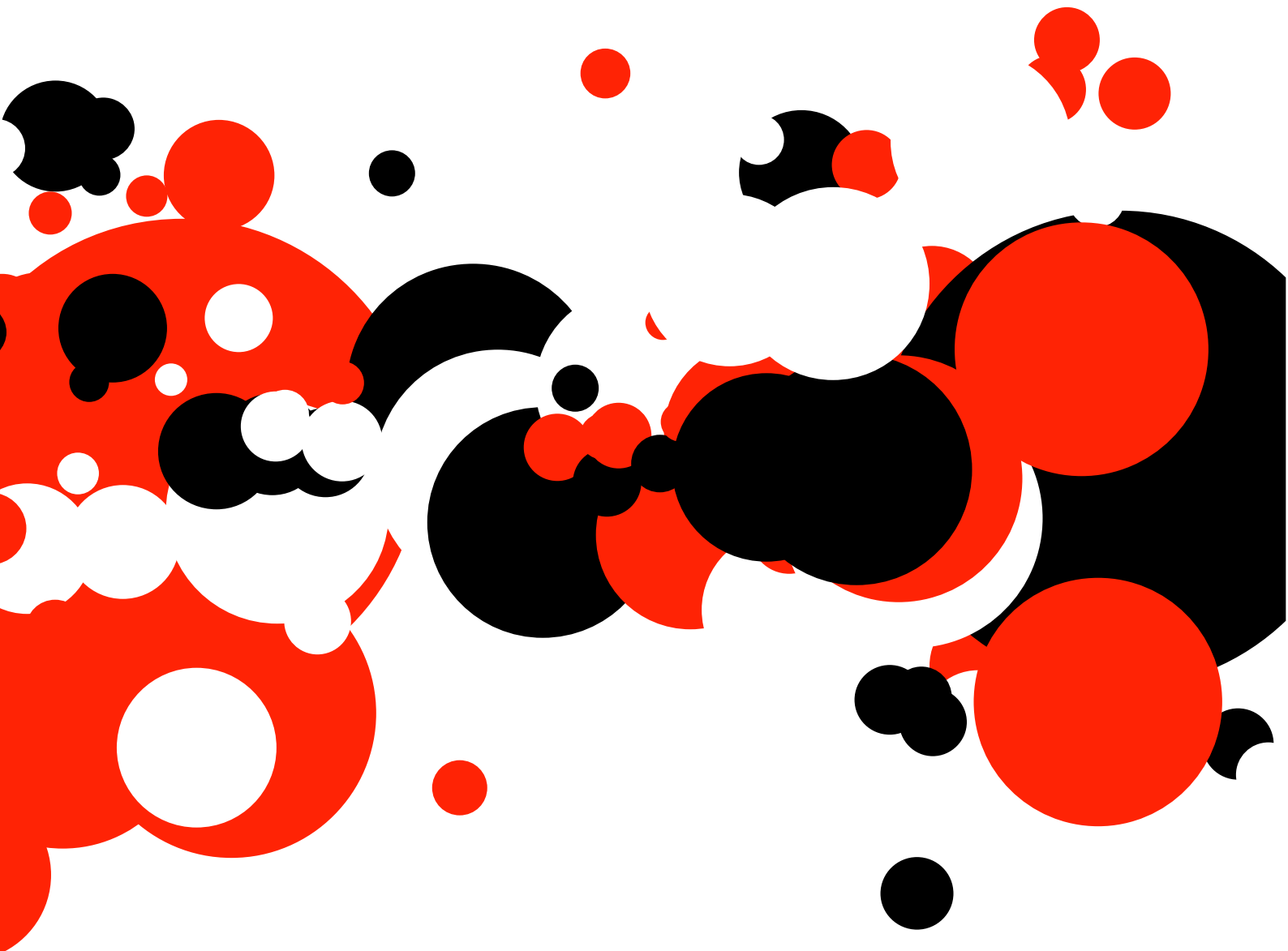




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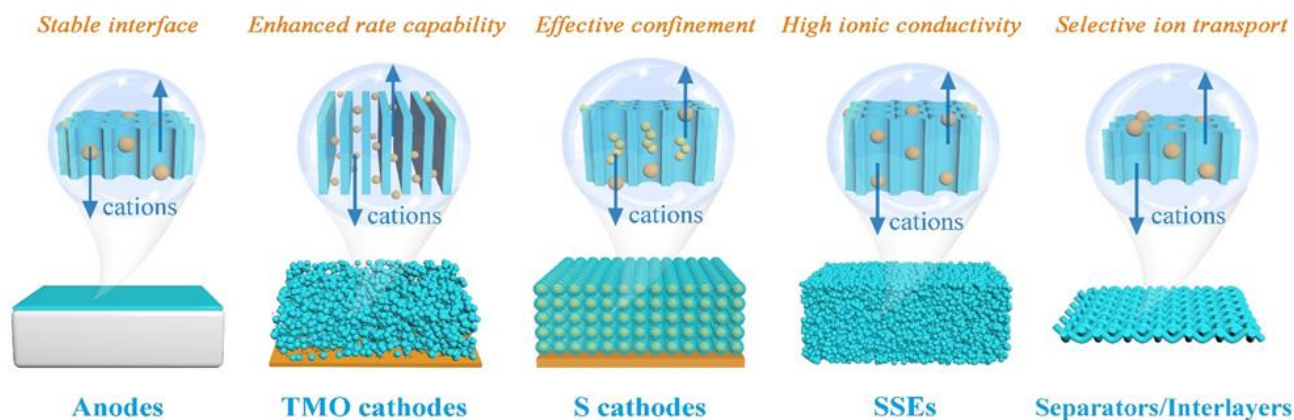
Centre for Clean Energy Technology

Research Highlights in 2024 (Jan-May)



1. Yao-Jie Lei, Lingfei Zhao, Wei-Hong Lai, Zefu Huang, Bing Sun, Pauline Jaumaux, Kening Sun, Yun-Xiao Wang, **Guoxiu Wang***, “Electrochemistry Coupling in Subnanometer Pores/Channels for Rechargeable Batteries”, **Chemical Society Review**, 2024, IF=46.20. DOI:10.1039/D3CS01043K

