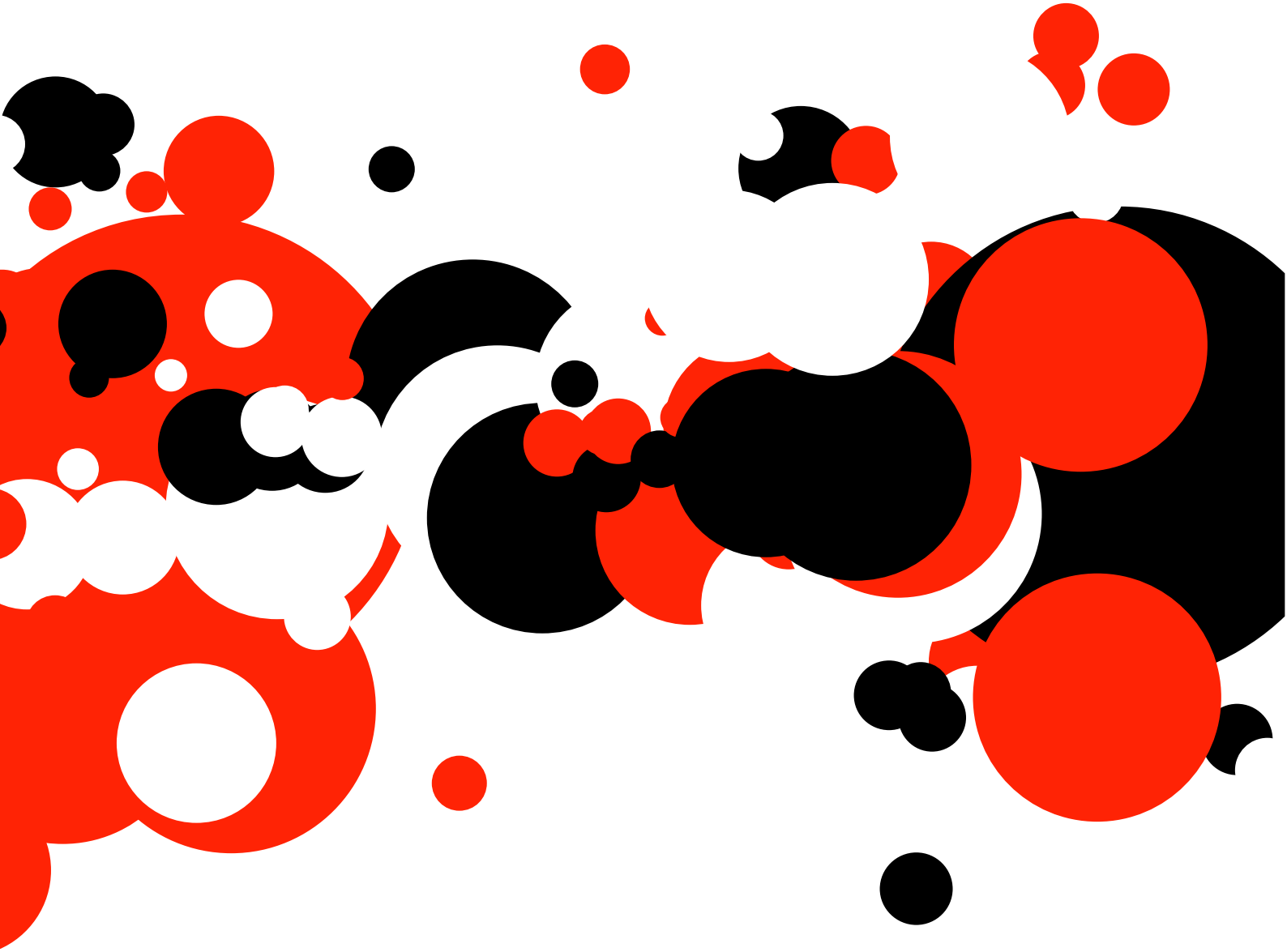
University of Technology Sydney

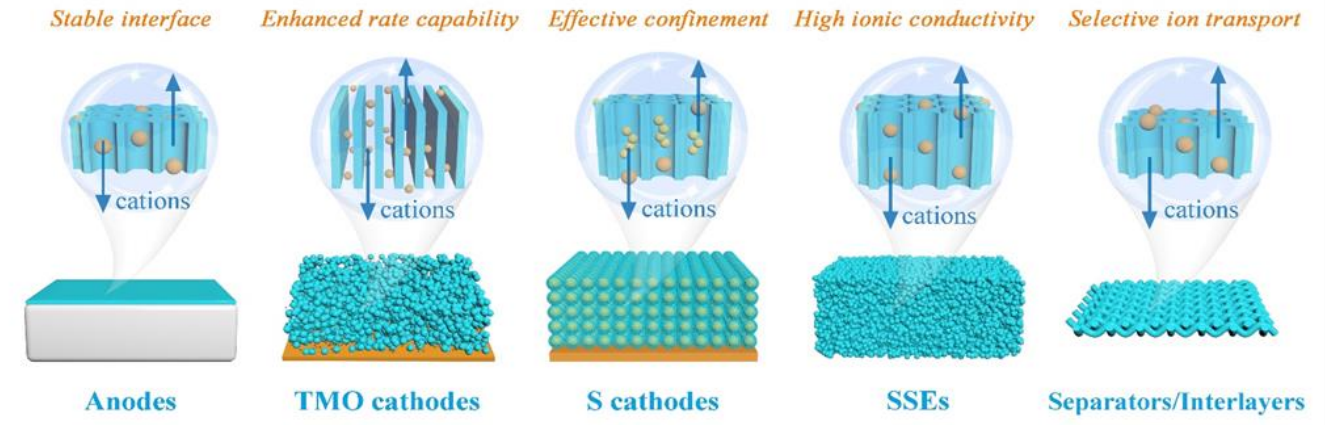
# Centre for Clean Energy Technology

## Research Highlights in 2024



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**, 53, 3829-3895, 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.

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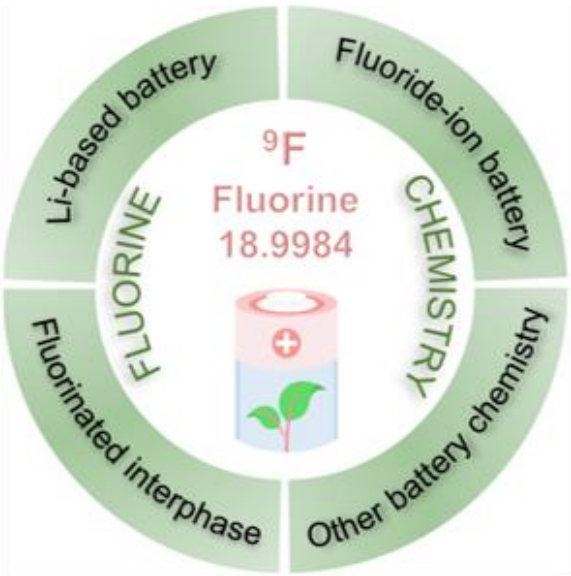
2. Yao Wang[†], Xu Yang[†], Yuefeng Meng[†], Zuxin Wen, Ran Han, Xia Hu, Bing Sun, Feiyu Kang, Baohua Li[*], Dong Zhou[*], Chunsheng Wang[*], **Guoxiu Wang**[*], "Fluorine Chemistry in Rechargeable Batteries: Challenges, Progress and Perspectives", **Chemical Reviews**, 124, 3494–3589, 2024, IF=62.1.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

