

In-Plane Molecular Cord Bridging as a Method for Boosting MXene Fiber Strength and Toughness

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This is a well-executed study that provides a clear and convincing explanation of the proposed toughening mechanism in MXene fibers. The experimental results are comprehensive, and the reported mechanical and multifunctional performance is impressive. However, similar enhancement mechanisms, particularly those involving in-plane bridging and intersheet reinforcement in MXene-based fibers and films, have already been demonstrated in earlier works (see comment 1). As a result, the conceptual novelty of the present strategy appears more incremental. Additionally, the method employed, including thermal densification, has been demonstrated previously (Nature Communications 13 (2022) 4564).

Given the relatively incremental novelty, I would not recommend this manuscript for publication in Nature Communications, as works in this journal family are expected to emphasize advances in fundamental understanding. Much of the fundamental mechanisms discussed in the submitted manuscript have already been well established. The study may, however, be suitable for journals that focus on record mechanical or multifunctional performance, as it demonstrates excellent performance enhancements arising from the combination of new materials.

Additional specific comments are as follows:

1. Early studies by Wan et al. and Usman et al. (Science 374 (2021) 96–99; Nature Communications 13 (2022) 4564; 2D Materials 9 (2022) 044003) pioneered sequential in-plane and interlayer bridging of MXene nanosheets to enhance stress transfer, densification, and toughness. While their performance metrics were lower, these works established the core mechanistic principles of sequential reinforcement, including load sharing, delayed crack initiation, and energy dissipation. Many recent reports build on this same framework, often achieving incremental improvements rather than fundamentally new concepts. The authors should clearly articulate how the present molecular cord strategy advances beyond these foundational studies and justify its novelty in the context of established sequential bridging mechanisms.
2. Carbon nanotubes (CNTs) are well known for imparting high mechanical strength; however, it would be valuable to clarify whether the observed molecular cord bridging mechanism is intrinsically dependent on CNTs or whether other one-dimensional additives could achieve a similar strengthening effect. In many composite systems, the inclusion of CNTs is often considered a straightforward or expected route to mechanical reinforcement, which can obscure the specific contribution of the proposed mechanism. From a mechanistic perspective, the exceptionally high Young's modulus of CNTs may even be excessive, as it could partially constrain interfacial deformation and limit the full activation of the molecular cord bridging process. Additionally, the cost-effectiveness of CNTs as a reinforcement is a recurring concern in previous studies. This suggests that more compliant or chemically tunable one-dimensional additives may be equally, or potentially more, effective in enabling this mechanism. Clarifying this distinction would strengthen the mechanistic interpretation and broaden the design space for future material systems.
3. Previous studies have attributed the balanced mechanical properties and enhanced strength of MXene based composites not only to interflake connectivity but also to the intrinsic anisotropy and alignment of the MXene sheets. In this context, it is important to clarify whether flake alignment contributes to the observed mechanical performance in the present system. The X ray scattering data provided in the Supporting Information do not indicate any improvement in alignment. Can the authors provide further clarification on this point?

4. The lack of a clearly defined target application limits the broader appeal of the work. While the study demonstrates record mechanical and electrical performance, it is not immediately clear which specific type of supercapacitor or practical use is intended. For example, is the system designed for supercapacitors that undergo repeated bending or for applications that primarily require high tensile strength? Clarifying the intended application and explaining how the molecular cord strategy addresses a specific functional or practical need would strengthen the manuscript. Providing this context would help position the study within a purposeful materials design framework, rather than appearing solely as a combination of high-performance components aimed at surpassing existing benchmarks.

Reviewer #2

(Remarks to the Author)

Review comments on NCOMMS-25-94906-T (Pushing Strength and Toughness of MXene fibers to New Limits via In-Plane Molecular Cord Bridging)

This study presents a paradigm-shifting strategy to fabricate high-performance MXene-based fibers by addressing the fundamental bottleneck of weak in-plane edge-to-edge interactions between MXene nanosheets. Inspired by the cord-bound architecture of ancient lamellar armor, the authors employed carboxymethyl cellulose-wrapped single-walled carbon nanotubes (CS) as "molecular cords" to bridge adjacent MXene flakes via robust Mg^{2+} -mediated ionic coordination bonds. Through a synergistic process of wet-spinning, ionic crosslinking, and controlled hot-pressing, the resulting fibers (containing only 0.12 wt% CS) achieved a remarkable tensile strength of 1.48 ± 0.06 GPa, an unprecedented toughness of 131.0 ± 8.7 MJ m⁻³, and an exceptional electrical conductivity exceeding 10,400 S cm⁻¹. These are great achievements in MXene based fiber research. However, for this paper to be published in Nature Communications, it is essential to provide additional supplementation and rigorous verification regarding the following academic concerns. Therefore, the manuscript can be further considered after major revision. The comments are as follows.

1. (Line 99) The manuscript employs carboxymethyl cellulose (CMC) to form strong ionic coordination bonds; however, the rationale for selecting CMC over other ion-rich or carboxylate-containing polymers is not sufficiently discussed. A brief discussion in the Introduction part highlighting the advantages of CMC, such as its coordination capability, interfacial compatibility, and resulting performance benefits, would further clarify why CMC is particularly well suited for this system.
2. (Line 152) The authors state that CMC wraps the single-walled carbon nanotubes (SWNTs); however, the specific interactions responsible for this wrapping are not clearly described. While SEM images of fractured fiber cross-sections suggest effective CNT dispersion and mechanical bridging, these observations do not directly evidence CMC wrapping of SWNTs, particularly in the solution state. Further mechanistic discussion and supporting experimental evidence (direct or indirect) are necessary to substantiate the claimed CMC wrapping.
3. (Line 163) The authors report that the viscosity of the MXene-based spinning dopes exceeds 1000 Pa·s at low shear rates. In conventional wet-spinning, the viscosity of dopes is typically maintained below 100 Pa·s to ensure stable extrusion and prevent structural defects. Given this exceptionally high viscosity, the authors should clarify how they achieved stable fiber formation through a 26 G needle at a rate of 5 mL·h⁻¹ without encountering issues such as high back-pressure or stable extrusion. It is recommended to provide capillary rheometry data or jet profile images at the nozzle tip to prove the flow stability and shear-thinning behavior under actual spinning conditions.
4. (Line 178) Regarding the structural characteristics shown in Supplementary Figures 7 and 8, the authors report that C-MXene fibers exhibit a more densified structure with lower porosity compared to neat MXene fibers. However, a significant discrepancy arises as the orientation factor of C-MXene (0.80) is substantially lower than that of neat MXene (0.91), despite its higher compactness. This inverse correlation suggests that the inclusion of CMC polymer chains alone may induce steric hindrance or disrupt the nematic liquid crystalline order during the spinning process. The authors should provide a clear physical explanation for this complex trend of orientation in C-MXene and further elucidate how the incorporation of CS (SWCNTs) effectively restores the alignment to 0.91 in the CS-MXene system.
5. (Line 212) Regarding the XPS analysis in Fig. 2g, the absence of Mg 1s and survey spectra is a critical omission, as these are essential to verify the successful incorporation and chemical state of the Mg^{2+} crosslinkers. Furthermore, relying solely on C 1s shifts is insufficient to prove the proposed mechanism; the authors must provide high-resolution Ti 2p and O 1s spectra to offer direct experimental evidence of coordination at the Ti-OH and -COOH sites.
6. (Line 214) For the FT-IR analysis in Fig. 2h, the authors attribute the subtle redshifts (2–3 cm⁻¹) of the Ti-OH and -COO⁻ peaks to the formation of ionic coordination bonds. However, such small shifts are often within the margin of instrumental resolution or baseline noise unless specific high-precision parameters are employed. To justify the physical significance of these minor wavenumber differences, the authors must provide detailed measurement parameters, including the spectral resolution (e.g., 1 or higher), the number of scans, and the peak-fitting methodology used to determine the exact peak positions. Furthermore, given the reliance on 2D-COS to monitor these interactions, it is crucial to demonstrate that the original 1D spectra were acquired with sufficient precision to support such a high-sensitivity correlation analysis."
7. (Line 245) In tensile property data, the authors report exceptional mechanical strength in GPa. However, as shown in Fig.2d and Supplementary Fig.6, the fibers possess a substantially microporous and layered structure, which presents significant challenges in accurately determining the cross-sectional area (S) and density (ρ). According to Supplementary

Note 1, the area (S) was measured from SEM images; yet, for such porous fibers, SEM-based geometric measurements can lead to an overestimation of the effective stress. The authors should clarify whether the 'apparent area' or 'effective material area' was used for GPa conversion. Furthermore, given that density is a critical parameter for porosity calculations (Equation S4), the specific methodology for determining the fiber density (e.g., via linear density or pycnometer) must be verified to ensure the reliability of the reported mechanical properties. Moreover, the tensile properties were evaluated using more than four samples for each fiber. While this provides preliminary insight, the number of tested specimens appears relatively limited to ensure robust statistical reliability, especially considering the inherent variability in fiber tensile testing.

