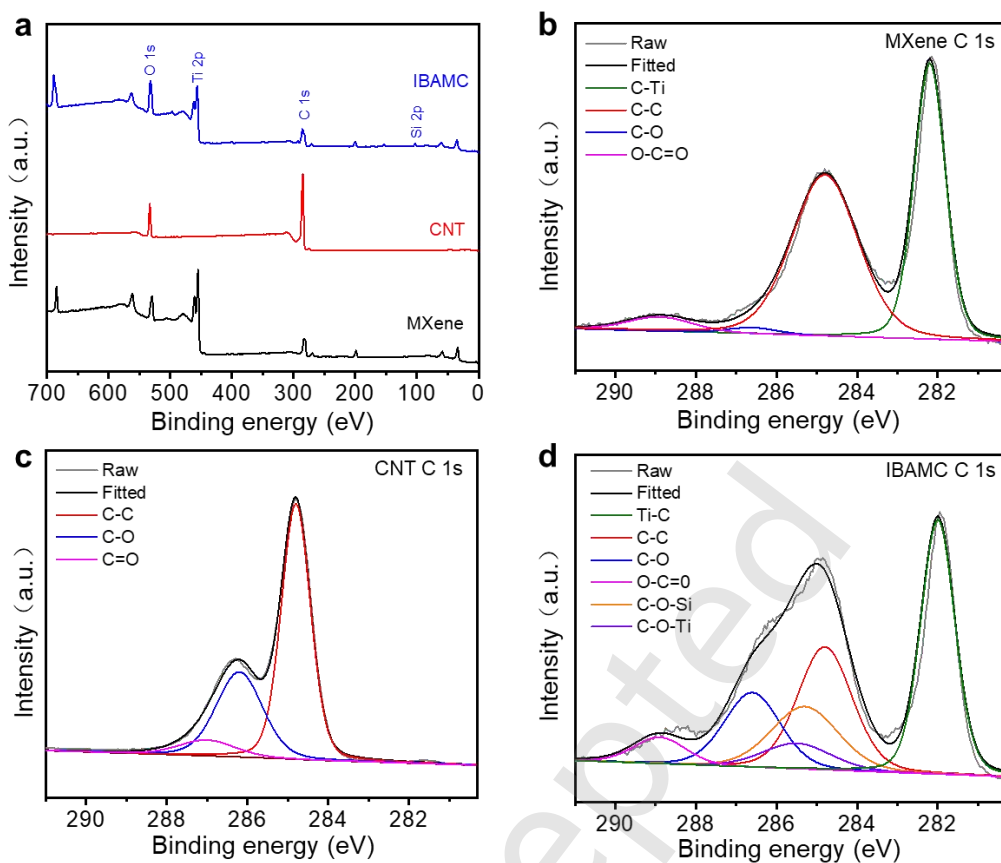


Figure S12. XPS spectra and component analysis for (a) MXene, CNT, and IBAMC films; C 1s spectra of (b) MXene film, (c) CNT, and (d) IBAMC films.