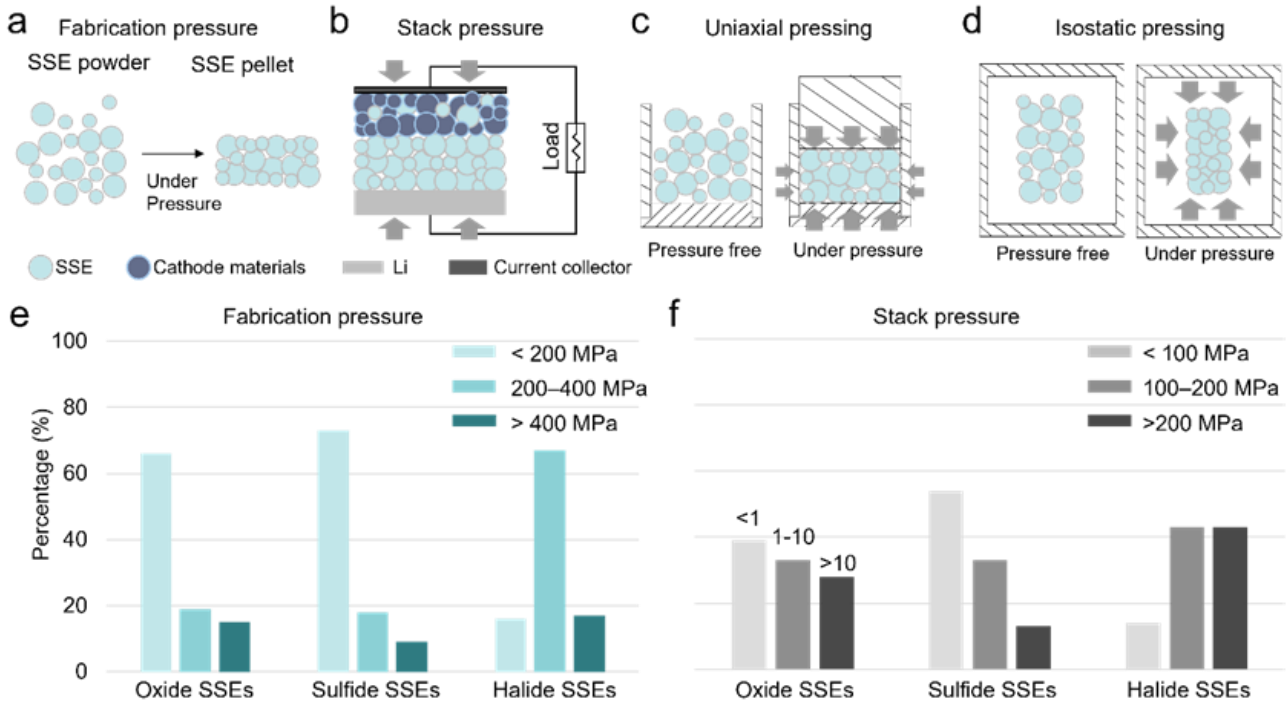
**ABSTRACT:** The renewable energy industry highly demands rechargeable batteries featured with low manufacturing cost, abundant resources, high energy density, high safety, wide operation temperature, and long lifespan. Utilizing fluorine chemistry to re-design the battery configurations/components 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.

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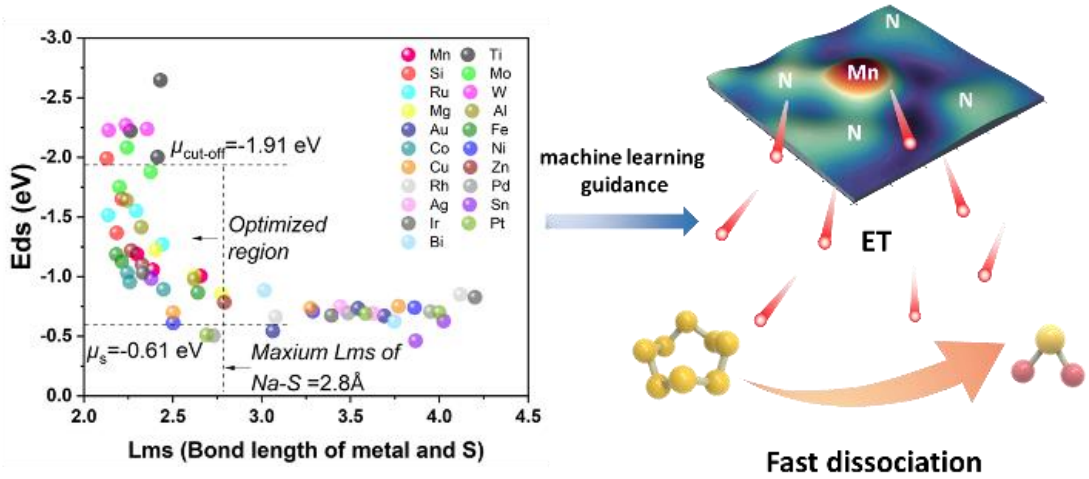
4. Yao-Jie Lei, Xinxin Lu, Hirofumi Yoshikawa, Daiju Matsumura, Yameng Fan, Lingfei Zhao, Jiayang Li, Shijian Wang, Qinfen Gu, Hua-Kun Liu, Shi-Xue Dou, Devaraj Shanmukaraj, Teofilo Rojo, Wei-Hong Lai*, Michel Armand*, Yun-Xiao Wang*, **Guoxiu Wang***, "Understanding the charge transfer effects of single 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.

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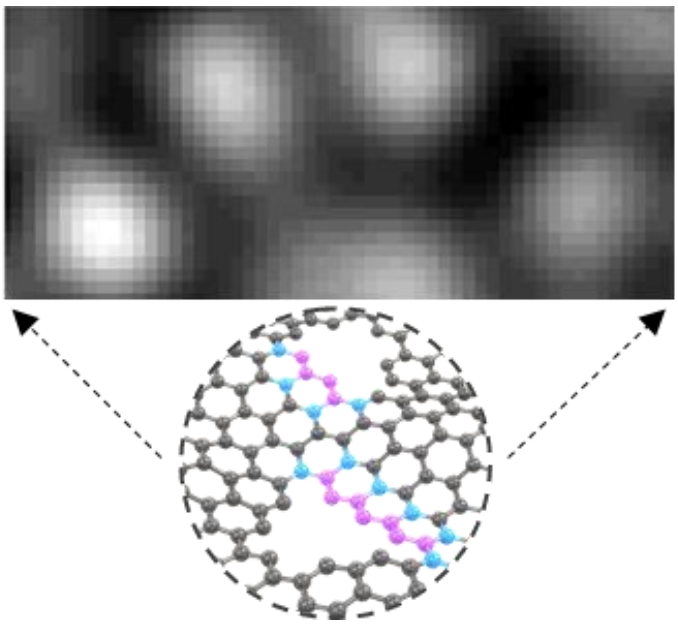
5. Jiufeng Ruan,‡ Yao-Jie Lei,‡ Yameng Fan, Marcela Chaki Borras, Zhouxin Luo, Zichao Yan, Bernt Johannessen, Qinfen Gu, Konstantin Konstantinov, Wei Kong Pang, Wenping Sun, Jia-Zhao Wang, Hua-Kun Liu, Wei-Hong Lai, Yun-Xiao Wang, Shi-Xue Dou, "Linearly Interlinked Fe-$N_x$-Fe Single Atoms Catalyze High-Rate Sodium-Sulfur Batteries", **Advanced Materials**, 36, 2312207, 2024, IF=29.4, DOI: 10.1002/adma.202312207