8. (Line 251) The authors attribute the reduction in mechanical and electrical properties at 0.16 wt% CS loading to potential aggregation acting as stress concentrators. However, considering that the difference between 0.12 wt% and 0.16 wt% is extremely minute (only a 0.04 wt% increment), such a dramatic drop in performance—approximately 30% in both strength and conductivity—requires more rigorous validation. To support this claim, the authors should provide direct evidence of CS aggregation at this specific concentration through solution-phase analysis (e.g., DLS, Zeta potential, or Rheological characterization) or cite specific references that establish the critical stability limits of CMC-stabilized SWCNTs in high-concentration MXene dispersions. Without such evidence, the explanation remains speculative.

9. (Line 287) Regarding the unexpected decrease in electrical resistance after 50,000 cycles, the authors' attribution to 'structural reconfiguration' remains purely speculative without post-test structural characterization. To substantiate this claim, the authors should provide comparative WAXD data acquired after the cycling test. Specifically, a quantitative comparison of the Herman's orientation factor and the full-width at half-maximum (FWHM) of the (002) peak is required to demonstrate that cyclic mechanical loading indeed leads to enhanced alignment or superior stacking order of the MXene nanosheets. Without such evidence, it is difficult to distinguish whether the observed resistance change originates from intrinsic structural improvement or uncompensated geometric deformations of the fiber.

10. (Line 532) The synthesis and purification of MXene nanosheets involve multiple centrifugation steps (5000 rpm, 8000 rpm, and 3500 rpm) to obtain a 'homogeneous suspension'. Given that the final fiber performance is highly sensitive to the uniformity of the building blocks, the authors should provide quantitative data regarding the yield of single-layer MXene relative to the initial Ti_3AlC_2 powder. Furthermore, the claim of 'homogeneity' should be substantiated with statistical size distribution data (e.g., from DLS or large-scale AFM analysis) and Zeta potential measurements to demonstrate the dispersibility and colloidal stability of the MXene nanosheets before the spinning process. This information is crucial for assessing the scalability and reproducibility of the proposed molecular cord strategy.

11. (Line 544) Clearer specifications of key experimental details, such as the quantitative ratio between SWNT and CMC, as well as a more detailed description of the experimental procedures, would further enhance the clarity, transparency, and reproducibility of the manuscript.

12. (Line 549) The authors employed a hot-pressing temperature of 70 °C, which exceeds the boiling point of the residual methanol (~65 °C) used in the crosslinking bath. While this process successfully reduced the porosity from 45.8% to 21.4%, such rapid solvent evaporation above the boiling point could potentially induce internal vapor pressure, leading to microstructural defects or non-uniform pore distribution. The authors should clarify whether this side effect affected the structural integrity of the fibers and justify why 70 °C was selected over a more conservative temperature below the boiling point of methanol. Additionally, further discussion is needed on whether this thermal energy might lead to excessive annealing, influencing the mechanical flexibility and the stability of the ionic coordination knots.

Overall, these results significantly surpass the current benchmarks for MXene-based fibers. However, the aforementioned concerns—ranging from the rheological paradox of the spinning dope and the structural inconsistencies in orientation-porosity relations to the insufficiency of direct spectroscopic evidence for ionic coordination—must be rigorously addressed. Resolving these issues validate the proposed strengthening mechanism as a robust and reproducible paradigm in fiber science.

Reviewer #3

(Remarks to the Author)

This work addresses a fundamental and long-standing limitation of MXene fibers by directly resolving weak in-plane edge-to-edge connectivity between MXene nanosheets, rather than relying solely on interlayer reinforcement. Inspired by laced lamellar armor, the authors introduce a molecular cord bridging strategy which simultaneously enhances stress transfer, enables controlled slippage, and suppresses crack initiation. Comprehensive structural, spectroscopic, mechanical, fatigue, and simulation analyses convincingly validate the proposed in-plane and interlayer bridging mechanism. However, the following points need to be addressed before publication.

1. Although the achieved properties are impressive, the manuscript does not sufficiently foreground that the primary advance is the direct resolution of weak in-plane edge-to-edge connectivity between MXene nanosheets. Much of the Introduction frames the work as incremental interfacial strengthening, which risks obscuring the genuinely new conceptual contribution.

2. A more explicit, quantitative benchmarking against leading MXene, graphene, and carbon nanotube fibers, particularly considering additive content and processing severity is necessary to convincingly establish superiority.

3. The absence of thermal stability and temperature-dependent mechanical or electrical data is a notable omission, particularly given the proposed applications in functional and structural systems. Without this information, the operational envelope and robustness of the molecular cord-bridged architecture remain unclear.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have satisfactorily addressed the comments. The manuscript can now be published

Reviewer #2

(Remarks to the Author)

The manuscript has been revised extensively and it is in the near shape of publication in Nature Communications. However, it appears although the authors provided a number of data set, it has to be justified accordingly (i.e. comparison with the previously reported Mxene fiber properties and origin of improved toughness). As a minor point, there is typo to describe azimuthal angle (sometimes it is denoted as ϕ or Φ).

Reviewer #3

(Remarks to the Author)

The authors have addressed the reviewer comments in a satisfactory manner. I recommend publication, provided that the concerns raised by the other reviewers have also been adequately addressed.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The manuscript has been revised according to the comments, and it is recommended to be published in Nature Communications.

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Reply to the Reviewer#1:

Comments: Early studies by Wan et al. and Usman et al. (Science 374 (2021) 96–99; Nature Communications 13 (2022) 4564; 2D Materials 9 (2022) 044003) pioneered sequential in-plane and interlayer bridging of MXene nanosheets to enhance stress transfer, densification, and toughness. While their performance metrics were lower, these works established the core mechanistic principles of sequential reinforcement, including load sharing, delayed crack initiation, and energy dissipation. Many recent reports build on this same framework, often achieving incremental improvements rather than fundamentally new concepts. The authors should clearly articulate how the present molecular cord strategy advances beyond these foundational studies and justify its novelty in the context of established sequential bridging mechanisms.

REPLIES: Many thanks for the suggestions! We agree that these studies established important mechanistic principles for laminated architectures, including improved stress transfer, delayed crack initiation, and enhanced energy dissipation. At the same time, we respectfully note that the key scientific question and the core mechanical bottleneck addressed in our manuscript are different. **Our strategy is not intended as another incremental step in “sequential bridging.”** **Instead, it is a targeted solution to the MXene-specific in-plane edge-to-edge weakness,** which we identify as the primary source of stress concentration and premature failure in MXene fibers and films. In other words, the framework represented by the studies cited by the reviewer emphasizes sequential reinforcement more broadly, whereas our central contribution is to make edge-bridging the primary design axis and to demonstrate how controlling the in-plane edge junctions fundamentally changes the deformation and failure pathway.

Specifically, the “molecular cord” strategy advances beyond prior sequential bridging by introducing a continuous, long-range in-plane bridging network rather than relying on discrete or stepwise junction strengthening. We employ micrometer-long CMC-wrapped SWCNT cords that span multiple MXene sheets and act as load-bearing “laces” across sheet edges, while Mg^{2+} -mediated coordination creates robust crosslinking “knots” between the cords and MXene surface terminations. This architecture is crucial because it simultaneously achieves (i) effective stress redistribution across edge-to-edge discontinuities (the dominant weak links) and (ii) interlayer stabilization, but without over-constraining the interfaces. In contrast to approaches that mainly

densify or strengthen the laminate yet may suppress interlayer sliding, our cord–knot motif is designed to be strong-yet-dynamic, enabling sustained load transfer while preserving slidable interfaces for energy dissipation. **As a result, the toughening mechanism is not merely “stronger bonding” within an existing sequential-bridging paradigm, but a shift toward cord-governed crack-bridging and distributed deformation**, which we corroborate by multiscale evidence (e.g., shear strain redistribution and crack-bridging behavior consistent with cord pull-out and ductile fracture features, as supported by simulations and fracture morphology analyses).