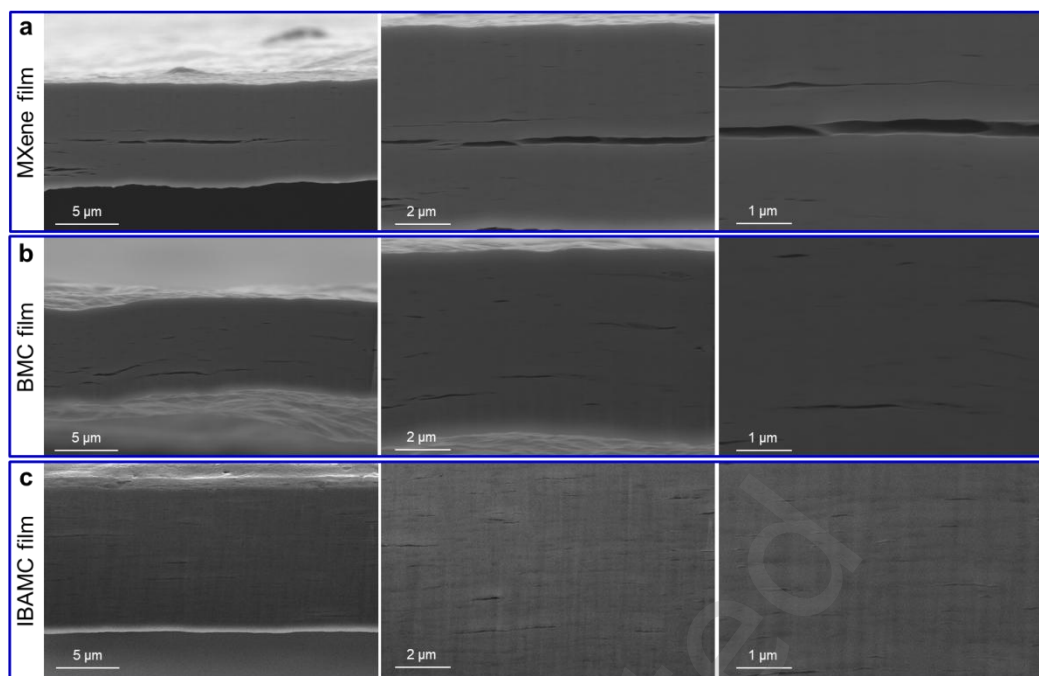


Figure S13. SEM images of the IBAMC film cross-sections obtained through FIB cutting: (a) MXene film, (b) BMC films, and (c) IBAMC films.

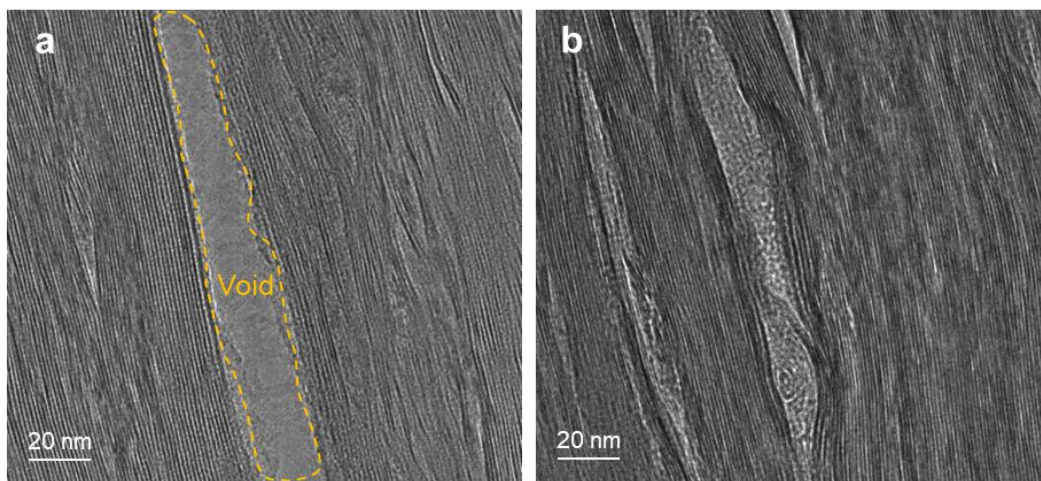


Figure S14. High-resolution transmission electron microscopy (HR-TEM) images of (a) the MXene film and (b) the IBAMC film.