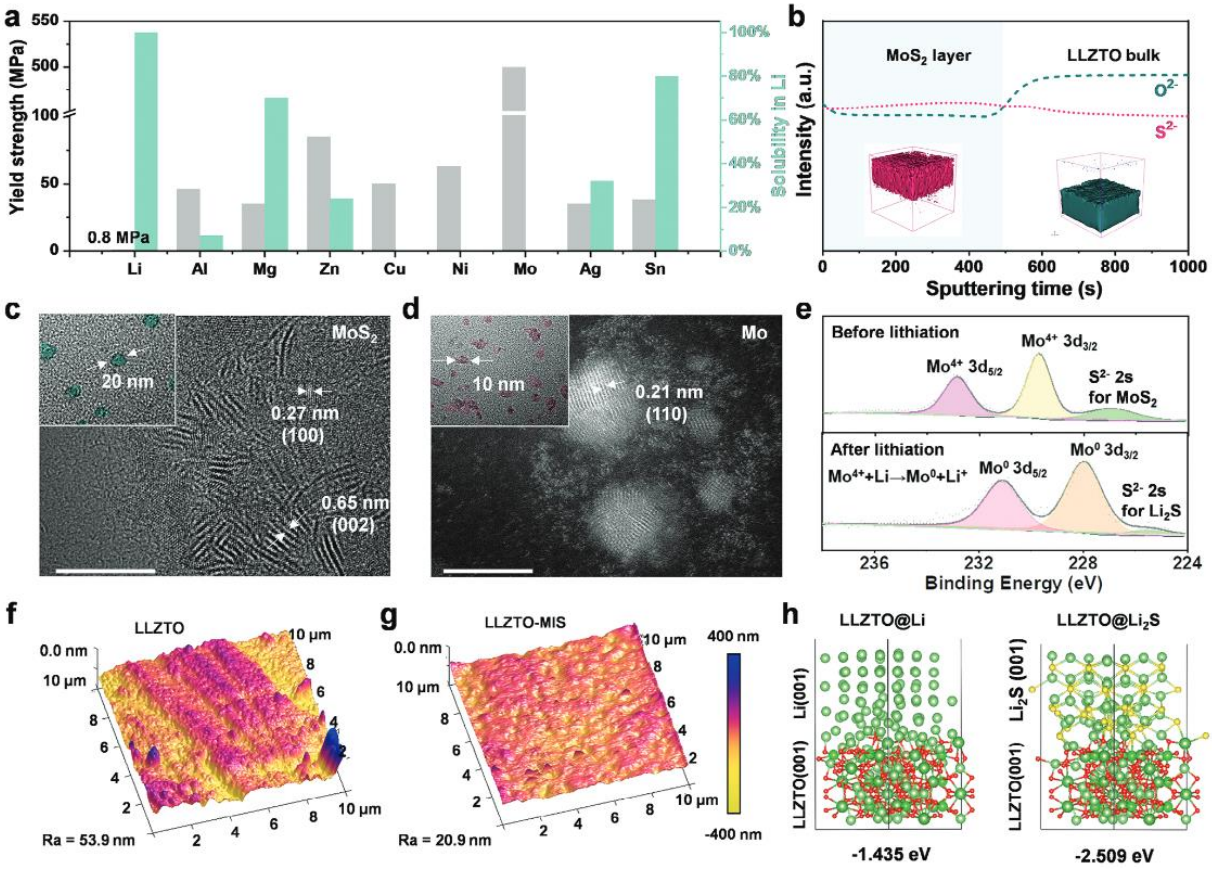
**ABSTRACT:** Linearly interlinked single atoms offer unprecedented physiochemical properties, but their synthesis for practical applications still poses significant challenges. Herein, we presented linearly interlinked iron single-atom catalysts that loaded onto interconnected carbon channels as cathodic 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

6. 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**, 36, 2308275, 2024. 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 $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ (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.

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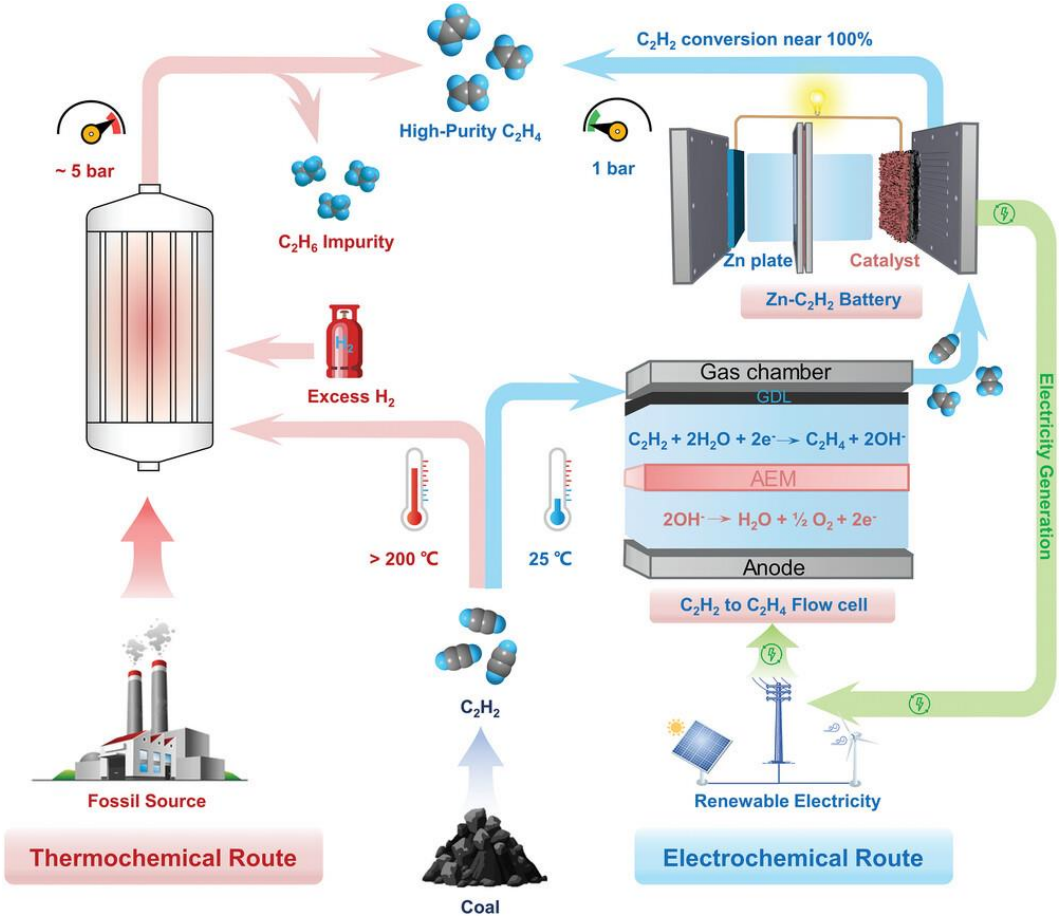
7. Zeliang Wu, Jinqiang Zhang, Qihui Guan, Xing Liu, Hanting Xiong, Shixia Chen, Wei Hong, Dongfang Li, Yaojie Lei, Shuguang Deng, Jun Wang, **Guoxiu Wang***, "Near 100% Conversion of Acetylene to High-purity Ethylene at Ampere-Level Current", **Advanced Materials**, 36, 2408681, 2024. IF= 29.4. DOI: 10.1002/adma.202408681.