Finally, our results demonstrate that this concept is not an incremental tuning of established sequential bridging, but a distinct structure–mechanism–property linkage achieved at ultralow additive content while maintaining multifunctionality: the cord network operates at only 0.12 wt% yet it enables a combination of ultrahigh strength and toughness together with high electrical conductivity and exceptional cyclic durability. To address the reviewer’s request more explicitly, we have revised the Introduction and Discussion in the revised manuscript to (i) clearly acknowledge Wan *et al.* and Usman *et al.* as pioneering sequential-bridging studies, and (ii) articulate more directly that **our novelty lies in elevating in-plane edge-bridging** to the central design principle via a continuous molecular-cord network with dynamic coordination knots, thereby resolving the MXene-specific edge-junction limitation rather than making a purely incremental improvement within the same sequential-bridging framework (Highlight on Pages 4 and 5).

Comments: Additionally, the method employed, including thermal densification, has been demonstrated previously (Nature Communications 13 (2022) 4564).

REPLIES: Thanks for the comments! In our present work, thermal densification is not presented as the primary innovation, nor is it sufficient to account for the observed strengthening and toughening. Instead, we use it as an adopted processing step to reduce residual porosity following our previously published work (*Nature Communications* 2025, 16, 9645), so that the intrinsic role of our molecular-cord-enabled edge-bridging can be evaluated under a comparable densified-state baseline. Importantly, the key advance of our method lies in introducing a continuous cord network

with dynamic coordination “knots” that specifically stabilizes in-plane edge-to-edge junctions and enables simultaneous efficient load transfer and energy dissipation—an effect that cannot be achieved by densification alone.

To avoid any ambiguity, we have revised the manuscript to explicitly acknowledge the prior use of thermal densification and clarify its role in our workflow as an auxiliary, non-novel processing step (Highlight on Page 25). We also emphasized that all relevant control samples were processed with the same densification protocol, and that the performance gains raised from the cord-mediated edge-bridging mechanism rather than from thermal densification itself.

Comments: Carbon nanotubes (CNTs) are well known for imparting high mechanical strength; however, it would be valuable to clarify whether the observed molecular cord bridging mechanism is intrinsically dependent on CNTs or whether other one-dimensional additives could achieve a similar strengthening effect. In many composite systems, the inclusion of CNTs is often considered a straightforward or expected route to mechanical reinforcement, which can obscure the specific contribution of the proposed mechanism. From a mechanistic perspective, the exceptionally high Young's modulus of CNTs may even be excessive, as it could partially constrain interfacial deformation and limit the full activation of the molecular cord bridging process. Additionally, the cost-effectiveness of CNTs as a reinforcement is a recurring concern in previous studies. This suggests that more compliant or chemically tunable one-dimensional additives may be equally, or potentially more, effective in enabling this mechanism. Clarifying this distinction would strengthen the mechanistic interpretation and broaden the design space for future material system.

REPLIES: Thank you for the suggestions! Our molecular-cord strategy requires (i) high strength and modulus to prevent failure under dynamic loading, (ii) a sufficiently high aspect ratio to span across multiple MXene sheets, and (iii) strong and stable interfacial interactions to enable effective load transfer while still permitting interfacial energy dissipation. SWCNTs were selected primarily because they combine excellent intrinsic mechanical properties with an ultrahigh aspect ratio, which makes them particularly suitable as long-range bridging backbones within the laminated MXene framework. However, SWCNTs alone cannot realize our mechanism. Pristine SWCNTs are difficult to individually disperse and tend to bundle/aggregate, which would prevent the

formation of a homogeneous, continuous bridging network. Moreover, the intrinsic interaction between SWCNTs and MXene is mainly weak van der Waals attraction, which is insufficient for efficient stress transfer and for stabilizing the edge-to-edge junctions that dominate failure. For these reasons, we do not rely on SWCNTs as a “straightforward filler,” but instead introduce CMC to wrap SWCNTs and form well-dispersed, micrometer-long cords, while simultaneously providing abundant functional groups that strengthen the interface with MXene (and further enable coordination crosslinking). Thus, the CMC-enabled dispersion and interfacial bonding are indispensable to activate the cord-bridging effect; without this chemical and structural design, SWCNT addition would not produce the same mechanism or performance.

Regarding the concern that the high modulus of SWCNTs might over-constrain interfacial deformation, **we note that our cords are introduced at an ultralow loading and behave as flexible, lacing-like bridges rather than rigid bars.** The cord network is designed to redistribute stress and bridge developing microcracks while still allowing the MXene interfaces to slide and dissipate energy. In practice, the very small SWCNT fraction and the cord-like configuration do not suppress interfacial deformation; instead, they provide localized load sharing and crack bridging without locking the layered structure. Finally, on cost-effectiveness, we emphasize that the CMC-SWCNT content is extremely low (0.12 wt% in our optimized formulation), so the overall cost impact is limited compared with conventional CNT-reinforced composites, while the performance gains arise from the architecture-enabled mechanism rather than from large CNT fractions.

More broadly, we agree with the reviewer that the mechanism should not be viewed as intrinsically CNT-exclusive. In principle, other one-dimensional additives with sufficient aspect ratio and appropriate surface chemistry could serve as alternative “cord backbones,” provided they can be uniformly dispersed and can establish strong yet non-brittle interfaces with MXene. We have provided further clarification on why CNTs were chosen in this study, and discuss the criteria for potential substitutes (e.g., dispersibility, and chemically tunable interfacial interactions) to broaden the future design space (Highlight on Page 7).

Comments: Previous studies have attributed the balanced mechanical properties and enhanced strength of MXene based composites not only to interflake connectivity but also to the intrinsic

anisotropy and alignment of the MXene sheets. In this context, it is important to clarify whether flake alignment contributes to the observed mechanical performance in the present system. The X ray scattering data provided in the Supporting Information do not indicate any improvement in alignment. Can the authors provide further clarification on this point?

REPLIES: Many thanks for the suggestions! We fully agree that flake anisotropy and alignment are well-established factors influencing the mechanical performance of MXene-based fibers and films, as demonstrated in the literature, including our previous studies (*Adv. Mater.* 2023, 35, 2209527; *Nat. Commun.* 2025, 16, 9645). As shown in **Supplementary Fig.9**, the neat MXene fiber already exhibits a high degree of alignment, reaching 0.91, which is among the higher values reported for MXene fibers (*Nat. Commun.* 2022, 13, 4564; *ACS Nano* 2021, 15, 7821-7832). Meanwhile, its tensile strength reaches 235 MPa, outperforming most previously reported neat MXene fibers (*Adv. Mater.* 2023, 35, 2209527; *Nat. Commun.* 2020, 11, 2825). We believe that this excellent performance mainly originates from our optimized ink preparation, wet-spinning, and post-treatment processes. These results also indicate that, in the present system, the highly aligned nanosheet arrangement indeed provides an important structural basis for the good mechanical performance.

On this basis, we further compared the alignment characteristics of pristine MXene fibers and CS-MXene fibers. The results show that the two samples possess essentially the same degree of alignment, indicating that the introduction of CS does not further improve the alignment of MXene nanosheets. In other words, although high alignment is an important prerequisite for achieving good mechanical properties in this system, the additional mechanical enhancement observed from pristine MXene fibers to CS-MXene fibers cannot be attributed to improved alignment. We therefore conclude that the performance gain mainly originates from the “molecular cord” bridging mechanism introduced by CS, which enhances load transfer and cooperative deformation between adjacent nanosheets, thereby enabling the simultaneous improvement of strength and toughness.

To address the reviewer’s concern, we have clarified the role of flake alignment in the revised manuscript and provide a clearer comparison of the alignment and ordering between CS–MXene and C–MXene in the Supporting Information. We will further **emphasize that the improved mechanical performance arises from a synergistic effect of alignment improvement and edge bridging, with edge bridging remaining the dominant mechanism** (**Highlight on Page 8**).

Comments: The lack of a clearly defined target application limits the broader appeal of the work. While the study demonstrates record mechanical and electrical performance, it is not immediately clear which specific type of supercapacitor or practical use is intended. For example, is the system designed for supercapacitors that undergo repeated bending or for applications that primarily require high tensile strength? Clarifying the intended application and explaining how the molecular cord strategy addresses a specific functional or practical need would strengthen the manuscript. Providing this context would help position the study within a purposeful materials design framework, rather than appearing solely as a combination of high-performance components aimed at surpassing existing benchmarks.