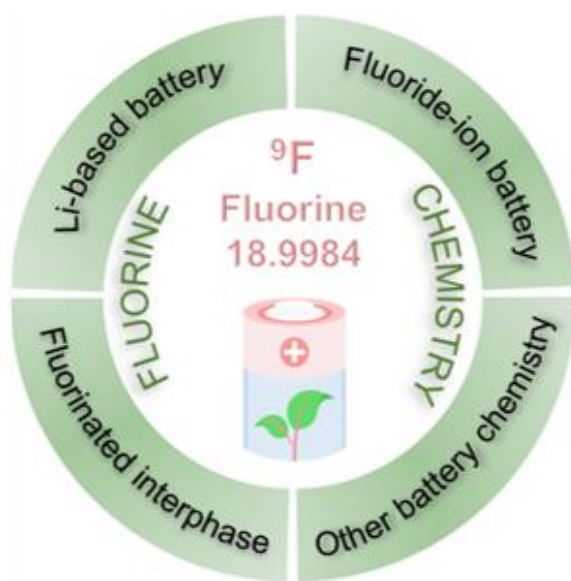
ABSTRACT: Subnanometer pores/channels (SNPCs) play crucial roles in regulating electrochemical redox reactions for rechargeable batteries. The delicate designed and tailored porous structure of SNPCs not only provides ample space for ion storage but also facilitates efficient ion diffusion within the electrodes in batteries, which can greatly improve the electrochemical performances. However, due to current technological limitations, it is challenging to synthesize and control the quality, storage, and transport of nanopores at subnanometer scale, as well as to understand the relationship between SNPCs and performances. In this review, we systematically classify and summarize materials with SNPCs from a structural perspective, dividing them into one-dimensional (1D) SNPCs, two-dimensional (2D) SNPCs, and three-dimensional (3D) SNPCs. We also unveil the unique physicochemical properties of SNPCs and analyse electrochemical couplings in SNPCs for rechargeable batteries, including cathodes, anodes, electrolytes, and functional materials. Finally, we discussed the challenges that SNPCs may face in electrochemical reactions in batteries and proposed future research directions.



<https://pubs.rsc.org/en/Content/ArticleLanding/2024/CS/D3CS01043K>

2. Yao Wang[†], Xu Yang[†], Yuefeng Meng[†], Zuxin Wen, Ran Han, Xia Hu, Bing Sun, Feiyu Kang, Baohua Li^{*}, Dong Zhou^{*}, Chunsheng Wang^{*}, **Guoxiu Wang^{*}**, “Fluorine Chemistry in Rechargeable Batteries: Challenges, Progress and Perspectives”, **Chemical Reviews**, 124, 3494–3589, 2024, IF=62.1. DOI: 10.1021/acs.chemrev.3c00826

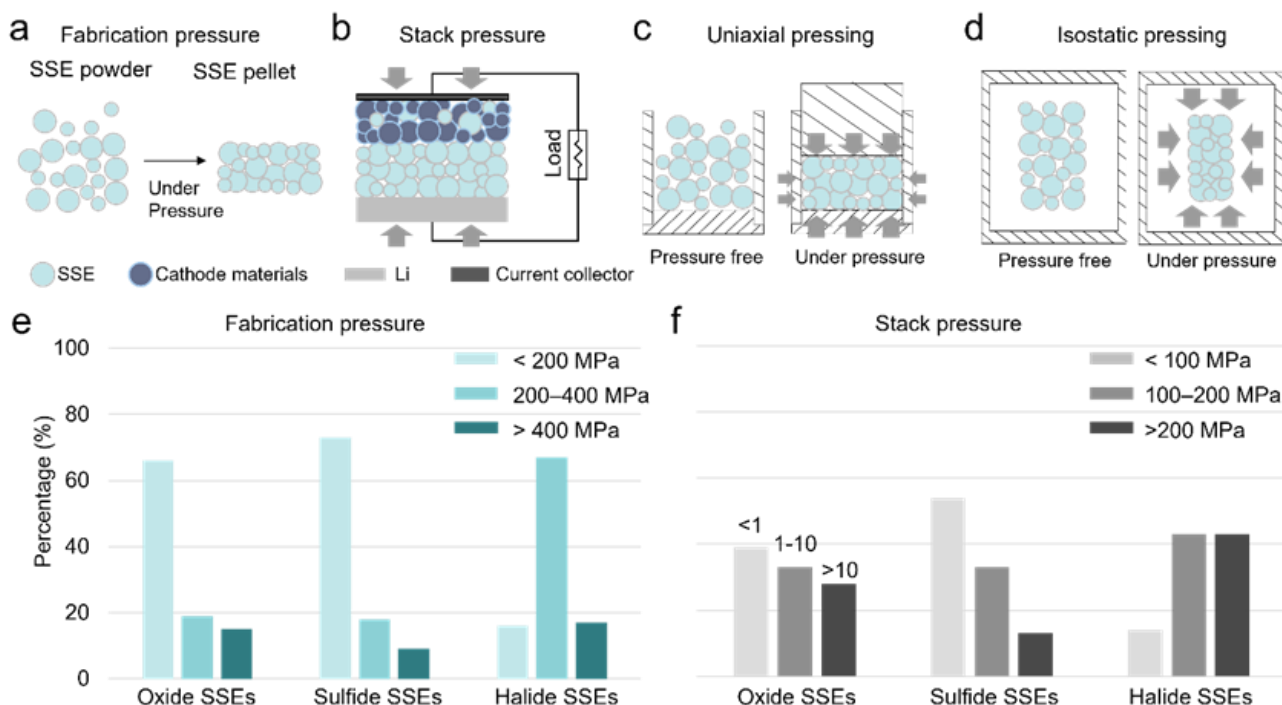
ABSTRACT: The renewable energy industry highly demands rechargeable batteries featured with low manufacturing cost, abundant resources, high energy density, high safety, wide operation temperature, and long lifespan. Utilizing fluorine chemistry to re-design the battery configurations/components is considered a critical strategy to fulfil these requirements, owing to the natural abundance, robust bond strength and extraordinary electronegativity of fluorine, and the high free energy of fluoride formation, which enables the fluorinated components with cost-effectiveness, non-flammability and intrinsic stability. Particularly, fluorinated materials and electrode|electrolyte interphases have been demonstrated to significantly affect reaction reversibility/kinetics, safety and temperature tolerance of rechargeable batteries. However, the underlining principles governing material design and the mechanistic insights of interphases at the atomic level have been largely overlooked. Spanning from exploring fluorine-containing electrodes, fluorinated electrolyte constituents and other fluorinated battery components for metal ion-shuttle batteries, to constructing fluoride-ion batteries, dual-ion batteries and beyond chemistries, this review aims to provide a comprehensive understanding of the structure-property interaction, the features of fluorinated interphases, and the cutting-edge techniques for elucidating the role of fluorine chemistry in rechargeable batteries. Further, we present current challenges and promising strategies for employing fluorine chemistry, aiming to advance the electrochemical performance, wide-temperature operation, and safety attributes of rechargeable batteries.