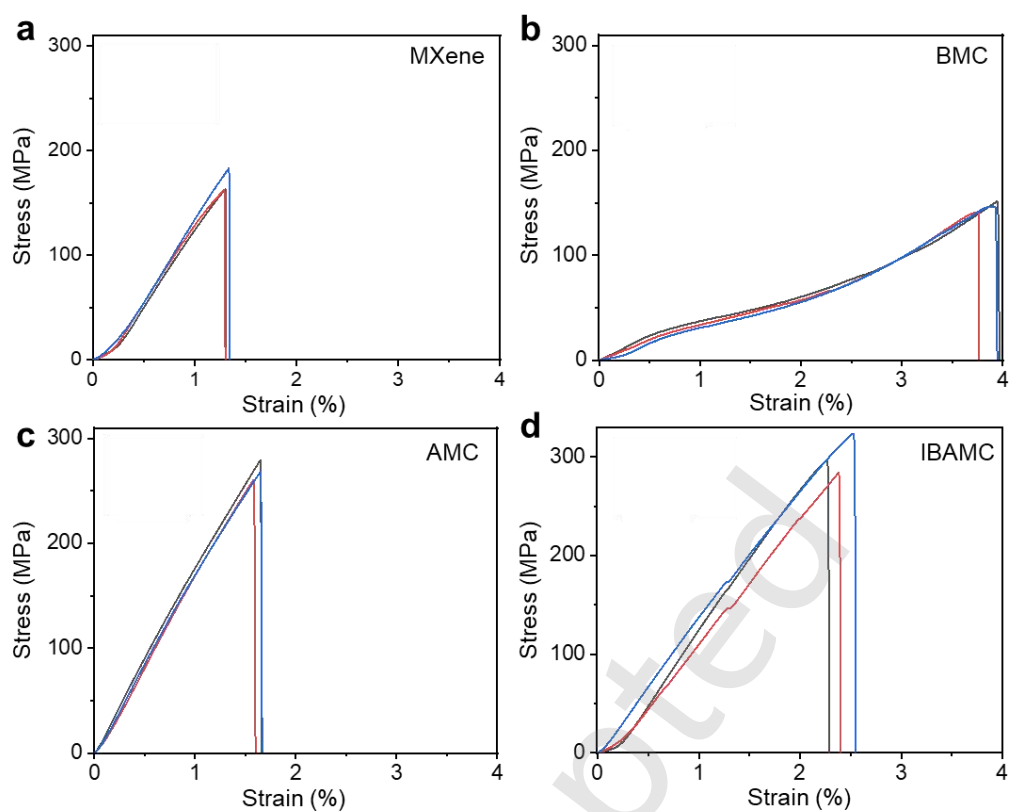


Figure S15. The typical stress-strain curves are represented as follows: (a) MXene film, (b) BMC film, (c) AMC film, and (d) IBAMC film.

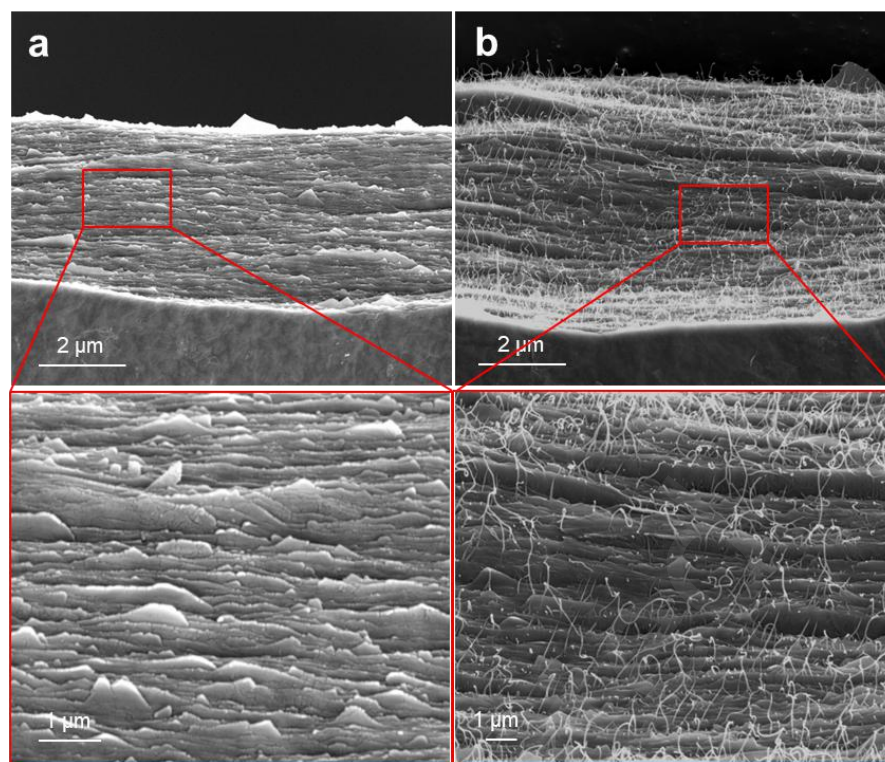


Figure S16. Inclined-view SEM images of the fracture surface of the MXene and AMC films. (a) MXene film, (b) AMC film.