REPLIES: Thanks for this insightful suggestion! We agree that clarifying the intended application context will strengthen the positioning and broader appeal of the work. In the revised manuscript, we have more clearly articulated that the combination of high strength, high toughness, and high electrical conductivity demonstrated here is particularly relevant for structural and wearable fiber-shaped energy storage systems, such as fiber-shaped supercapacitors and conductive yarns integrated into textiles. In practical scenarios, such fibers are not subjected to a single loading mode. Instead, they must withstand repeated bending, twisting, knotting, and weaving, while also tolerating tensile loads during fabrication and service. **Therefore, our design is not aimed solely at systems requiring high tensile strength, nor exclusively at devices undergoing bending, but rather at fiber electrodes that operate under coupled mechanical deformation and electrochemical function.** The molecular cord strategy directly addresses this practical need by enhancing crack resistance, fatigue tolerance, and load redistribution while preserving continuous conductive pathways—thereby improving structural reliability under realistic mechanical conditions.

At the same time, we would like to clarify that the primary objective of this study is to establish and validate a mechanism-driven reinforcement and toughening strategy centered on edge-bridging, rather than to present a fully integrated device demonstration. Many studies on MXene and, more broadly, functional materials based on two-dimensional materials primarily focus on elucidating the structure–mechanism–property relationships underlying strengthening and

toughening, while treating practical applications as the overall background and driving motivation for the research. Many high-impact studies on MXene and other 2D structural conductors likewise focus on establishing structure–mechanism–property relationships for strengthening/toughening, with practical applications mentioned only as general motivation rather than being demonstrated as a specific end-use prototype. For example, Wan *et al.* reported a sequential-bridging strategy to densify MXene films, achieving markedly improved tensile strength and toughness while maintaining high electrical conductivity and strong EMI-shielding performance (*Science*, 2021, 374, 96-99). The main contribution of that work was to elucidate the mechanism of bridging-induced densification and establish the corresponding structure–property relationship, rather than to focus on specific application scenarios or device-level demonstrations. In addition, Li *et al.* reported a nanoconfined polymerization strategy to produce tough, near-zero-hysteresis gels with significantly enhanced crack-propagation resistance under large deformation, attributing the improved performance to nanochannel confinement and strong hydrogen-bond interactions that alleviate stress concentration at crack tips. The study primarily focused on elucidating the reinforcement and toughening mechanism and demonstrating its generality across different gel systems, rather than defining a specific target application or presenting a device-level demonstration (*Nat. Mater.* 2024, 23, 131–138). Consistent with this established materials-design paradigm, our manuscript is positioned to resolve a fundamental MXene-specific failure origin (in-plane edge-to-edge weakness) and to demonstrate that the molecular-cord architecture reshapes deformation and fracture pathways, thereby providing a robust multifunctional fiber platform that can be readily adopted in various fiber-based energy-storage and wearable-electronics configurations. In response to the reviewer’s comment, we have revised the Introduction and Conclusion in the revised manuscript to more explicitly connect the molecular cord strategy with specific functional demands in fiber-shaped supercapacitors, wearable electronics, and structurally reliable conductive textiles (Highlight on Pages 5 and 18).

Reply to the Reviewer#2:

Comments: This study presents a paradigm-shifting strategy to fabricate high-performance MXene-based fibers by addressing the fundamental bottleneck of weak in-plane edge-to-edge interactions between MXene nanosheets. Inspired by the cord-bound architecture of ancient lamellar armor, the authors employed carboxymethyl cellulose-wrapped single-walled carbon nanotubes (CS) as "molecular cords" to bridge adjacent MXene flakes via robust Mg^{2+} -mediated ionic coordination bonds. Through a synergistic process of wet-spinning, ionic crosslinking, and controlled hot-pressing, the resulting fibers (containing only 0.12 wt% CS) achieved a remarkable tensile strength of 1.48 ± 0.06 GPa, an unprecedented toughness of 131.0 ± 8.7 MJ m⁻³, and an exceptional electrical conductivity exceeding 10,400 S cm⁻¹. These are great achievements in MXene based fiber research.

REPLIES: Thanks a lot for the positive comments!

Comments: (Line 99) The manuscript employs carboxymethyl cellulose (CMC) to form strong ionic coordination bonds; however, the rationale for selecting CMC over other ion-rich or carboxylate-containing polymers is not sufficiently discussed. A brief discussion in the Introduction part highlighting the advantages of CMC, such as its coordination capability, interfacial compatibility, and resulting performance benefits, would further clarify why CMC is particularly well suited for this system.

REPLIES: We sincerely appreciate the reviewer's valuable suggestion. In the revised manuscript, we have added a brief discussion in the Introduction to clarify the rationale for selecting carboxymethyl cellulose (CMC) as the molecular cord (Highlight on Pages 5 and 7). **CMC offers three key advantages that make it particularly well suited for this system. First, as a highly abundant anionic polyelectrolyte, CMC exhibits excellent water solubility, processability, and strong coordination capability, which are essential for constructing stable ionic crosslinking networks. Second, the abundant carboxyl and hydroxyl groups along the CMC chains enable the formation of strong coordination bonds and hydrogen bonds with MXene nanosheets,**

ensuring robust interfacial anchoring. Third, **CMC can efficiently wrap around carbon nanotubes through hydrophobic and hydrogen-bonding interactions, promoting their uniform dispersion and effective integration into the MXene matrix.** These combined features—coordination capability, interfacial compatibility, and dispersing efficiency—render CMC an ideal molecular cord for synergistically bridging MXene and CNTs in the spinning dope system. Additionally, ongoing work in our group is exploring these alternative carboxylate-containing polymer systems.

Comments: (Line 152) The authors state that CMC wraps the single-walled carbon nanotubes (SWNTs); however, the specific interactions responsible for this wrapping are not clearly described. While SEM images of fractured fiber cross-sections suggest effective CNT dispersion and mechanical bridging, these observations do not directly evidence CMC wrapping of SWNTs, particularly in the solution state. Further mechanistic discussion and supporting experimental evidence (direct or indirect) are necessary to substantiate the claimed CMC wrapping.

REPLIES: Thank you for the suggestions! To demonstrate the functionalization and wrapping of SWCNTs by CMC, we characterized the aqueous dispersions of pristine SWCNTs and CMC-SWCNT through macroscopic dispersion observation, Zeta potential measurement, and HRTEM imaging (Highlight on Page 7). As shown in Supplementary Fig.1, pristine SWCNTs are difficult to disperse in water, whereas CMC functionalization enables effective dispersion of SWCNTs, consistent with previous reports (*Constr. Build. Mater.*, 2023, 377, 131104; *Powder Technol.*, 2022, 404, 117505; *J. Phys. Chem. C* 2016, 120, 22694-22701). This improvement can be attributed to the adsorption of CMC chains onto the SWCNT surface through noncovalent interactions, including hydrophobic interactions and possible π - π interactions, which enhance interfacial affinity and steric stabilization (*Adv. Fiber Mater.* 2024, 6, 758-771). Zeta potential analysis further reveals that the surface charge of SWCNTs decreases significantly from -5.8 mV to -50.9 mV after CMC functionalization, indicating successful wrapping of CMC around the SWCNTs (Supplementary Fig.1).

More direct structural evidence is provided by HRTEM imaging (Supplementary Fig.2). Compared with pristine SWCNTs, which exhibit clean and well-defined boundaries, the CMC-

SWCNTs display roughened edges and a thin amorphous coating layer surrounding the nanotube walls, which can be attributed to the adsorption of CMC chains. These results from dispersion behavior, surface charge analysis, and microscopic imaging collectively support the effective wrapping of CMC around SWCNTs in the aqueous system.

Comments: (Line 163) The authors report that the viscosity of the MXene-based spinning dopes exceeds 1000 Pa·s at low shear rates. In conventional wet-spinning, the viscosity of dopes is typically maintained below 100 Pa·s to ensure stable extrusion and prevent structural defects. Given this exceptionally high viscosity, the authors should clarify how they achieved stable fiber formation through a 26 G needle at a rate of 5 mL·h⁻¹ without encountering issues such as high back-pressure or stable extrusion. It is recommended to provide capillary rheometry data or jet profile images at the nozzle tip to prove the flow stability and shear-thinning behavior under actual spinning conditions.

REPLIES: We sincerely appreciate the reviewer's insightful suggestion regarding the high viscosity of the spinning dopes. To evaluate the extrusion stability under practical spinning conditions, we estimated the apparent wall shear rate at the needle using a circular tube laminar flow model. The shear rate can be expressed as (Equation S3, Supplementary Note 1):

$$\gamma = \frac{4Q}{\pi R^3}$$

where Q is the extrusion rate (5 mL·h⁻¹) and R is the inner radius of the needle (220 μm). Based on this model, the apparent wall shear rate is calculated to be approximately 166.2 s⁻¹.