<https://pubs.acs.org/doi/10.1021/acs.chemrev.3c00826>

3. Xia Hu, Zhijia Zhang, Xiang Zhang, Yao Wang, Xu Yang, Xia Wang, Miryam Fayena–Greenstein, Hadas Alon Yehezkel, Steven Langford, Dong Zhou*, Baohua Li*, **Guoxiu Wang***, Doron Aurbach*, “External-pressure–electrochemistry coupling in solid-state lithium metal batteries”, **Nature Reviews Materials**, 2024, IF= 83.5. DOI: 10.1038/s41578-024-00669-y

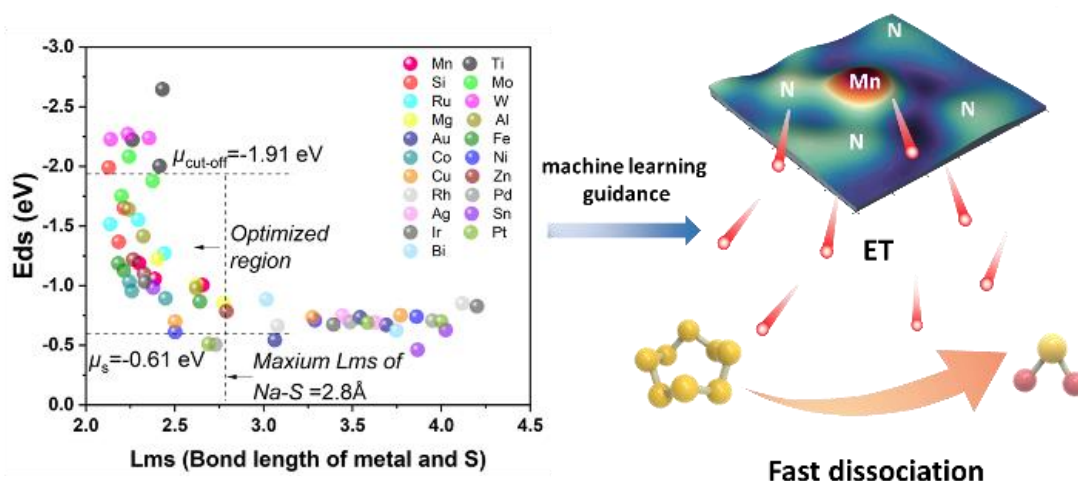
ABSTRACT: Solid–state lithium metal batteries (SSLBs) using inorganic solid–state electrolytes (SSEs) have attracted extensive scientific and commercial interest owing to their potential to provide higher energy density and safety than conventional Li–ion batteries. These batteries are subject to external pressure during both their manufacturing processes (fabrication pressure) and their current operation (stack pressure). This pressure not only affects the intrinsic properties of both the electrolytes (such as ionic conductivity and electrochemical voltage window) and electrodes (such as ion transport and structural variation) but also determines the cyclability and safety of the whole battery. Hence, understanding the effect of pressure is essential for designing high-performance SSLBs. This Review aims to elucidate the coupling between external pressure and electrochemistry in these batteries. We summarize the effects of external pressure on SSEs and electrodes, and on the interfaces between the components. We analyze the overall electrochemical performance and safety of the batteries under external pressure. Finally, we clarify the dominant challenges in achieving pressure–proof and low–pressure SSLBs, laying out a perspective for future breakthroughs.



<https://www.nature.com/articles/s41578-024-00669-y>

4. Yao-Jie Lei, Xinxin Lu, Hirofumi Yoshikawa, Daiju Matsumura, Yameng Fan, Lingfei Zhao, Jiayang Li, Shijian Wang, Qinfen Gu, Hua-Kun Liu, Shi-Xue Dou, Devaraj Shanmukaraj, Teofilo Rojo, Wei-Hong Lai*, Michel Armand*, Yun-Xiao Wang*, **Guoxiu Wang***, “Understanding the charge transfer effects of single atoms for boosting the performance of Na-S batteries”, Nature communications, 15, 3325, 2024, IF=16.6. DOI: 10.1038/s41467-024-47628-3.

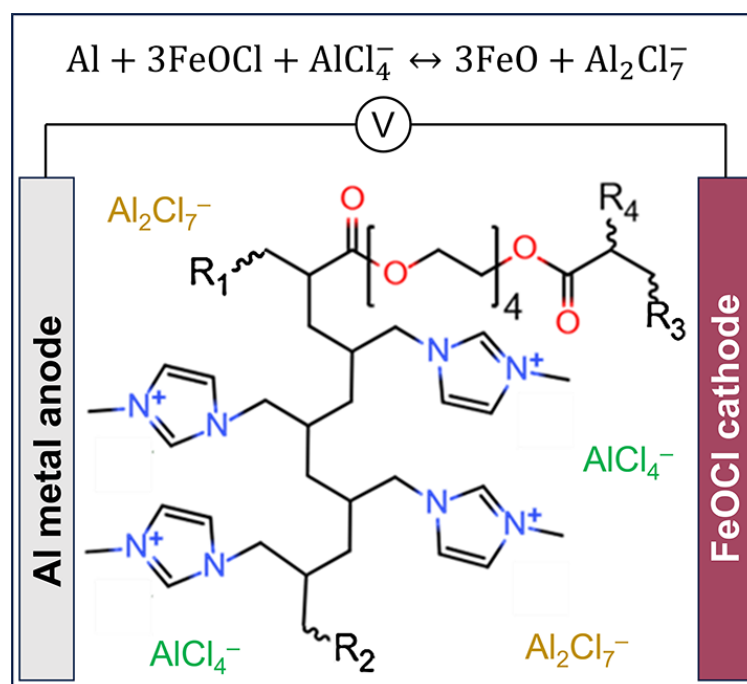
ABSTRACT: The effective flow of electrons through bulk electrodes is crucial for achieving high-performance batteries, although the poor conductivity of homocyclic sulfur molecules results in high barriers against the passage of electrons through electrode structures. This phenomenon causes incomplete reactions and the formation of metastable products. To enhance the performance of the electrode, it is important to place substitutable electrification units to accelerate the cleavage of sulfur molecules and increase the selectivity of stable products during charging and discharging. Herein, we develop a single-atom-charging strategy to address the electron transport issues in bulk sulfur electrodes. The establishment of the synergistic interaction between the adsorption model and electronic transfer helps us achieve a high level of selectivity towards the desirable short-chain sodium polysulfides during the practical battery test. These finding indicates that the atomic manganese sites have an enhanced ability to capture and donate electrons. Additionally, the charge transfer process facilitates the rearrangement of sodium ions, thereby accelerating the kinetics of the sodium ions through the electrostatic force. These combined effects improve pathway selectivity and conversion to stable products during the redox process, leading to superior electrochemical performance for room temperature sodium-sulfur batteries.