**ABSTRACT:** Direct production of high-purity ethylene from acetylene using renewable energy through electrocatalytic semi-hydrogenation presents a promising alternative to traditional thermocatalytic processes. However, the low conversion of acetylene results in a significant amount of acetylene impurities in the product, necessitating additional purification steps. Herein, a tandem electrocatalytic system that integrates acetylene electrolyzer and zinc-acetylene battery units for high-purity ethylene production is designed. The ultrathin CuO nanoribbons with enriched oxygen vacancies ($CuO_{1-x}$ NRs) as electrocatalysts achieve a remarkable 93.2% Faradaic efficiency of ethylene at an ampere-level current density of 1.0 A $cm^{-2}$ in an acetylene electrolyzer, and the power density reaches 3.8 mW $cm^{-2}$ in a zinc-acetylene battery under acetylene stream. Moreover, the tandem electrocatalysis system delivers a single-pass acetylene conversion of 99.998% and ethylene selectivity of 96.1% at a high current of 1.4 A. Experimental data and calculations demonstrate that the presence of oxygen vacancies accelerates water dissociation to produce active hydrogen atoms while preventing the over-hydrogenation of ethylene. Furthermore, techno-economic analysis reveals that the tandem system can dramatically reduce the overall ethylene production cost compared to the conventional thermocatalytic processes. A novel strategy for complete acetylene-to-ethylene conversion under mild conditions, establishing a non-petroleum route for the production of ethylene is reported.

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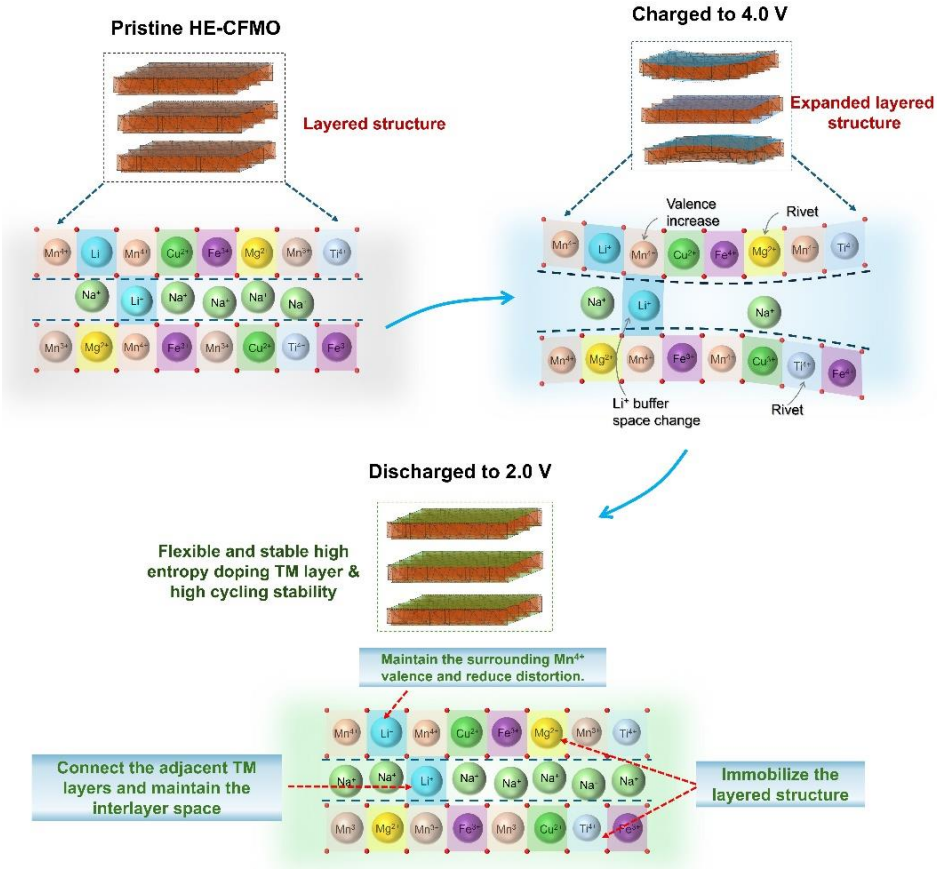
8. Zefu Huang[+], Shijian Wang[+], Xin Guo*, Frederick Marlton, Yameng Fan, Wei-Kong Pang, Tao Huang, Jun Xiao, Dongfang Li, Hao Liu, Qinfen Gu, Cheng-Chieh Yang, Chung-Li Dong, Bing Sun*, and **Guoxiu Wang***, "High-Entropy Layered Oxide Cathode Materials with Moderated Interlayer Spacing and Enhanced Kinetics for Sodium-Ion Batteries", **Advanced Materials**, 36, 2410857, 2024. IF=29.4, DOI: 10.1002/adma.202410857.

**ABSTRACT:** Sodium-ion batteries (SIBs) with low cost and environmentally friendly features have recently attracted significant attention for renewable energy storage. Sodium layer oxides stand out as a type of promising cathode material for SIBs owing to their high capacity, good rate performance, and high compatibility for manufacturing. However, the poor cycling stability of layer oxide cathodes due to structure distortion greatly impacts their practical applications. Herein, we designed a high entropy doped Cu, Fe and Mn-based layered oxide (HE-CFMO), $Na_{0.95}Li_{0.05}Mg_{0.05}Cu_{0.20}Fe_{0.22}Mn_{0.35}Ti_{0.13}O_2$ for high-performance SIBs. The HE-CFMO cathode possesses high-entropy transition metal (TM) layers with a homogeneous stress distribution, providing a moderated interlayer spacing to maintain the structure stability and enhance $Na^+$ ion diffusion. In addition, Li doping in TM layers increases the Mn valence state, which effectively suppresses the Jahn-Teller effect, thus stabilizing the layered structure during cycling. Furthermore, the use of nontoxic and low-cost raw materials benefits future commercialization and reduces the risk of environmental pollution. As a result, the HE-CFMO cathode exhibits a super cycling performance with a 95% of capacity retention after 300 cycles. This work provides a promising strategy to improve the structure stability and reaction kinetics of cathode materials for SIBs.

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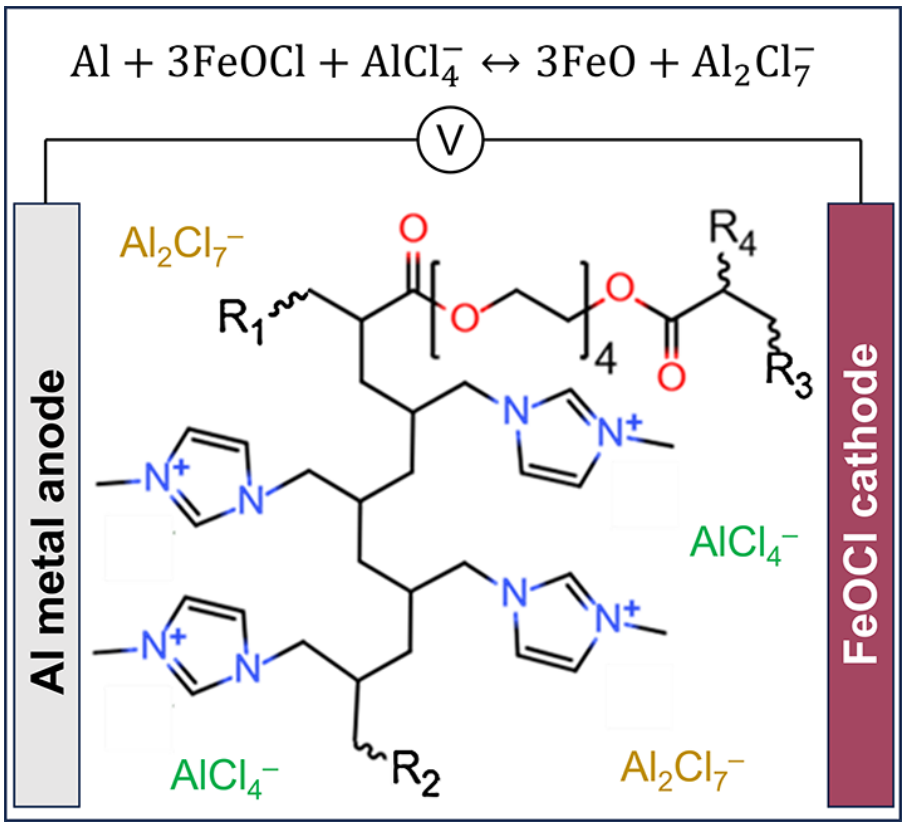
9. Xu Yang, Zhiqiang Fu, Ran Han, Yaojie Lei, Shijian Wang, Xin Zhao, Yuefeng Meng, Hao Liu, Dong Zhou, Doron Aurbach, and **Guoxiu Wang***, "Design of Solid Polycationic Electrolyte to Enable Durable Chloride-Ion Batteries", **Angewandte Chemie International Edition**, 63, e202405750, 2024. IF= 16.6. 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 ($1.3\times10^{-2}$ S $cm^{-1}$ at 25 °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.