Importantly, although the dope exhibits a high viscosity (>1000 Pa·s) at low shear rates, it shows pronounced shear-thinning behavior. As shown in Supplementary Fig.5, the viscosity decreases to below 100 Pa·s at the estimated shear rate (~166 s⁻¹), which falls within the typical range for conventional wet-spinning processes. This indicates that the high low-shear viscosity does not hinder extrusion under actual spinning conditions. Furthermore, we recorded the extrusion behavior at the needle tip. As shown in the Supplementary Movie 1, the spinning dope forms a continuous, uniform jet with a smooth surface and no observable fluctuations, providing direct evidence of flow stability under actual spinning conditions. Collectively, these results confirm the

favorable shear-thinning behavior and extrusion stability of the spinning dope. Additionally, we have also supplemented the discussion in the main manuscript (Highlight on Page 7, Supplementary Movie 1, and Supplementary Note 1).

Comments: (Line 178) Regarding the structural characteristics shown in Supplementary Figures 7 and 8, the authors report that C-MXene fibers exhibit a more densified structure with lower porosity compared to neat MXene fibers. However, a significant discrepancy arises as the orientation factor of C-MXene (0.80) is substantially lower than that of neat MXene (0.91), despite its higher compactness. This inverse correlation suggests that the inclusion of CMC polymer chains alone may induce steric hindrance or disrupt the nematic liquid crystalline order during the spinning process. The authors should provide a clear physical explanation for this complex trend of orientation in C-MXene and further elucidate how the incorporation of CS (SWCNTs) effectively restores the alignment to 0.91 in the CS-MXene system.

REPLIES: We sincerely thank the reviewer for this insightful comment. In the revised manuscript, we have provided additional discussion and supporting experimental results to explain this structural evolution. First, the enhanced compactness of the C-MXene fibers mainly arises from the role of CMC as a macromolecular binder during wet spinning and subsequent drying. CMC is a flexible polymer that typically adopts an isotropic random-coil conformation in solution. During fiber formation and solvent evaporation, CMC chains interact with MXene flakes through hydrogen bonding and interfacial interactions, which helps transfer capillary contraction stress to the stacked sheet network and fill inter-flake voids (*Science* 2021, 374, 96-99). As a result, the C-MXene fibers exhibit a denser structure with lower porosity. **However, higher compactness does not necessarily correspond to higher orientation.**

The orientation factor reflects the degree of long-range ordered alignment of MXene flakes along the fiber axis, and its determining mechanism is not entirely the same as the densification process. In the neat MXene dispersion, rigid 2D nanosheets form a highly ordered nematic liquid crystalline (LC) phase and align thoroughly along the flow direction under the shear flow field of wet spinning, yielding a high orientation factor (0.91). However, upon the introduction of CMC, its random-coil chain conformation and chain entanglement exert significant steric hindrance

effects on the parallel sliding, rotation, and rearrangement of MXene flakes during spinning. This disrupts the intrinsic order of the LC phase and weakens the synergistic alignment of the flakes along the macroscopic axis (*Macromolecules* 1978, 11, 1138-1141). Consequently, although the C-MXene fiber is denser, its orientation factor decreases from 0.91 to 0.80.

This behavior is consistent with polymer/2D material composite spinning systems. While the polymer component enhances interfacial bonding and promotes densification, its random-chain conformation and entanglement can introduce steric hindrance that interferes with the cooperative sliding and long-range ordered alignment of 2D flakes. In the absence of additional orientation-inducing mechanisms, such effects may partially reduce the orientational order along the fiber axis. In similar systems, post-drawing is often required to restore alignment and achieve higher structural order (*ACS Nano* 2021, 15, 13055-13064). These further highlight that densification and orientation enhancement do not necessarily occur simultaneously in 2D flake fiber systems.

To further verify this explanation, we prepared and characterized C-MXene fibers with a lower CMC content (CMC: 0.06%). WAXS results show that its orientation factor increased to 0.85, higher than that of the sample with 0.12% CMC (Supplementary Fig.10). Concurrently, we supplemented polarized optical microscopy (POM) images of the C-MXene spinning dope (Supplementary Fig.4). Compared with the neat MXene dispersion, the system exhibited a weaker liquid crystalline texture and fewer anisotropic domains after the addition of CMC, demonstrating that the introduction of CMC indeed perturbs the LC ordered alignment of MXene nanosheets. The above results indicate that while CMC promotes fiber densification, it also partially disrupts the orientational order of the MXene flakes due to the steric hindrance and chain entanglement effects of its macromolecular chains.

Regarding the recovery of the orientation factor to 0.91 in the CS-MXene fibers, we attribute this to the strong alignment tendency of SWCNTs in the shear flow field and their synergistic templating effect within the composite system. Because of their rigid one-dimensional structure, high aspect ratio, and strong flow responsiveness, SWCNTs can more readily align along the fiber axis during wet spinning. Once aligned, they likely serve as rigid anisotropic scaffolds that help guide the surrounding MXene flakes toward a more parallel arrangement through van der Waals interactions and interfacial coupling (*Adv. Mater.* 2024, 36, 2306129; *Sci. China: Technol. Sci.* 2019, 62, 1545). In this way, SWCNTs not only improve structural connectivity, but also help compensate for the orientational disruption caused by the CMC chains, thereby restoring the

overall alignment of the CS-MXene system to a level comparable to that of neat MXene fibers.

Based on the above analysis, we believe that the “high compactness but relatively low orientation” observed in the C-MXene fibers is not contradictory, but rather reflects the difference between two distinct structural evolution mechanisms: densification is mainly governed by CMC-assisted interlayer contraction and pore filling, whereas orientation depends more strongly on liquid-crystalline order and the cooperative rearrangement of flakes under flow. The incorporation of SWCNTs helps re-establish anisotropic alignment in the composite system, thus recovering the orientation factor. The corresponding discussion has been added to the revised manuscript (Highlight on Page 8 and Supplementary Note 4).

Comments: (Line 212) Regarding the XPS analysis in Fig. 2g, the absence of Mg 1s and survey spectra is a critical omission, as these are essential to verify the successful incorporation and chemical state of the Mg^{2+} crosslinkers. Furthermore, relying solely on C 1s shifts is insufficient to prove the proposed mechanism; the authors must provide high-resolution Ti 2p and O 1s spectra to offer direct experimental evidence of coordination at the Ti-OH and -COOH sites.

REPLIES: We sincerely appreciate the reviewer’s critical comment regarding the XPS analysis. W

We have performed additional XPS measurements. First, the survey spectra before and after Mg^{2+} crosslinking and high-resolution Mg 1s spectra of the CS-MXene fiber provided in **Supplementary Fig.13** in the revised SI, which confirms the successful incorporation of Mg^{2+} into the fiber structure. Furthermore, to provide direct experimental evidence of coordination bonding, we collected high-resolution Ti 2p and O 1s spectra (**Supplementary Fig.14**). Notably, the C-Ti-O peak in both the Ti 2p and O 1s spectra exhibits a distinct shift toward lower binding energy after Mg^{2+} crosslinking. This shift indicates redistribution of electron density the Ti-OH and -COOH sites upon coordination with Mg^{2+} , providing direct evidence of ionic bond formation at the interfaces between MXene and CMC. Together with the previously reported C 1s analysis, these newly acquired data collectively and robustly support the proposed coordination mechanism involving Mg^{2+} as the ionic crosslinker. We have also supplemented the discussion in the revised manuscript (**Highlight on Page 10**).

Comments: (Line 214) For the FT-IR analysis in Fig. 2h, the authors attribute the subtle redshifts (2–3 cm⁻¹) of the Ti-OH and -COO⁻ peaks to the formation of ionic coordination bonds. However, such small shifts are often within the margin of instrumental resolution or baseline noise unless specific high-precision parameters are employed. To justify the physical significance of these minor wavenumber differences, the authors must provide detailed measurement parameters, including the spectral resolution (e.g., 1 or higher), the number of scans, and the peak-fitting methodology used to determine the exact peak positions. Furthermore, given the reliance on 2D-COS to monitor these interactions, it is crucial to demonstrate that the original 1D spectra were acquired with sufficient precision to support such a high-sensitivity correlation analysis."