<https://www.nature.com/articles/s41467-024-47628-3>

5. Xu Yang, Zhiqiang Fu, Ran Han, Yaojie Lei, Shijian Wang, Xin Zhao, Yuefeng Meng, Hao Liu, Dong Zhou, Doron Aurbach, and **Guoxiu Wang***, “Design of Solid Polycationic Electrolyte to Enable Durable Chloride-Ion Batteries”, **Angewandte Chemie International Edition**, 2024, e202405750. IF= 16.6. DOI: 10.1002/anie.202405750.

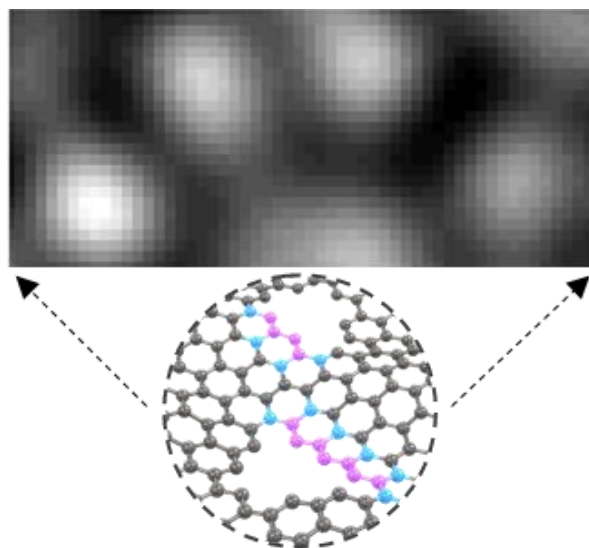
ABSTRACT: The high energy density and cost-effectiveness of chloride-ion batteries (CIBs) make them promising alternatives to lithium-ion batteries. However, the development of CIBs is greatly restricted by the lack of compatible electrolytes to support cost-effective anodes. Herein, we present a rationally designed solid polycationic electrolyte (SPE) to enable room-temperature chloride-ion batteries utilizing aluminum (Al) metal as an anode. This SPE endows the CIB configuration with improved air stability and safety (i.e. free of flammability and liquid leakage). A high ionic conductivity ($1.3 \times 10^{-2} \text{ S cm}^{-1}$ at $25 \text{ }^\circ\text{C}$) has been achieved by the well-tailored solvation structure of the SPE. Meanwhile, the solid polycationic electrolyte ensures stable electrodes|electrolyte interfaces, which effectively inhibit the growth of dendrites on the Al anodes and degradation of the FeOCl cathodes. The Al|SPE|FeOCl chloride-ion batteries showcased a high discharge capacity around 250 mAh g^{-1} (based on the cathodes) and extended lifespan. Our electrolyte design opens a new avenue for developing low-cost chloride-ion batteries.



<https://onlinelibrary.wiley.com/doi/10.1002/anie.202405750>

6. Jiufeng Ruan,[‡] Yao-Jie Lei,[‡] Yameng Fan, Marcela Chaki Borrás, Zhouxin Luo, Zichao Yan, Bernt Johannessen, Qinfen Gu, Konstantin Konstantinov, Wei Kong Pang, Wenping Sun, Jia-Zhao Wang, Hua-Kun Liu, Wei-Hong Lai, Yun-Xiao Wang, Shi-Xue Dou, “Linearly Interlinked Fe-N_x-Fe Single Atoms Catalyze High-Rate Sodium-Sulfur Batteries”, *Advanced Materials*, 2312207, 2024, IF=29.4, DOI: 10.1002/adma.202312207

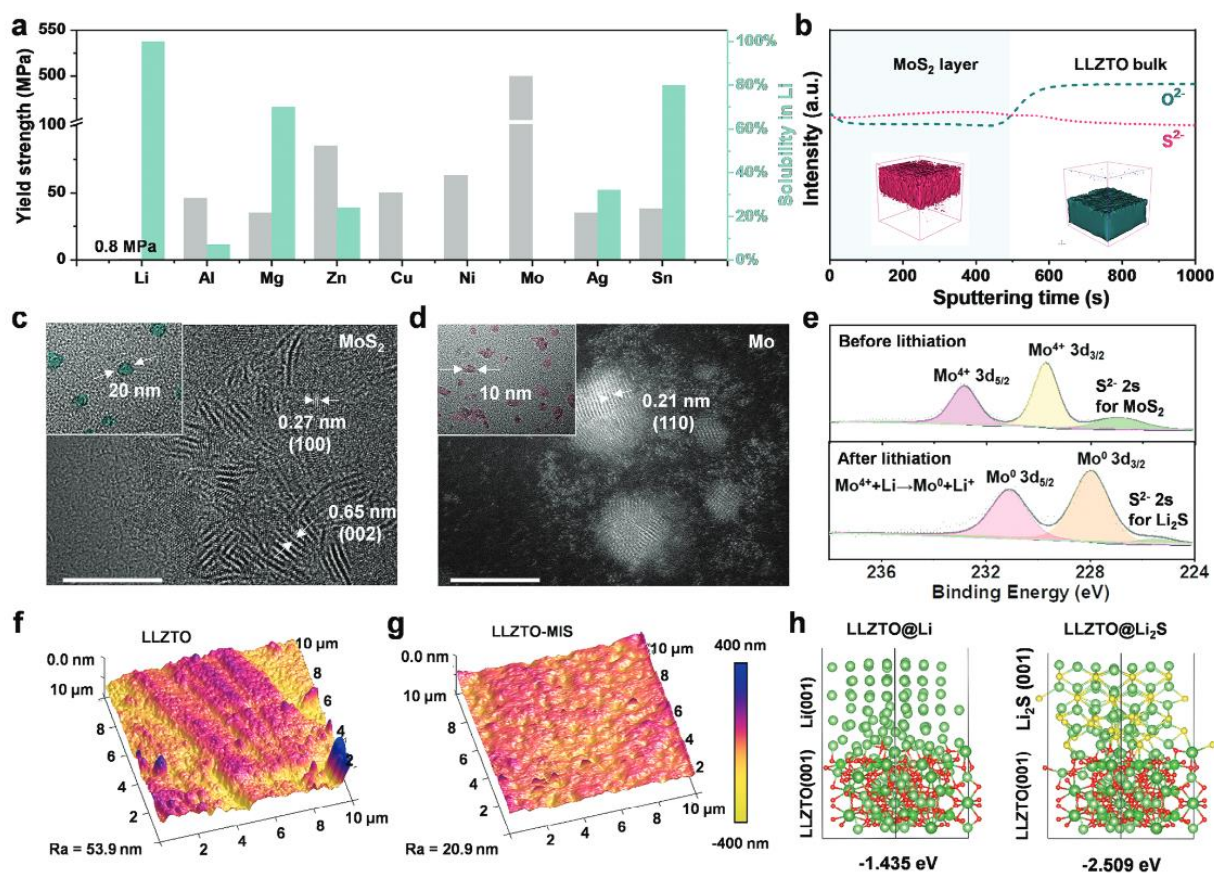
ABSTRACT: Linearly interlinked single atoms offer unprecedented physiochemical properties, but their synthesis for practical applications still poses significant challenges. Herein, we presented linearly interlinked iron single-atom catalysts that loaded onto interconnected carbon channels as cathodic sulfur hosts for room-temperature sodium-sulfur batteries. The interlinked iron single-atom exhibit unique metallic iron bonds that facilitate the transfer of electrons to the sulfur cathode, thereby accelerating the reaction kinetics. Additionally, the columnated and interlinked carbon channels ensure rapid Na⁺ diffusion kinetics to support high-rate battery reactions. By combining the iron atomic chains and the topological carbon channels, the resulting sulfur cathodes demonstrate effective high-rate conversion performance while maintaining excellent stability. Remarkably, even after 5000 cycles at a current density of 10 A g⁻¹, the Na-S battery retains a capacity of 325 mAh g⁻¹. This work could open a new avenue in the design of catalysts and carbon ionic channels, paving the way to achieve sustainable and high-performance energy devices.