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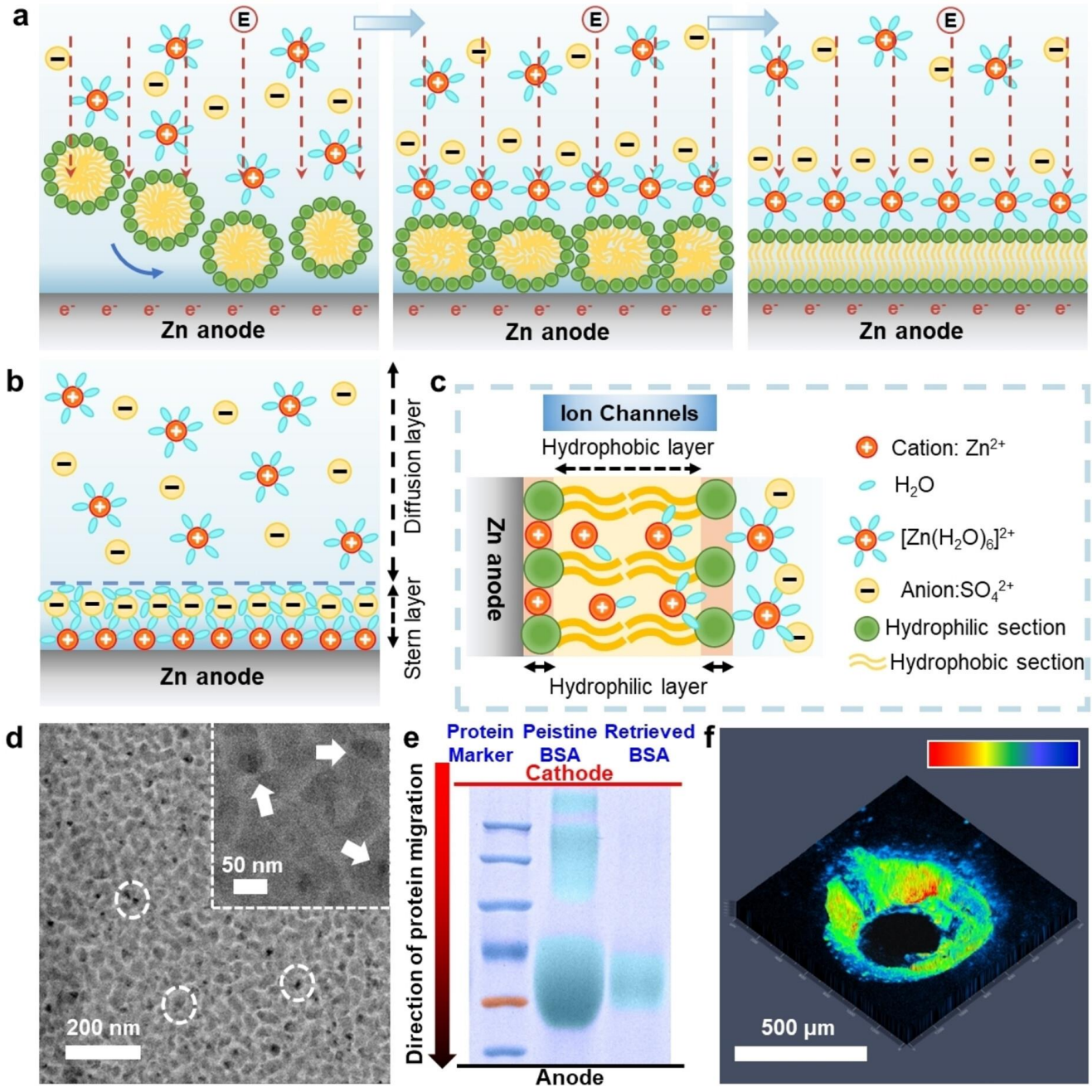
10. Jiahui Lu, Tianyi Wang, Jian Yang, Xin Shen, Huan Pang, Bing Sun, **Guoxiu Wang*,** Chengyin Wang, "Multifunctional Self-Assembled Bio-Interfacial Layers for High-Performance Zinc Metal Anodes", **Angewandte Chemie International Edition**, 63, e202409838, 2024. IF=16.6. DOI: 10.1002/anie.202409838

**ABSTRACT:** Bovine serum albumin (BSA) is employed as an electrolyte additive in the aqueous $ZnSO_4$ electrolyte, which can form a bio-interfacial layer on the Zinc (Zn) anode via a self-assembly process. The BSA-based interfacial layer with hydrophilic and hydrophobic fragments forms unique ion channels, which regulate the migration of Zn ions and facilitate their desolvation process, significantly diminishing parasitic reactions and contributing to improved cycling performance.