REPLIES: We sincerely appreciate the reviewer's valuable suggestion regarding the FT-IR analysis. To ensure the accuracy and reliability of the FT-IR data, we repeated the measurements using the Thermo Scientific Nicolet IS50 spectrometer with higher precision parameters: 32 scans and a spectral resolution of 1 cm⁻¹. The results obtained under these conditions are consistent with the original data. Notably, the observed peak shifts (2–9 cm⁻¹) are greater than the instrumental resolution, indicating that these changes are experimentally distinguishable rather than arising from baseline noise. Since the measured infrared curves have a high signal-to-noise ratio and the peaks of the main functional groups are simple in shape, we obtained the peak positions by reading the minimum values of the peaks from the raw data curves.

As shown in Fig. 2h, the Ti-OH stretching vibration shifts progressively from 3434.6 cm⁻¹ (neat MXene fibers) to 3432.2 cm⁻¹ and 3425.4 cm⁻¹ for CS-MXene fibers before and after crosslinking, respectively. Meanwhile, the -COO⁻ peak of CMC shifts from 1620.4 cm⁻¹ to 1618.4 cm⁻¹ upon Mg²⁺ coordination. These systematic shifts are consistent with coordination interactions involving Mg²⁺. Building on these high-quality spectra, we performed two-dimensional correlation spectroscopy (2D-COS) analysis. As shown in Fig. 2i, the 2D-COS map exhibits strong autopeaks at 3425 cm⁻¹ (Ti-OH) and 1618 cm⁻¹ (-COO⁻) in the range of 1500–3600 cm⁻¹, highlighting the pronounced sensitivity of these functional groups to Mg²⁺ binding. These data have been updated accordingly in the revised manuscript (Highlight in Figure 2h and 2i on Page 9 and 10).

Comments: (Line 245) In tensile property data, the authors report exceptional mechanical strength in GPa. However, as shown in Fig.2d and Supplementary Fig.6, the fibers possess a substantially microporous and layered structure, which presents significant challenges in accurately determining the cross-sectional area (S) and density (ρ). According to Supplementary Note 1, the area (S) was measured from SEM images; yet, for such porous fibers, SEM-based geometric measurements can lead to an overestimation of the effective stress. The authors should clarify whether the 'apparent area' or 'effective material area' was used for GPa conversion. Furthermore, given that density is a critical parameter for porosity calculations (Equation S4), the specific methodology for determining the fiber density (e.g., via linear density or pycnometer) must be verified to ensure the reliability of the reported mechanical properties. Moreover, the tensile properties were evaluated using more than four samples for each fiber. While this provides preliminary insight, the number of tested specimens appears relatively limited to ensure robust statistical reliability, especially considering the inherent variability in fiber tensile testing.

REPLIES: We sincerely appreciate the reviewer's insightful comments regarding the mechanical property characterization, particularly concerning the cross-sectional area determination, density measurement, and statistical reliability. In response, we have conducted additional analyses and clarifications as detailed below. First, for tensile strength calculation, we used the effective load-bearing cross-sectional area rather than the apparent geometric area. Because the fibers contain internal pores, using the apparent cross-sectional area does indeed lead to inaccurate stress calculations. Therefore, cross-sectional SEM images were analyzed using ImageJ, and the internal void regions were excluded to obtain the effective material area for stress conversion (*ACS Cent. Sci.* 2020, 6, 254-265; *Science* 2024, 384, 1318-1323). For each sample condition, at least four independent fiber cross-sections were analyzed to reduce measurement uncertainty. This procedure has now been explicitly clarified in **Supplementary Note 1**.

Second, the fiber density was determined using the linear density method. Specifically, fibers of known length were weighed using a high-precision microbalance (precision: 0.01 mg), and the linear density was calculated. The density was then derived by combining the linear density with the geometric cross-sectional area obtained from SEM images. This approach is widely adopted for fiber specimens and is consistent with the porosity calculation formula (Equation S5). We have now explicitly stated this methodology in the revised Supplementary Information (**Supplementary**

Note 1) to ensure clarity and reproducibility.

Finally, we thank the reviewer for their attention to statistical reliability. In our work, the measured mechanical data exhibited good consistency with relatively low standard deviations, indicating good reproducibility of the experimental results. In fact, during our experimental investigation, tensile tests were repeatedly conducted on samples prepared from different batches to further verify the reproducibility and statistical reliability of the mechanical performance. In the field of fiber tensile testing, the use of four parallel specimens per sample condition is widely accepted (*Adv. Mater.* 2023, 35, 2305807; *Science* 2024, 384, 1318-1323; *Nat. Commun.* 2026, 17, 1277). The mechanical data presented in our study exhibited good consistency with low standard deviations, confirming the reproducibility of the measurements.

Comments: (Line 251) The authors attribute the reduction in mechanical and electrical properties at 0.16 wt% CS loading to potential aggregation acting as stress concentrators. However, considering that the difference between 0.12 wt% and 0.16 wt% is extremely minute (only a 0.04 wt% increment), such a dramatic drop in performance—approximately 30% in both strength and conductivity—requires more rigorous validation. To support this claim, the authors should provide direct evidence of CS aggregation at this specific concentration through solution-phase analysis (e.g., DLS, Zeta potential, or Rheological characterization) or cite specific references that establish the critical stability limits of CMC-stabilized SWCNTs in high-concentration MXene dispersions. Without such evidence, the explanation remains speculative.

REPLIES: We sincerely appreciate the reviewer's insightful comment regarding the observed performance drop at 0.16 wt% CS loading. First, we note that during the preparation of the spinning dope, the CS-MXene dispersion at 0.16 wt% CS loading exhibited visible small aggregates (Supplementary Fig.18), a phenomenon that becomes even more pronounced at higher concentrations. Given the stringent dispersion requirements for wet spinning, the presence of localized aggregates can introduce pore defects during fiber formation, thereby leading to a significant deterioration in both mechanical and electrical properties. We would like to clarify that although the absolute increase in CS content from 0.12 wt% to 0.16 wt% appears small, it corresponds to a ~33% relative increase, which can significantly affect the dispersion stability of

high-aspect-ratio carbon nanotubes. For such nanomaterials with high aspect ratio, even slight increases in concentration can lead to a sharp rise in effective volume fraction and interparticle interactions, thereby promoting aggregation (*Mater. Res. Express*, 2018, 5, 015044).

To further provide evidence of this aggregation behavior, we characterized the liquid crystalline properties of the spinning dopes using polarized optical microscopy (POM). As shown in **Supplementary Fig.4**, compared with the CS-MXene-0.12 wt% dispersion, the CS-MXene-0.16 wt% system exhibits significantly weakened and more heterogeneous liquid crystalline textures, indicating reduced dispersion uniformity and disruption of ordered alignment. Thus, these observations suggest that the system approaches a dispersion stability threshold near 0.16 wt% CS loading, where aggregation begins to occur and disrupts both structural integrity and conductive pathways. We have clarified this point in the revised manuscript (**Highlight on Page 12**).

Comments: (Line 287) Regarding the unexpected decrease in electrical resistance after 50,000 cycles, the authors' attribution to 'structural reconfiguration' remains purely speculative without post-test structural characterization. To substantiate this claim, the authors should provide comparative WAXD data acquired after the cycling test. Specifically, a quantitative comparison of the Herman's orientation factor and the full-width at half-maximum (FWHM) of the (002) peak is required to demonstrate that cyclic mechanical loading indeed leads to enhanced alignment or superior stacking order of the MXene nanosheets. Without such evidence, it is difficult to distinguish whether the observed resistance change originates from intrinsic structural improvement or uncompensated geometric deformations of the fiber.

REPLIES: We sincerely thank the reviewer for this valuable suggestion regarding the origin of the resistance decrease after cyclic loading. To address this concern, we performed additional WAXS characterization on the fibers after cyclic stretching to probe their structural evolution. As shown in **Supplementary Fig.26**, the fibers exhibit higher orientation factor (~ 0.92) and a narrower FWHM of 21.61° after 10,000 stretch-release cycles, comparable to that of the pristine state, indicating that the MXene nanosheets remain highly aligned even after repeated deformation. This suggests that the internal alignment is well preserved under cyclic loading. These results indicate that cyclic mechanical loading does not deteriorate the structural order of the fibers and may

facilitate subtle structural rearrangements that improve inter-sheet contact. Such structural stabilization and possible local reorganization could contribute to the observed decrease in electrical resistance.