<https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202312207>

7. Xia Hu, Jiahao Yu, Yao Wang, Weiqian Guo, Xiang Zhang, Michel Armand, Feiyu Kang, **Guoxiu Wang***, Dong Zhou, Baohua Li, “A Lithium Intrusion–Blocking Interfacial Shield for Wide–Pressure–Range Solid–State Lithium Metal Batteries”, **Advanced Materials**, 35, 2308275, 2023. IF= 30.85. 10.1002/adma.202308275

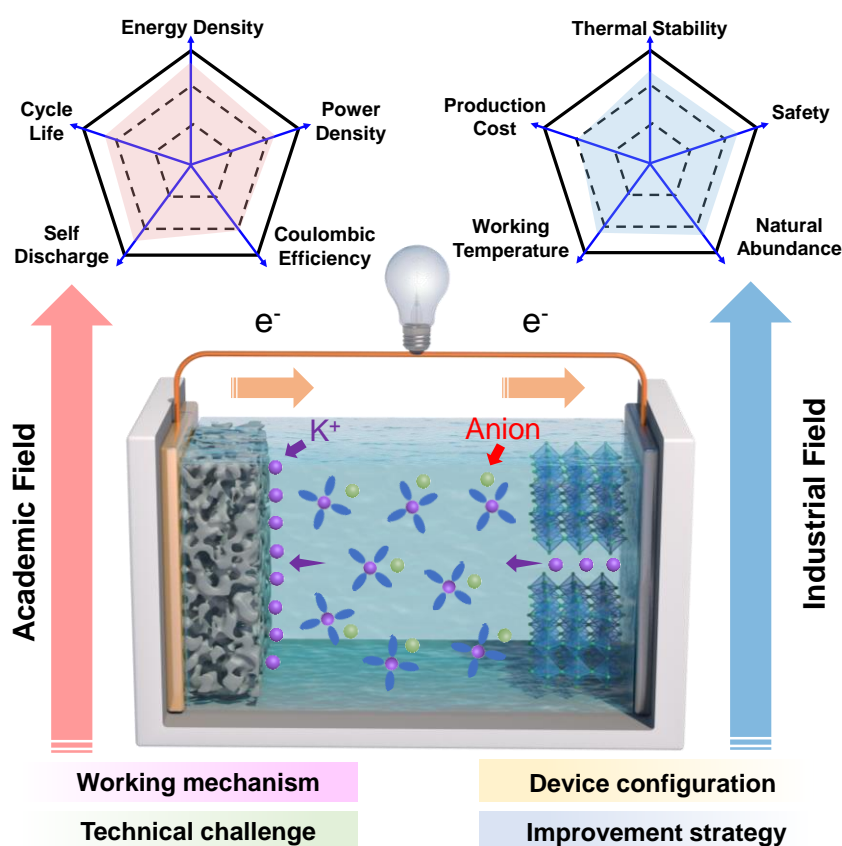
ABSTRACT: Lithium garnets are considered as promising solid-state electrolytes for next-generation solid-state Li metal batteries (SSLBs). However, the Li intrusion driven by external stack pressure triggers premature of Li metal batteries. Herein, for the first time, an in situ constructed interfacial shield is reported to efficiently inhibit the pressure-induced Li intrusion in SSLBs. Theoretical modeling and experimental investigations reveal that high-hardness metallic Mo nanocrystals inside the shield effectively suppress Li dendrite growth without alloy hardening-derived interfacial contact deterioration. Meanwhile the electrically insulated Li_2S as a shield component considerably promotes interfacial wettability and hinders Li dendrite penetration into the bulk of garnet electrolyte. Interfacial shield-protected $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ (LLZTO)-based cells exhibit significantly enhanced cyclability without short circuits under conventional pressures of ≈ 0.2 MPa and even at high pressure of up to 70 MPa; which is the highest endurable stack pressure reported for SSLBs using garnet electrolytes. These key findings are expected to promote the wide-pressure-range applications of SSLBs.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.202308275>

8. Shuoqing Zhao, Guohao Li, Bohan Zhang, Tianming Li, Mingchuan Luo, Bing Sun, **Guoxiu Wang*** Shaojun Guo, “Technological roadmap for potassium-ion hybrid capacitors”, *Joule*, 8, P922-943, 2024, IF=39.8. DOI: 10.1016/j.joule.2024.03.006