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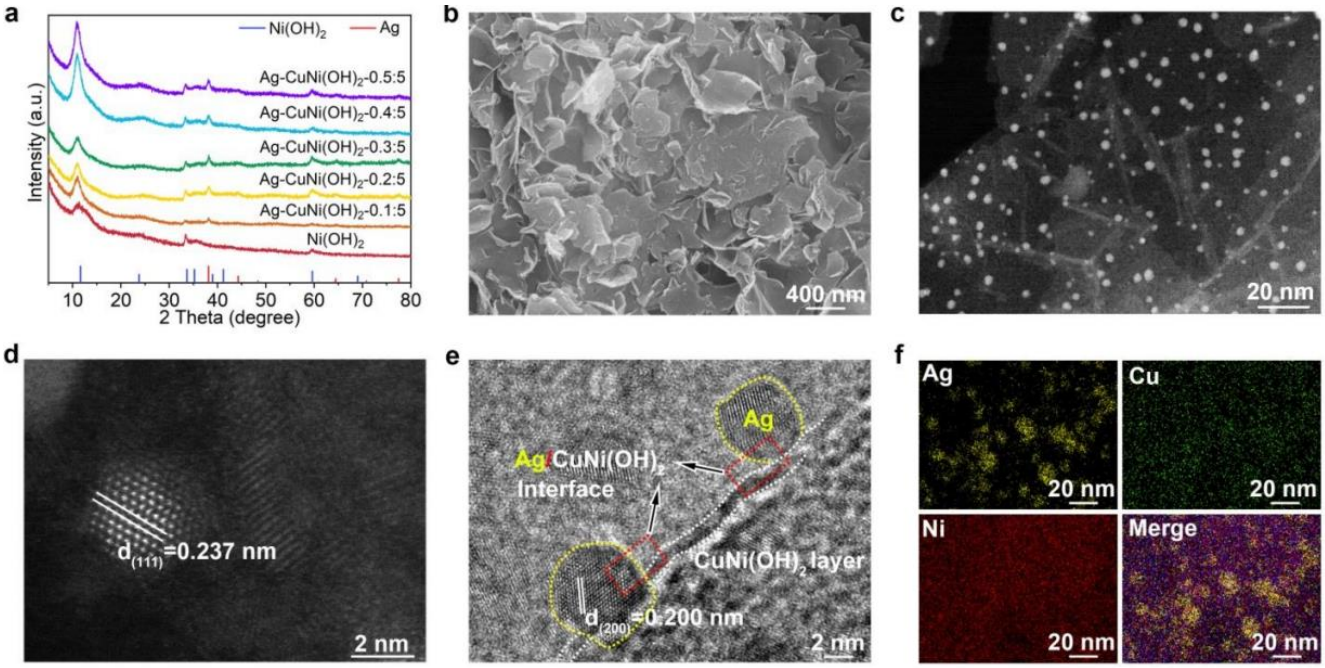
11. Wei Ye, Ye Zhang, Liang Chen, Fangfang Wu, Yuanhui Yao, Wei Wang, Genping Zhu, Gan Jia, Zhongchao Bai, Shi Xue Dou, Peng Gao, Nana Wang, **Guoxiu Wang***, "A Strongly Coupled Metal/Hydroxide Heterostructure Cascades Carbon Dioxide and Nitrate Reduction Reactions toward Efficient Urea Electrosynthesis", **Angewandte Chemie International Edition**, 63, e202410105, 2024. IF=16.6. DOI: 10.1002/anie.202410105

**ABSTRACT:** The direct coupling of nitrate ions and carbon dioxide for urea synthesis presents an appealing alternative to the Bosch–Meiser process in industry. The simultaneous activation of carbon dioxide and nitrate, however, as well as efficient C–N coupling on single active site, poses significant challenges. Here, we propose a novel metal/hydroxide heterostructure strategy based on synthesizing an Ag-CuNi$(OH)_2$ composite to cascade carbon dioxide and nitrate reduction reactions for urea electrosynthesis. The strongly coupled metal/hydroxide heterostructure interface integrates two distinct sites for carbon dioxide and nitrate activation, and facilitates the coupling of *CO (on silver, where * denotes an active site) and *NH2 (on hydroxide) for urea formation. Moreover, the strongly coupled interface optimizes the water splitting process and facilitates the supply of active hydrogen atoms, thereby expediting the deoxyreduction processes essential for urea formation. Consequently, our Ag-CuNi$(OH)_2$ composite delivers a high urea yield rate of 25.6 mmol $gcat^{-1}$ $h^{-1}$ and high urea Faradaic efficiency of 46.1%, as well as excellent cycling stability. This work provides new insights into the design of dual-site catalysts for C–N coupling, considering their role on the interface.

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12. Chao Liu, Caichao Ye, Tianning Zhang, Jiheng Tang, Kunpeng Mao, Long Chen, Liang Xue, Jingwen Sun, Wenqing Zhang, Xin Wang, Pan Xiong, **Guoxiu Wang*,** Junwu Zhu, **2024**, "Bio-inspired Double Angstrom-Scale Confinement in Ti-deficient $Ti_{0.87}O_2$ Nanosheet Membranes for Ultrahigh-performance Osmotic Power Generation", **Angewandte Chemie International Edition**, 63 (4), e202315947. IF=16.6. DOI: 10.1002/anie.202315947.

**ABSTRACT:** We design a Ti-deficient $Ti_{0.87}O_2$ nanosheet membrane with biological double angstrom-scale confinement (DAC) ion-permselective channels, named DAC-$Ti_{0.87}O_2$, for high-efficiency osmotic power generators. Benefiting from the precisely designed DAC ion-permselective channels, the DAC-$Ti_{0.87}O_2$ membrane achieves an unprecedented power density of up to 17.8 W $m^{-2}$ by mixing 0.5/0.01 M NaCl solution, far exceeding the reported macroscopic-scale membranes.


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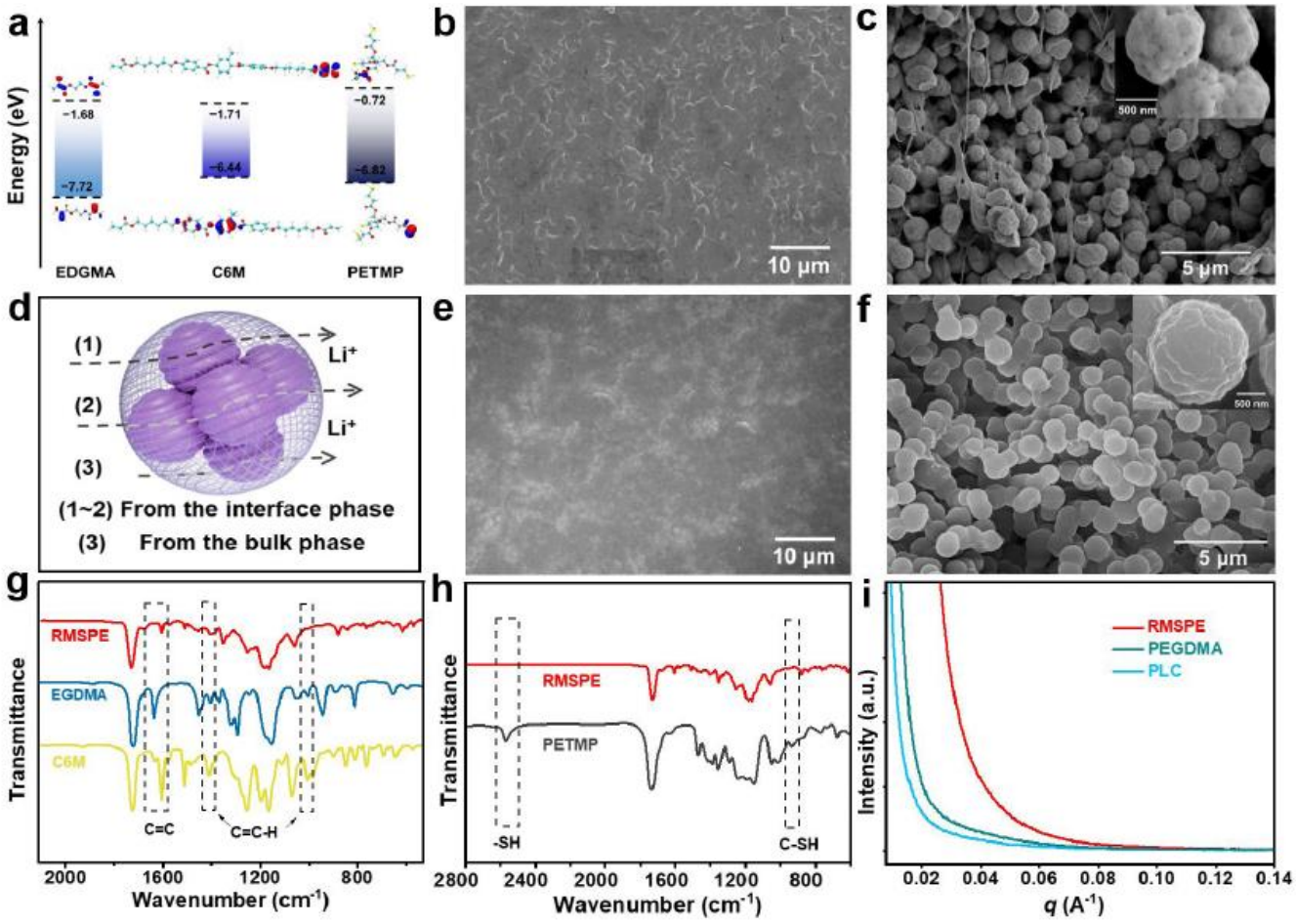
13. Weiting Ma, Xiurui Cui, Yong Chen, Shuang Wan, Shunshun Zhao, Jiajun Gong, **Guoxiu Wang***, Shimou Chen, "Designing a Refined Multi-Structural Polymer Electrolyte Framework for Highly Stable Lithium-Metal Batteries", **Angewandte Chemie International Edition**, 63, e202415617, 2024. IF=16.6. DOI: 10.1002/anie.202415617.