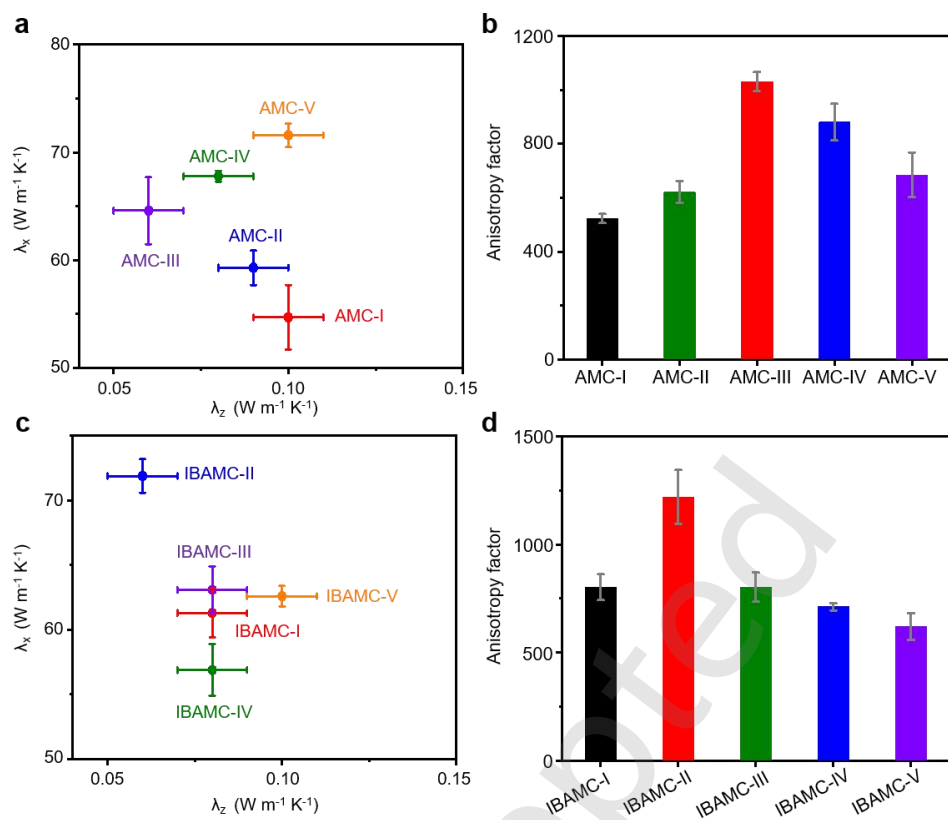


Figure S17. The thermal anisotropy performance of AMC and IBAMC films. λ_x and λ_z of (a) AMC-I to AMC-V films and (c) IBAMC-I to IBAMC-V films. Anisotropy factors of (b) AMC-I to AMC-V films and (d) IBAMC-I to IBAMC-V films.