At the same time, we acknowledge that the resistance change may also be partially influenced by geometric factors (e.g., slight densification or strain-induced dimensional changes), which cannot be completely excluded at this stage. Therefore, we have revised the manuscript to adopt a more cautious interpretation, stating that the resistance decrease is likely associated with stress-induced structural rearrangement and preserved nanosheet alignment. The additional discussion has been added to the revised manuscript (Highlight on Page 13).

Comments: (Line 532) The synthesis and purification of MXene nanosheets involve multiple centrifugation steps (5000 rpm, 8000 rpm, and 3500 rpm) to obtain a 'homogeneous suspension'. Given that the final fiber performance is highly sensitive to the uniformity of the building blocks, the authors should provide quantitative data regarding the yield of single-layer MXene relative to the initial Ti_3AlC_2 powder. Furthermore, the claim of 'homogeneity' should be substantiated with statistical size distribution data (e.g., from DLS or large-scale AFM analysis) and Zeta potential measurements to demonstrate the dispersibility and colloidal stability of the MXene nanosheets before the spinning process. This information is crucial for assessing the scalability and reproducibility of the proposed molecular cord strategy.

REPLIES: We sincerely appreciate the reviewer's valuable comments regarding the quality of MXene nanosheets. In response, we have provided additional data to quantitatively assess the yield, uniformity, and dispersibility of the MXene building blocks. The yield of single-layer MXene relative to the initial Ti_3AlC_2 powder is approximately 30–35%. Large-scale AFM analysis (Supplementary Fig.3a-3c) reveals that the MXene nanosheets exhibit a uniform lateral size distribution, with an average size of $1.9 \pm 0.6 \mu m$. Furthermore, the dispersibility and colloidal stability of the MXene suspension were evaluated by Zeta potential measurements. As shown in Supplementary Fig.3d, the Zeta potential of the MXene dispersion is -30.8 mV, indicating good dispersion stability. Additionally, we have also supplemented the discussion in the main manuscript (Highlight on Pages 7 and 23).

Comments: (Line 544) Clearer specifications of key experimental details, such as the quantitative ratio between SWNT and CMC, as well as a more detailed description of the experimental procedures, would further enhance the clarity, transparency, and reproducibility of the manuscript.

REPLIES: We sincerely appreciate the reviewer's valuable comments. We deeply appreciate the reviewer's constructive feedback regarding the completeness of our experimental details. We completely agree that clarity and reproducibility are paramount. However, we would like to clarify that the SWCNTs used in our study were derived from a highly stable, commercially available single-walled carbon nanotube aqueous dispersion (TUBALL™, SWCNTs, Batch 01RW02.N1, manufactured by OCSiAl). In this commercial formulation, CMC is pre-added by the manufacturer as a necessary surfactant/dispersant (the mass ratio of SWCNT to CMC is approximately 1:1.5) to ensure the uniform and stable suspension of the highly entangled SWCNTs in water. This detail has already been clarified in the "Methods" of the manuscript (Highlight on Page 24).

Comments: (Line 549) The authors employed a hot-pressing temperature of 70 °C, which exceeds the boiling point of the residual methanol (~65 °C) used in the crosslinking bath. While this process successfully reduced the porosity from 45.8% to 21.4%, such rapid solvent evaporation above the boiling point could potentially induce internal vapor pressure, leading to microstructural defects or non-uniform pore distribution. The authors should clarify whether this side effect affected the structural integrity of the fibers and justify why 70 °C was selected over a more conservative temperature below the boiling point of methanol. Additionally, further discussion is needed on whether this thermal energy might lead to excessive annealing, influencing the mechanical flexibility and the stability of the ionic coordination knots.

REPLIES: We sincerely appreciate the reviewer's valuable comment. The design of the hot-pressing parameters is based on our previous work (*Adv. Mater.* 2023, 35, 2209527; *Nat. Commun.* 2025, 16, 9645), in which a temperature of 70 °C was selected to more effectively remove the interlayer solvent and avoid additional pore defects during subsequent vacuum drying. Although

70 °C is slightly above the boiling point of methanol, the application of a high pressure of 11 kPa ensures structural compactness. To more rigorously evaluate the influence of the hot-pressing temperature on the microstructure, mechanical properties, and stability of coordination bonds in the fibers, we prepared CS-MXene fibers using a hot-pressing temperature of 60 °C and conducted a series of comparative tests (Supplementary Fig. 22 and Fig. 23).

The microstructures of the fibers were characterized by XRD and SEM. As shown in Supplementary Fig.22, the fibers processed at the two different temperatures exhibit comparable interlayer spacing, porosity, and overall microstructure. XPS and FT-IR analyses reveal nearly identical peak positions and chemical states for the fibers obtained under both conditions, confirming the stability of ionic coordination knots at the hot-pressing temperature of 70 °C. Moreover, the stress–strain curves of the two types of fibers show no significant difference in mechanical properties, indicating that the slightly elevated temperature does not compromise the mechanical flexibility or structural integrity of the fibers. These results collectively demonstrate that the hot-pressing temperature of 70 °C effectively reduces porosity without introducing microstructural defects or destabilizing the coordination bonds, thereby supporting its suitability for fabricating dense and structurally robust fibers. Additionally, we have also supplemented the discussion in the main manuscript (Highlight on Page 12).

Reply to the Reviewer#3:

Comments: This work addresses a fundamental and long-standing limitation of MXene fibers by directly resolving weak in-plane edge-to-edge connectivity between MXene nanosheets, rather than relying solely on interlayer reinforcement. Inspired by laced lamellar armor, the authors introduce a molecular cord bridging strategy which simultaneously enhances stress transfer, enables controlled slippage, and suppresses crack initiation. Comprehensive structural, spectroscopic, mechanical, fatigue, and simulation analyses convincingly validate the proposed in-plane and interlayer bridging mechanism.

REPLIES: Thank you very much for the positive comments!

Comments: Although the achieved properties are impressive, the manuscript does not sufficiently foreground that the primary advance is the direct resolution of weak in-plane edge-to-edge connectivity between MXene nanosheets. Much of the Introduction frames the work as incremental interfacial strengthening, which risks obscuring the genuinely new conceptual contribution.

REPLIES: We thank the reviewer for this insightful comment! We fully agree that the key advance of this work is the direct resolution of weak in-plane edge-to-edge connectivity between MXene nanosheets, rather than a simple incremental improvement in interfacial strengthening strategies. **In the revised manuscript, we have clarified that the central novelty of this work lies in identifying in-plane edge-junction weakness as a critical mechanical bottleneck in MXene fibers and addressing it through the molecular cord bridging strategy.** We have revised the Introduction and Discussion accordingly to better highlight this conceptual contribution and to avoid framing the work as merely incremental interfacial reinforcement. Specifically, the central challenge targeted in this work is the intrinsic in-plane edge-junction weakness of MXene assemblies, which we identify as a major origin of stress concentration and premature mechanical failure. Unlike conventional strategies that mainly focus on reinforcing interlayer interactions or improving overall interfacial adhesion, our molecular cord design specifically targets the edge-to-edge discontinuities between adjacent MXene nanosheets. By doing so, it establishes a continuous load-transfer pathway across in-plane weak junctions while simultaneously preserving the

dynamic interlayer sliding needed for energy dissipation. We believe this distinction is essential, because it shifts the design logic from general “interfacial strengthening” to the more specific and conceptually important regulation of in-plane edge connectivity (Highlight on Pages 3 and 4).

Comments: A more explicit, quantitative benchmarking against leading MXene, graphene, and carbon nanotube fibers, particularly considering additive content and processing severity is necessary to convincingly establish superiority.

REPLIES: Thanks for this valuable suggestion! We agree that a more explicit and quantitative comparison with state-of-the-art MXene, graphene, and carbon nanotube fibers is important for more convincingly establishing the significance of our work. Therefore, we have revised the table and added a comparative analysis of ink concentration/additive content, coagulation bath, and post-treatment/processing conditions. We believe this revised analysis provides a clearer and more quantitative basis for evaluating the advantages and limitations of our approach relative to leading MXene-, graphene-, and carbon nanotube-based fibers. The relevant additions can be found in Supplementary Tables 1 and 2.

Comments: The absence of thermal stability and temperature-dependent mechanical or electrical data is a notable omission, particularly given the proposed applications in functional and structural systems. Without this information, the operational envelope and robustness of the molecular cord-bridged architecture remain unclear.