**ABSTRACT:** Rational structural designs of solid polymer electrolytes featuring rich interface-phase morphologies can improve electrolyte connection and rapid ion transport. However, these rigid interfacial structures commonly result in diminished or entirely inert ionic conductivity within their bulk phase, compromising overall electrolyte performance. Herein, a multi-component ion-conductive electrolyte was successfully designed based on a refined multi-structural polymer electrolyte (RMSPE) framework with uniform Li+ solvation chemistry and rapid $Li^+$ transporting kinetics. The RMSPE is constructed via polymerization-induced phase separation based on a rational combination of lithiophilic components and rigid/flexible chain units with significant hydrophobic/hydrophilic contrasts. Further refined by coating a robust polymer network, this all-organic design endows a homogenous micro-nano porous structure, providing a novel framework favorable for rapid ion transport in both its soft interfacial and bulk phases. The RMSPE exhibited excellent ion conductivity of 1.91 mS cm−1 at room temperature and a high $Li^+$ transference number of 0.7. Assembled symmetrical Li cells realized stable cycling for over 2400 h at 3.0 mA $cm^{-2}$. $LiFePO_4$ full batteries demonstrated a long lifespan of 3300 cycles with a capacity retention of 93.5% and stable cycling performance at −35 °C. This innovative design concept offers a promising perspective for achieving high-performance polymer-based Li metal batteries.

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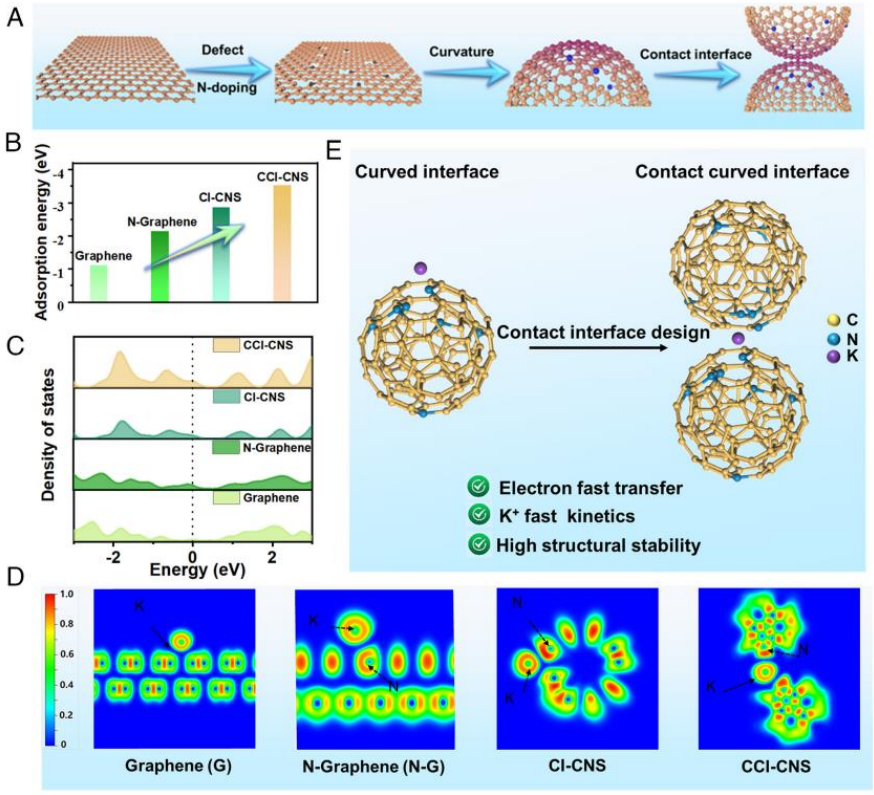
14. Xuan Li, Yaxin Wang, Junxiong Wu, Lijuan Tong, Shuling Wang, Xiaoyan Li, Chuanping Li, Manxi Wang, Manxian Li, Weiwei Fan, Xiaochuan Chen, Qinghua Chen, **Guoxiu Wang***, and Yuming Chen, "Engineering contact curved interface with high-electronic-state active sites for high-performance potassium-ion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

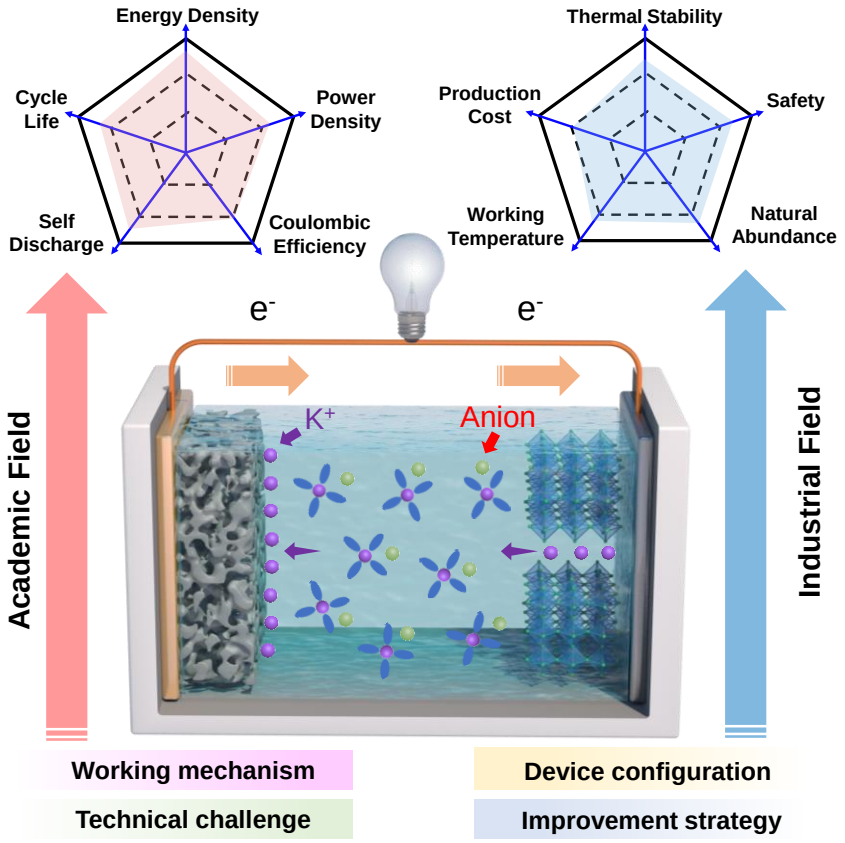
**ABSTRACT:** Potassium-ion batteries (PIBs) have attracted ever-increasing interest due to the abundant potassium resources and low cost, which are considered a sustainable energy storage technology. However, the graphite anodes employed in PIBs suffer from low capacity and sluggish reaction kinetics 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