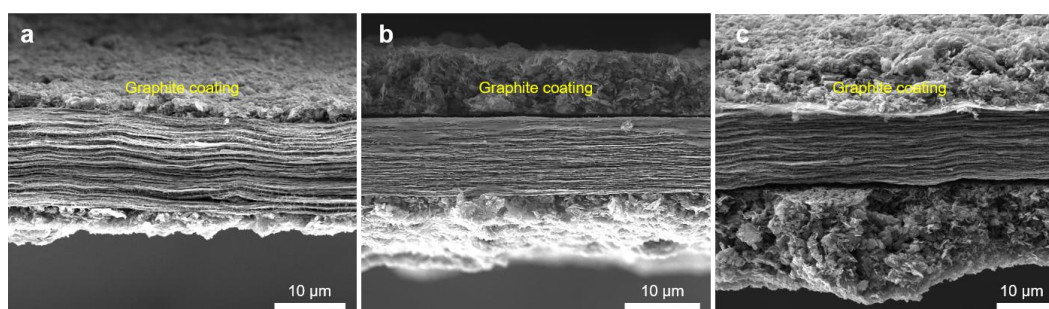


Figure S18. SEM images of the tensile fracture surfaces of the IBAMC films with different graphite coating thickness. (a) $3.1 \pm 0.6 \mu\text{m}$, (b) $5.6 \pm 1.3 \mu\text{m}$, and (c) $11.7 \pm 2.7 \mu\text{m}$.

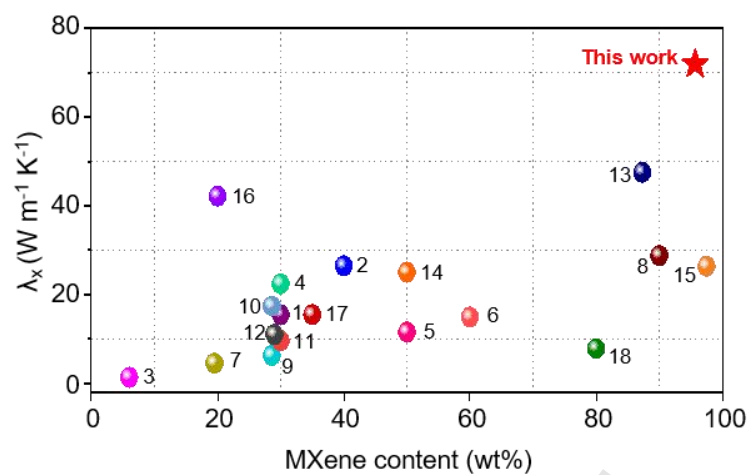


Figure S19. Comparison of λ_x and MXene content of the IBAMC film with those of other MXene films.

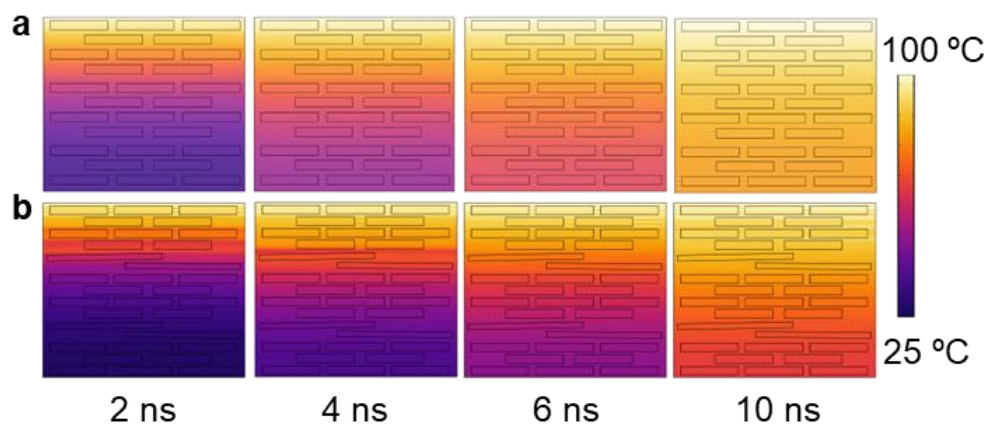


Figure S20. Finite element simulated temperature evolution through plane during 10 nanoseconds (ns) heating. (a) MXene film, (b) IBAMC film.