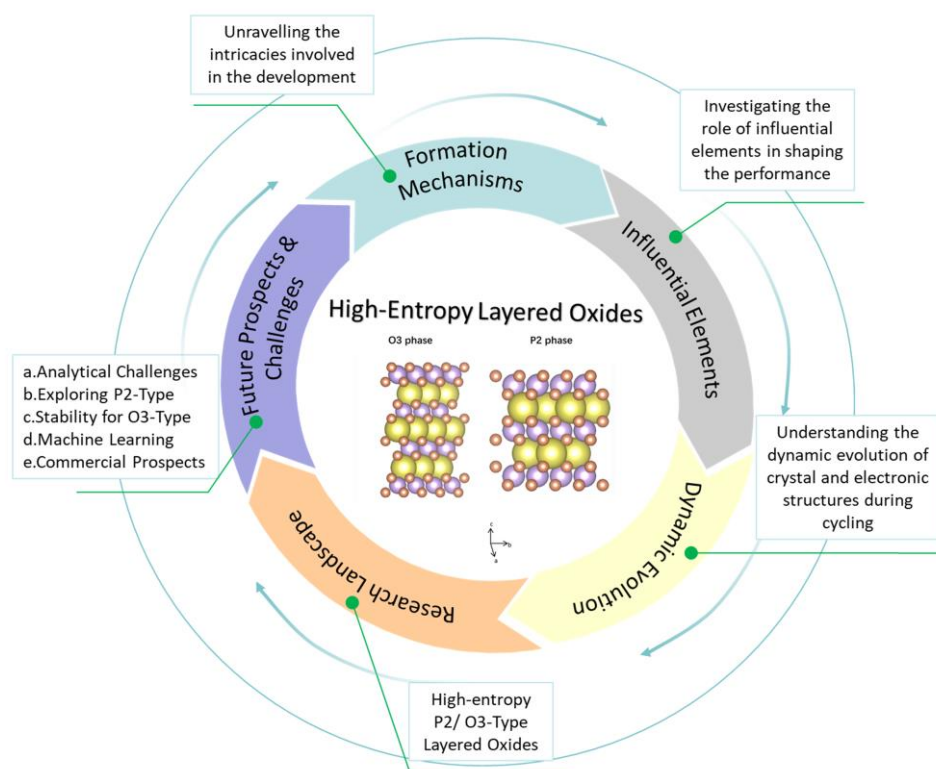
ABSTRACT: Potassium-ion hybrid capacitors (PIHCs) are in principle advantageous over the traditional metal-ion hybrid capacitors (MIHCs) in terms of low cost, safety and reliability, holding enormous potential for the massive market, yet remains largely an uncharted field. Herein, we provide a comprehensive review of recent advances on PIHCs, including fundamental understanding, device configurations and design rationale. Then, technical hurdles that essentially impede the development of PIHCs are discussed, accompanied by corresponding improvement strategies for all aspects of PIHCs. Lastly, how far are we from the lab bench to the consumer market is highlighted, and a comprehensive roadmap starting from cooperative components integration and ending with the future direction of next-generation PIHCs for grid-scale energy storage is clearly described. We anticipate that this review will timely arouse strong interest and attention from both academic and industrial communities to this rising technology and makes a revolutionary step forward toward commercial applications.



[https://www.cell.com/joule/abstract/S2542-4351\(24\)00136-3](https://www.cell.com/joule/abstract/S2542-4351(24)00136-3)

9. Hong Gao, Jiayi Li, Fan Zhang, Congcong Li, Jun Xiao, Xinming Nie*, Guilai Zhang, Yang Xiao, Dingyi Zhang, Xin Guo, Yong Wang, Yong-Mook Kang, **Guoxiu Wang***, Hao Liu, “Revealing the Potential and Challenges of High-Entropy Layered Cathodes for Sodium-Based Energy Storage”, **Advanced Energy Materials**, 2304529, 2024, IF=27.8. DOI: 10.1002/aenm.202304529

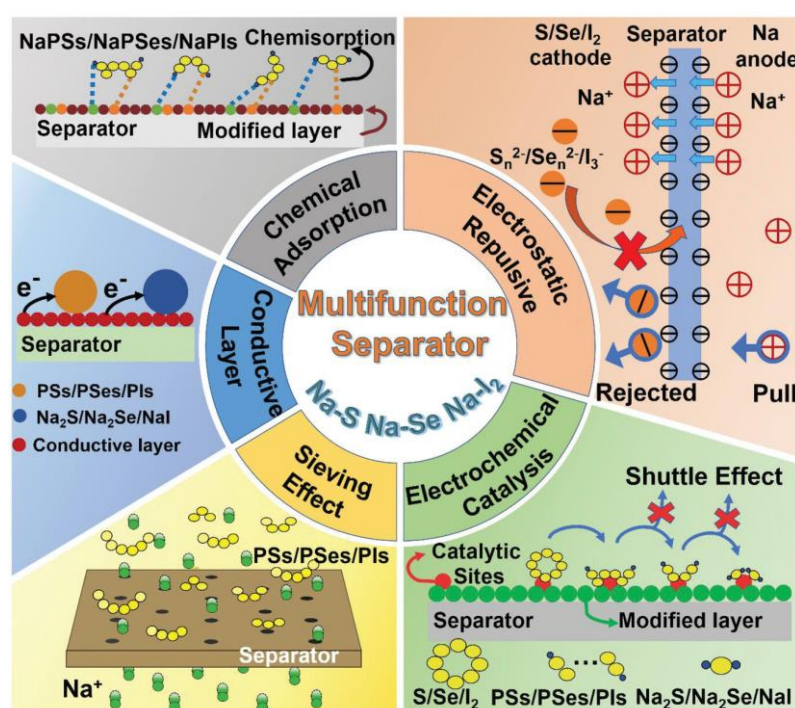
ABSTRACT: Sodium-ion batteries (SIBs) reflect a strategic move for scalable and sustainable energy storage. The focus on high-entropy (HE) cathode materials, particularly layered oxides, has ignited scientific interest due to the unique characteristics and effects to tackle their shortcomings, such as inferior structural stability, sluggish reaction kinetics, severe Jahn-Teller effects induced lattice distortion, and poor oxygen reversibility at high voltage. This review focuses on high-entropy oxide materials, highlighting their fundamentals, design principles, and application in layered oxide cathodes for SIBs. It delves into the growth mechanism, composition-properties correlations, and the functional roles of high-entropy design in enhancing the performance of layered oxide cathodes. Furthermore, it furnishes a comprehensive survey of recent advancements and persisting challenges within the domain of layered high-entropy cathode materials, as well as offers insights into potential future research directions in line with the current state of knowledge.