REPLIES: Thank you for your insightful questions regarding the thermal stability and temperature-dependent performance of the proposed molecular cord-bridged architecture, which is indeed critical for evaluating its potential in functional and structural systems. In response to this comment, we have conducted additional experiments to assess the thermal stability and temperature-dependent mechanical and electrical properties of the fibers. As shown in the newly added thermogravimetric analysis (TGA) results (Supplementary Fig. 25a), the fibers exhibit excellent thermal stability within the temperature range from ambient conditions up to 150 °C,

with no significant weight loss or decomposition was observed. This indicates that the molecular cord-bridged architecture remains intact under moderate thermal exposure.

We further evaluated the mechanical properties at 0 °C, 25 °C, 50 °C, and 100 °C. The stress-strain curves shown in [Supplementary Fig. 25b](#) indicate that the fibers exhibit no significant deterioration in mechanical performance over this temperature range, suggesting that the structural integrity and coordination interactions remain stable under varying thermal conditions.

In addition, the electrical conductivity of the fibers was measured at the same four temperatures. The results show a gradual increase in conductivity with rising temperature ([Supplementary Fig. 25c](#)), which can be attributed to the intrinsic temperature-dependent resistance decrease of MXene. Additionally, we have also supplemented the discussion in the main manuscript ([Highlight on Page 13](#)).

Reviewer #2

Comments: The manuscript has been revised extensively and it is in the near shape of publication in Nature Communications. However, it appears although the authors provided a number of data set, it has to be justified accordingly (i.e. comparison with the previously reported Mxene fiber properties and origin of improved toughness). As a minor point, there is typo to describe azimuthal angle (sometimes it is denoted as φ or Φ).

REPLIES: Thank you for pointing out this problem. To better justify our data and clarify the significance of the improved mechanical performance, we have reorganized the comparison of previously reported MXene fibers and summarized the representative strengthening and toughening strategies in the **Supplementary Table 1**. Furthermore, we have also clarified that the improved toughness mainly originates from the molecular-cord bridging strategy (**Highlighted in pages 5 and 15**). Specifically, high-aspect-ratio molecular cords bridge and mechanically interlock adjacent MXene layers, enabling efficient stress transfer, shear-strain redistribution. Most critically, when localized stresses exceed critical levels, the SWCNTs actively bridge developing microcracks, transforming what would be catastrophic fracture in neat MXene fibers into distributed, energy-absorbing modes. Together, these effects synergistically enhance the toughness of the CS-MXene fibers.

In addition, we have carefully checked the main text and Supporting Information and unified the notation of the azimuthal angle as φ throughout the revised manuscript (**Supplementary Note 1, Fig. 9, Fig. 10 and Fig. 26**).

Checklist:

1. We would like to propose a revised title to comply with our formatting requirements and improve the accessibility of your work: In-Plane Molecular Cord Bridging as a Method for Boosting MXene Fiber Strength and Toughness. Please edit the title in your manuscript files accordingly. Note that further minor changes may be made during the production process, and you will be able to check these in the proofs.

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

2. Please remove acronyms and abbreviations from the abstract.

REPLIES: Thank you for pointing out this problem. We have revised these as required in the revised manuscript.

3. Please shorten the abstract to 150 words or fewer.

REPLIES: Thank you for the comments. We have revised these as required in the revised manuscript.

4. Line 30: please change 'record' to 'enhanced'

REPLIES: Thank you for this correction. We have revised these as required in the revised manuscript.

5. Line 35: please delete "outperforming most reported MXene and carbon nanomaterial-based fibers"

REPLIES: Thank you for this correction. We have modified it in the manuscript.

6. Line 39: please change "This molecular cord strategy provides a general design paradigm for maximizing the mechanical performance of fibers derived from two-dimensional nanosheets." to "This molecular cord strategy could potentially be applied

to maximize the mechanical performance of fibers derived from other two-dimensional nanosheets"

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

7. Atomic orbital notations (sp, d, etc.) and corresponding XPS labels should be typeset in italics throughout the main text, figures and Supplementary Information, whereas all accompanying superscripts/subscripts should be typeset in roman font.

Please remove 'record' (line 123); 'record-breaking' (line 71)

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

8. Please state in the legends how many times each experiment was repeated independently with similar results. This is needed for all experiments, but is particularly important wherever results from representative experiments (such as micrographs) are shown. If space in the legends is limiting, this information can be included in a section titled "Statistics and Reproducibility" in the methods section.

Please revise this information in the legend(s) of figure(s) 2a, b, d-f; 4c-g

REPLIES: Thank you for pointing out this problem. We have indicated the number of repetitions of experiments yielding similar results in the legends accompanying representative data (such as SEM and AFM images).

9. All figures: please define acronyms at first use e.g. CS in Fig. 1 caption

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

10. Fig. 1: please include a key for interpreting atom colors

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

11. Fig. 1f,g,h: please remove red arrow

REPLIES: Thank you for this correction. We have revised these as required in the revised manuscript.

12. Fig. 2g,h, and any others: please change a.u. to arb. units

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

13. Fig. 2i: Please choose a perceptually uniform alternative to Rainbow color scheme (such as “Batlow” in: <https://www.nature.com/articles/s41467-020-19160-7>)

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

14. Fig. 2i: please quantify and label with units the color scale bar

REPLIES: Thank you for the comments. We have already provided this information in the caption for Figure 2i.

15. Fig. 2b: please specify in caption if this is e.g. strain at failure

REPLIES: Thank you for the comments. We have labeled it in Figure 3b.

16. Fig. 3c: For any bar plots with error bars, please ensure the caption includes how they were generated (e.g. if Standard Deviation, and number of samples); and where the number of samples is less than 5, that the individual data points also be plotted [See Fig. 4b in <https://www.nature.com/articles/s41467-024-45047-y> for an example].

REPLIES: Thank you for pointing out this problem. We have defined the error bars for Figure 3 and related figures and specified the exact number of repetitions in the experiments.

17. Fig. 3d: because ks is an unusual unit, recommend labeling as e.g. 's x10³' or labeling the axis as x10³

REPLIES: Thank you for pointing out this problem. We have modified it in the manuscript.

18. Fig. 5d: recommend moving the inset to a new panel in this figure to make (d) less visually busy

REPLIES: Thank you for pointing out this problem. We have reformatted Figure 5.

19. SI: Please delete line 25-26 about the movies, as this will be referenced externally in a different document

REPLIES: Thank you for pointing out this problem. We have modified it in the Supplementary Information.

20. SI: please ensure captions are not split across multiple pages e.g. Supp Fig 1, and that they are not split from their respective figure e.g. Supp Fig 7

REPLIES: Thank you for pointing out this problem. We have adjusted the layout so that the Supplementary Figures and their captions appear on the same page.

21. SI: Please separate tables from figures into their own Supplementary Tables e.g. Supp Fig 15 etc

REPLIES: Thank you for pointing out this problem. As requested, we have included the table and images, and have attached the table at the end of the supplementary information.

22. SI: e.g. Supp Table 1: For any tables that extend across multiple pages, please re-type the column header labels at the start of the table on the new page

REPLIES: Thank you for pointing out this problem. We have modified it in the Supplementary Information.

23. SI: Please remove refs to movies Line 341-344

REPLIES: Thank you for pointing out this problem. We have modified it in the

Supplementary Information.

24. We can waive word limits, so please move the Methods currently in the SI into the Methods section of the main text

REPLIES: We have moved the Methods in the SI into the Methods section of the main text

25. Please supply legends for each Supplementary Movie/Audio/Data file in your response here (not in the Supplementary Information file). Please label each files as Supplementary Movie/Audio/Data 1, etc.

REPLIES: The description of the video file is as follows:

Supplementary Movie 1: A close-up view of the wet spinning process at the nozzle tip.

Supplementary Movie 2: MD simulation for neat MXene fiber.

Supplementary Movie 3: MD simulation for C-MXene fiber.

Supplementary Movie 4: MD simulation for CS-MXene fiber.

26. The use or adaptation of previously published images is strongly discouraged. If this is unavoidable, please request the necessary rights documentation to re-use such material from the relevant copyright holders and return this to us when you submit your revised manuscript. Please check whether your manuscript or Supplementary Information contain third-party images, such as figures from the literature, stock photos, clip art or commercial satellite and map data. Please note that suspected third party content is present in figures 1a-e; 4a, b.

REPLIES: Thank you for your inquiry. We confirm that all elements in Figures 1a-e and 4a were originally created by the authors and no pre-existing or third-party elements were used. All assets were created using Photoshop and Cinema 4D, and then compiled in PowerPoint. Figure 4b shows the data from the molecular dynamics (MD) simulation. All MD calculations are implemented using the Large-scale Atomic-Molecular Massively Parallel Simulator (LAMMPS) software package.