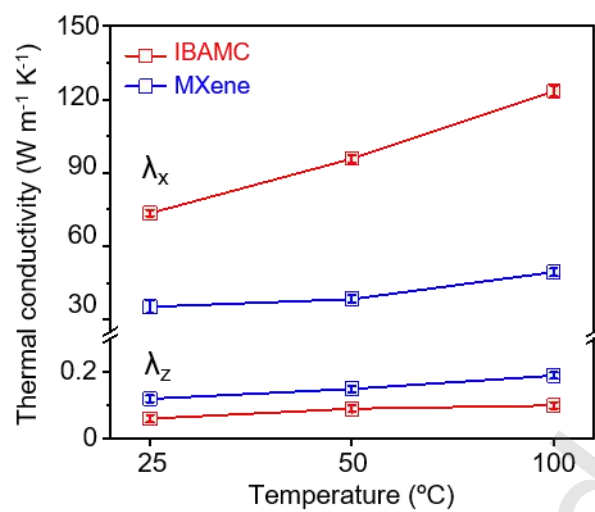


Figure S21. λ_x and λ_z of Mxene and IBAMC films at different temperatures.

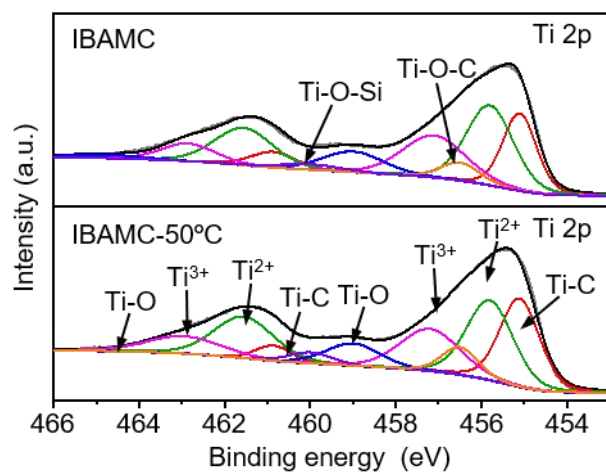


Figure S22. XPS spectra and component analysis for Ti 2p spectra of IBAMC and IBAMC-50°C films.

Supplementary tables

Table S1. The MXene, CNT, and (3,3,3-Trifluoropropyl)trimethoxy silane contents in AMC and IBAMC films determined by TGA measurements.

Sample	MXene content (wt%)	CNT content (wt%)	(3,3,3-Trifluoropropyl)trimethoxy silane content (wt%)
AMC-I	98.7	1.3	/
AMC-II	98.2	1.8	/
AMC-III	96.4	3.6	/
AMC-IV	92.7	7.3	/
AMC-V	88.7	11.3	/
IBAMC-I	95.7	3.6	0.7
IBAMC-II	95.2	3.6	1.2
IBAMC-III	95.1	3.5	1.4
IBAMC-IV	93.8	3.5	2.7
IBAMC-V	89.2	3.3	7.5

Table S2. λ_x , λ_z , anisotropy factor of IBAMC films and previously reported anisotropic thermally conductive films.