<https://onlinelibrary.wiley.com/doi/full/10.1002/aenm.202304529>

10. Jing Xu, Yashuang Qiu, Jianhao Yang, Haolin Li, Pingan Han, Yang Jin, Xiaoli Zhang, Hao Liu, Bing Sun, **Guoxiu Wang***, “Review of Separator Modification Strategies: Targeting Undesired Anion Transport in Room Temperature Sodium–Sulfur/Selenium/Iodine Batteries”, **Advanced Functional Materials**, 33, 2306206, 2023. IF= 19. DOI: 10.1002/adfm.202306206

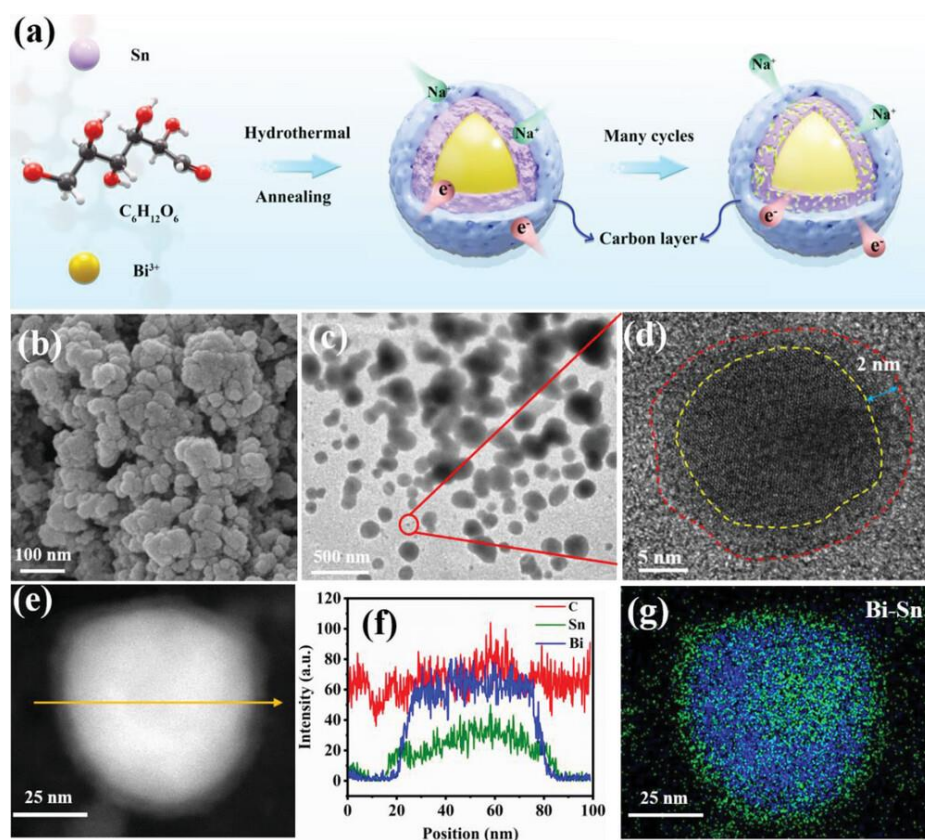
ABSTRACT: Rechargeable sodium–sulfur/selenium/iodine (Na–S/Se/I₂) batteries are regarded as promising candidates for large-scale energy storage systems, with the advantages of high energy density, low cost, and environmental friendliness. However, the electrochemical performances of Na–S/Se/I₂ batteries are still restricted by several inherent issues, including the “shuttle effect” of polysulfides/polyselenides/polyiodides (PSs/PSes/PIs), sluggish kinetics of the conversion reactions at the cathodes, and Na dendrite growth at the anodes. Among these challenges, uncontrolled “shuttle effect” of PSs/PSes/PIs is a major contributing factor for the irreversible loss of active cathode materials and severe side reactions on Na metal anodes, leading to rapid failure of the batteries. Separator modification has been demonstrated to be an effective strategy to suppress the shuttling of PSs/PSes/PIs. Herein, the latest achievement in modifying separators for high-performance Na–S/Se/I₂ batteries is comprehensively reviewed. The reaction mechanisms of each battery system are first discussed. Then, strategies of separator modification based on the different functions for regulating the transportation of PSs/PSes/PIs are summarized, including applying electrostatic repulsive interaction, introducing conductive layers, improving sieving effects, enhancing chemisorption capability, and adding efficient electrocatalysts. Finally, future perspectives on the practical application of modified separators in high-energy rechargeable batteries are provided.



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11. Jun Chen, Guilai Zhang, Jun Xiao, Jiayi Li, Yang Xiao, Dingyi Zhang, Hong Gao, Xin Guo, **Guoxiu Wang***, Hao Liu, “A Stress Self-Adaptive Bimetallic Stellar Nanosphere for High-Energy Sodium-Ion Batteries”, **Advanced Functional Materials**, 33, 2307959, 2023. IF= 19. DOI: 10.1002/ adfm.202307959