Number	Films	Content (wt%)	λ_x (W m ⁻¹ K ⁻¹)	λ_z (W m ⁻¹ K ⁻¹)	Anisotropy factor	Preparing method	Measuring method	Reference
1	GN/PU	10	10.68	0.46	23.2	Solution casting	Laser flash	45
2	PI/LC-GeF	15	4.21	0.63	6.7	Blade coating/imidization	Laser flash	46
3	NFC/Al ₂ O ₃ -rGO	40	11.5	0.5	23	Vacuum filtration	Laser flash	47
4	NFC/rGO	1	12.6	0.042	279	LbL assembly	Laser flash	10
5	rGO/CNF	50	7.3	0.13	56	Vacuum filtration	Laser flash	48
6	GN/PVA	10	13.8	0.6	23	LbL assembly	Laser flash	11
7	ANF/GN	20	20.54	0.11	187	Vacuum filtration	Laser flash	49
8	PPA@GNP-PVA/GNP	61	82.4	1.45	56.8	LbL assembly	Laser flash	5
9	ANF/GF	40	48.2	1.2	40.2	Sol-gel-film conversion	Laser flash	50
10	CNF/Ag-rGO	9.6	27.55	0.253	108.9	Vacuum filtration	Laser flash	51
11	rGO	100	61	0.09	675	Blade coating/high temperature	Laser flash	52
12	Graphene/PVP	10	81.2	5.1	15.9	Vacuum filtration	Laser flash	53
13	BNNS/PVA	83	67.6	3.57	18.9	Vacuum filtration	Laser flash	54
14	BNNS-VHP	85	73.5	0.25	297.4	Tape casting/Vacuum hot- pressing	Laser flash	55
15	BNNS/ANF	30	46.7	0.13	372	Vacuum filtration	Laser flash	12
16	BNNS/BP/CNF	85	72.5	0.9	80.6	Vacuum filtration	Laser flash	13
17	ANF/BNNS/AgNWs	50	9.44	0.75	12.6	Vacuum filtration	Laser flash	56
18	MXene/Ag/PVA	60	3.72	0.41	9.1	Pouring Evaporative self- assembly	Laser flash	57
19	MXene-PDA-Ag/NFC	30	7.32	1.34	5.4	Vacuum filtration	Laser flash	58
20	MXene/ANF/PVA/L M	/	13.2	0.71	18.5	Sol-gel-film conversion	Laser flash	22
21	MXene/ANF	30	15.5	0.82	19	Sol-gel-film conversion	Laser flash	59
22	MXene/CNF/LM	90	7.83	0.13	58	Vacuum filtration	Laser flash	60
23	MXene/CNT/CNF	50	25.06	1.64	15.3	Vacuum filtration	Laser flash	61
24	MXene/ND/CNF	28.6	17.43	0.55	32	Vacuum filtration	Laser flash	62
25	MXene/Pt	/	24.45	0.7	35	Vapor phase infiltration	Laser flash	63
26	MXene/MMT	90	28.8	0.27	107	Vacuum filtration	Laser flash	64
27	MXene/PBO	20	42.2	0.103	409.7	Sol-gel-film conversion	Laser flash	65
28	IBAMC	95.7	71.9	0.06	1220	Alternately layer-by-layer blade coating	Laser flash	This work

Table S3. Thermal conductivity (TC), thickness, temperature of battery (T_1), and temperature of battery wrapped with materials (T_2), the temperature reduction (T_3), cooling efficiency (η), and the specific cooling efficiency of IBAMC film for cooling 18650-type lithium-ion battery during 3 capacity (3C) discharge rate and previously reported phase change materials.

Composites	TC (W m ⁻¹ K ⁻¹)	Thickness (mm)	Discharge rate	T_1 (°C)	T_2 (°C)	T_3 (°C)	η (%)	Specific η (%/mm)	Reference
EG/OBC/PW	8.3	1.5	3C	61	49.1	11.9	19.5	13.0	40
TPU/Graphene	12.3	5	3.5C	49.8	44.1	5.7	11.4	2.3	31
CaCl ₂ /CF/PW/EG	3.9	3	3C	40.5	38	7.8	19.3	6.4	66
PEG/BN/CNT/SiC/ EG	0.95	3	3C	54	43.8	10.2	18.9	6.3	41
LA/SA/EVA/AlN	0.73	5	3C	46.1	32.1	14	30.4	6.1	67
PA/EG/SEPS	1.58	14	3C	51	43	8	15.7	1.1	68
AlN/PA/EG	1.4	4	3C	59.0	49.5	9.5	16.1	4.0	69
TPC-et	1.6	5	3C	44.6	41.1	3.5	7.8	1.6	70
IBAMC	71.9	0.014	3C	48.4	40.8	7.6	15.7	1121.6	This work

Table S4. Interlayer diffraction spacing of MXene, BMC, AMC, and IBAMC films.

Sample	<i>d</i> -spacing (nm)
MXene	1.20
BMC	1.30
AMC-I	1.22
AMC-II	1.21
AMC-III	1.22
AMC-IV	1.23
AMC-V	1.41
IBAMC-I	1.22
IBAMC-II	1.23
IBAMC-III	1.23
IBAMC-IV	1.24
IBAMC-V	1.21

Table S5. Tensile strength, strain, toughness, and Young's modulus of MXene, BMC, AMC, and IBAMC films.

Sample	Tensile Strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Young's modulus (GPa)
MXene	169.5±12	1.32±0.02	1.02±0.08	15.6±0.9
BMC	146.0±5.7	3.89±0.11	2.91±0.18	3.4±0.2
AMC	270.7±9.3	1.63±0.04	2.81±0.15	16.2±0.3
IBAMC	302.2±19.6	2.40±0.13	3.50±0.70	12.9±0.8

Table S6. λ_x , λ_z , density, specific heat capacity, and anisotropy factor of MXene, BMC, AMC, and IBAMC films.

Sample	Density (g cm ⁻³)	Specific heat capacity (J g ⁻¹ °C ⁻¹)	λ_x (W m ⁻¹ K ⁻¹)	λ_z (W m ⁻¹ K ⁻¹)	Anisotropy factor
MXene	3.28	1.12	37.1 ± 2.7	0.12 ± 0.01	302 ± 8
BMC	3.19	1.13	55.1 ± 3.0	0.14 ± 0.01	388 ± 9
AMC-I	3.45	1.13	54.7 ± 3.0	0.10 ± 0.01	523 ± 17
AMC-II	3.43	1.13	59.3 ± 1.6	0.09 ± 0.01	621 ± 40
AMC-III	3.33	1.14	64.6 ± 3.1	0.06 ± 0.01	1030 ± 36
AMC-IV	3.24	1.15	67.8 ± 0.5	0.08 ± 0.01	880 ± 68
AMC-V	3.14	1.17	71.6 ± 1.1	0.10 ± 0.01	685 ± 83
IBAMC-I	3.21	1.14	61.3 ± 1.9	0.08 ± 0.01	803 ± 60
IBAMC-II	3.31	1.14	71.9 ± 1.3	0.06 ± 0.01	1220 ± 124
IBAMC-III	3.34	1.14	63.1 ± 1.8	0.08 ± 0.01	803 ± 68
IBAMC-IV	3.37	1.14	56.9 ± 2.0	0.08 ± 0.01	712 ± 16
IBAMC-V	3.35	1.14	62.6 ± 0.8	0.10 ± 0.01	621 ± 62

Table S7. λ_x and λ_z of IBAMC film with different graphite coating thickness.

Sample	Graphite coating thickness (μm)	λ_x ($\text{W m}^{-1} \text{K}^{-1}$)	λ_z ($\text{W m}^{-1} \text{K}^{-1}$)
IBAMC	3.1 ± 0.6	71.5 ± 0.9	0.06 ± 0.01
	5.6 ± 1.3	71.9 ± 1.3	0.06 ± 0.01
	11.7 ± 2.7	69.1 ± 2.2	0.06 ± 0.01

The different graphite coating thickness IBAMC films show the similar anisotropic thermal conductivity, which demonstrates that the coating thickness does not affect the thermal conductivity.

Table S8. λ_x of IBAMC films and previously reported thermally conductive MXene films.

Number	MXene films	MXene Content (wt%)	λ_x ($\text{W m}^{-1} \text{K}^{-1}$)	Testing method	Reference
1	CNF/MXene/Agnws	35	15.53	Laser flash	71
2	MXene/Graphene	40	26.49	Laser flash	72
3	MXene/Co/PVDF	6	1.36	Laser flash	73
4	MXene/Ag/CNF	30	22.43	Laser flash	74
5	MXene/CNF	50	11.57	Laser flash	75
6	MXene/CNF	60	14.93	Laser flash	76
7	MXene/PVA	19.5	4.57	Laser flash	28
8	MXene/MMT	90	28.8	Laser flash	64
9	MXene/TPU	28.6	6.31	Laser flash	77
10	MXene/ANF	30	15.5	Laser flash	59
11	MXene/MCC	30	9.67	Laser flash	78
12	MXene/BNNS	29	11.02	Laser flash	79
13	MXene/PVA	87.3	47.6	Laser flash and Raman	80
14	MXene/CNT/CNF	50	25.06	Laser flash	61
15	MXene/SiO ₂	97.5	26.4	Laser flash	81
16	MXene/PBO	20	42.2	Laser flash	65
17	MXene/ND/CNF	28.6	17.43	Laser flash	62
18	MXene/CNF/LM	80	7.83	Laser flash	60
19	IBAMC	95.7	71.9	Laser flash	This work