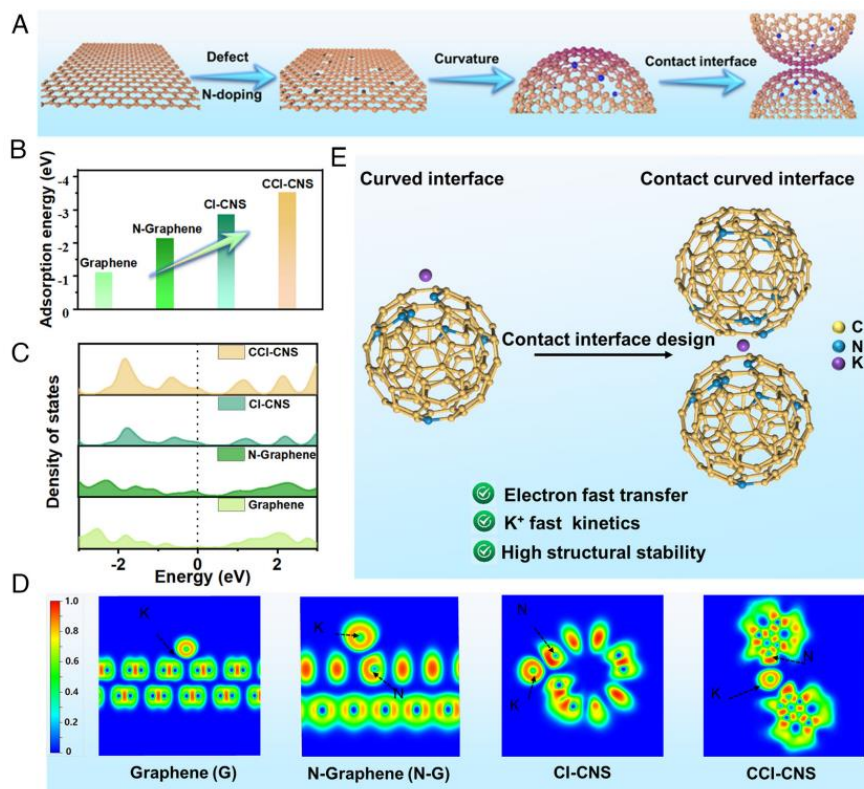
ABSTRACT: Bimetallic composites exhibit great potential as anode materials in advanced energy storage systems owing to their inherent tunability, cost-effectiveness, and simultaneous achievement of high specific capacity and low reaction potential. However, simple biphase mixing often fails to achieve satisfactory performance. Herein, an innovative stress self-adaptive bimetallic stellar nanosphere (50–200 nm) wherein bismuth (Bi) is fabricated, as a core, is seamlessly encapsulated by a tin (Sn) sheath (Sn-Bi@C). This well-integrated stellar configuration with bimetallic nature embodies the synergy between Bi and Sn, offering fortified conductivity and heightened sodium ion diffusion kinetics. Moreover, through meticulous utilization of finite element analysis simulations, a homogeneous stress distribution within the Sn-enveloped Bi, efficiently mitigating the structural strain raised from the insertion of Na⁺ ions, is uncovered. The corresponding electrode demonstrates remarkable cyclic stability, as it exhibits no capacity decay after 100 cycles at 0.1 A g⁻¹. Furthermore, it achieves an impressive 86.9% capacity retention even after an extensive 2000 cycles. When employed in a Na₃V₂(PO₄)₃ || Sn-Bi@C full cell configuration, it demonstrates exceptional capacity retention of 97.06% after 300 cycles at 1 A g⁻¹, along with a high energy density of 251.2 W h kg⁻¹.



<https://onlinelibrary.wiley.com/doi/10.1002/adfm.202307959>

12.Xuan Li, Yaxin Wang, Junxiong Wu, Lijuan Tong, Shuling Wang, Xiaoyan Li, Chuanping Li, Manxi Wang, Manxian Li, Weiwei Fan, Xiaochuan Chen, Qinghua Chen, **Guoxiu Wang***, and Yuming Chen, “Engineering contact curved interface with high-electronic-state active sites for high-performance potassium-ion batteries”, **Proceedings of the National Academy of Sciences (PNAS)**, 120, e2307477120, 2023. IF=12.78. DOI: 10.1073/pnas.2307477120

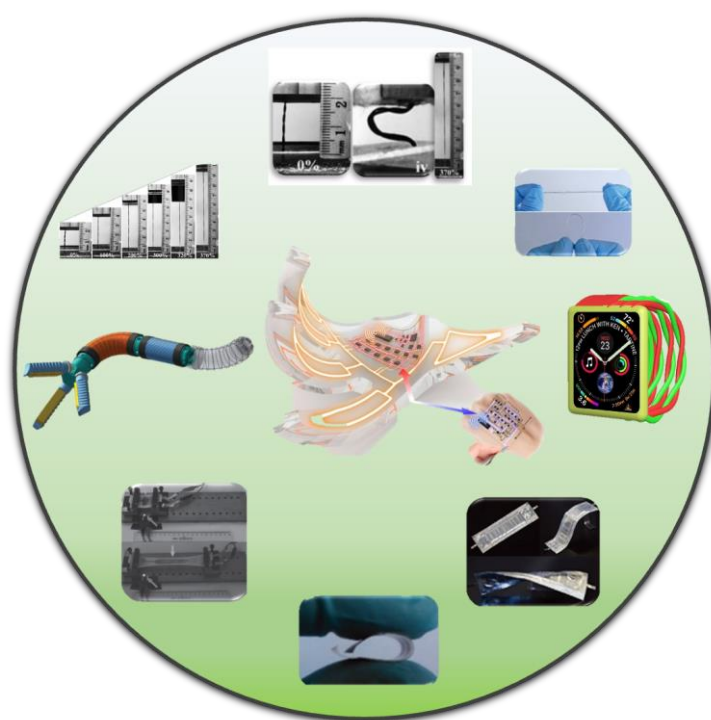
ABSTRACT: Potassium-ion batteries (PIBs) have attracted ever-increasing interest due to the abundant potassium resources and low cost, which are considered a sustainable energy storage technology. However, the graphite anodes employed in PIBs suffer from low capacity and sluggish reaction kinetics caused by the large radius of potassium ions. Herein, we report nitrogen-doped, defect-rich hollow carbon nanospheres with contact curved interfaces (CCIs) on carbon nanotubes (CNTs), namely CCI-CNS/CNT, to boost both electron transfer and potassium-ion adsorption. Density functional theory calculations validate that engineering CCIs significantly augments the electronic state near the Fermi level, thus promoting electron transfer. In addition, the CCIs exhibit a pronounced affinity for potassium ions, promoting their adsorption and subsequently benefiting potassium storage. As a result, the rationally designed CCI-CNS/CNT anode shows remarkable cyclic stability and rate capability. This work provides a strategy for enhancing the potassium storage performance of carbonaceous materials through CCI engineering, which can be further extended to other battery systems.



<https://www.pnas.org/doi/10.1073/pnas.2307477120>

13. Tao Huang, Xu Yang, Jun Xiao, Hong Gao, Yong Wang, Hao Liu, **Guoxiu Wang***, “Advancing Low-Dimensional Flexible Energy Devices for Wearable Technology”, **Journal of Materials Chemistry A**, 2024. IF=11.9. DOI: 10.1039/D4TA02241F

ABSTRACT: Flexible batteries and supercapacitors have witnessed remarkable progress in recent years, making them integral to the field of wearable technology. These energy devices offer the unique ability to seamlessly conform to the wearer's movements, ensuring uninterrupted power supply for an array of smart devices. To meet the pressing demand for high-efficiency wearable batteries, it is crucial to explore various forms of flexibility and their impact on practical applications. This perspective critically examines the recent advancements in one-dimensional and two-dimensional flexible energy devices, emphasizing their pivotal roles in enabling wearable technology to reach their full potential. Lastly, the challenges and opportunities associated with these devices are discussed in detail, underscoring the paramount importance of low-dimensional flexible energy solutions in the ever-evolving landscape of wearable devices.



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