15. 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

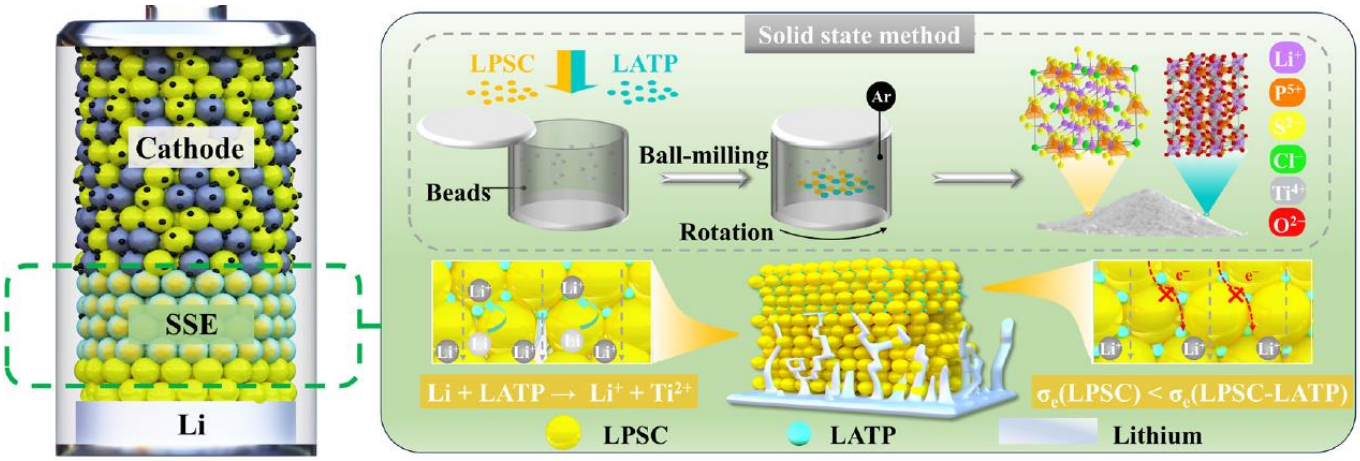
**ABSTRACT:** Potassium-ion hybrid capacitors (PIHCs) are in principle advantageous over the traditional metal-ion hybrid capacitors (MIHCs) in terms of low cost, safety and reliability, holding enormous potential for the massive market, yet remains largely an uncharted field. Herein, we provide a comprehensive 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

16. Ya Chen, Xin Gao, Zheng Zhen, Xiao Chen, Ling Huang, Deli Zhou, Tengfei Hu, Bozhen Ren, Runjing Xu, Jiayi Chen, Xiaodong Chen, Lifeng Cui, **Guoxiu Wang***, "The Construction of Multifunctional Solid Electrolytes Interlayer for Stabilizing $Li_6PS_5Cl$-based All-Solid-State Lithium Metal Batteries", **Energy & Environmental Science**, 17, 9288-9302, 2024. IF=27.8. DOI: 10.1039/D4EE03289F

**ABSTRACT:** The electrochemical performance of all-solid-state Li metal batteries (ASSLMBs) can be improved by resolving the challenges triggered by the uncontrolled growth of Li dendrites throughout the solid electrolytes (SEs). Herein, a well-defined composite of micro-$Li_6PS_5Cl$ (LPSC) and nano-$Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ (LATP) is designed as a LPSC–LATP interlayer sandwiched between LPSC electrolytes for ASSLMBs. This fabrication exhibits electron-blocking functionalities, which reduce the probability of reaction with $Li^+$ ions for the formation of anode-initiated and grain boundary (GB)-initiated dendrites. More importantly, it also creates localized eliminated micro-environments of Li dendrites through the high transient reactivity between them, and the remaining cracks can be dynamically and effectively filled by decomposition products, thereby clearly suppressing Li dendrite nucleation, propagation and penetration as well as simultaneously contributing to the enhancement of battery performance and stability. With this approach, a fine-tuned LPSC–LATP (8S–2O) interlayer enables symmetrical Li/LPSC/8S–2O/LPSC/Li cells to achieve an ultra-high critical current density (CCD) of over 5 mA $cm^{-2}$ at room temperature, and ultra-long-term cycling at a current density of 10 mA $cm^{-2}$ for over 1600 h. Additionally, ASSLMBs employing commercial $LiCoO_2$ cathodes can deliver exceptional durability, with an extremely high 85.6% retention of initial discharge capacity and coulombic efficiency (CE) of >99.6% after 1200 cycles at 1C (1.28 mA $cm^{-2}$). These experimental batteries demonstrate the application potential of this configuration of SEs for the commercialization of ASSLMBs.

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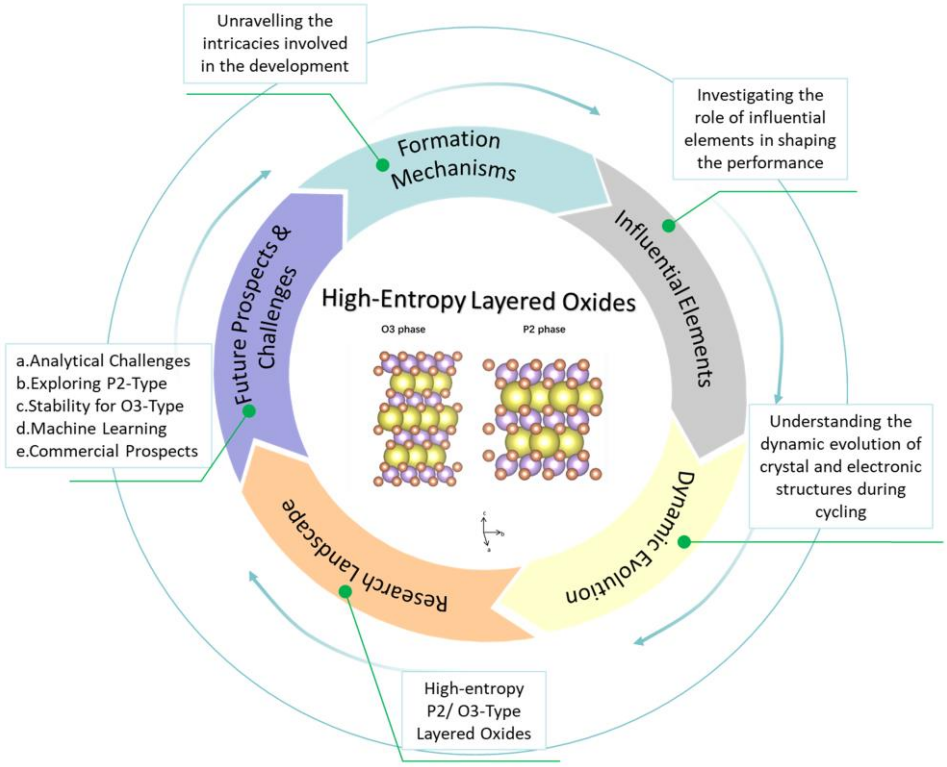
17. Hong Gao, Jiayi Li, Fan Zhang, Congcong Li, Jun Xiao, Xinming Nie*, Guilai Zhang, Yang Xiao, Dingyi Zhang, Xin Guo, Yong Wang, Yong-Mook Kang, **Guoxiu Wang***, Hao Liu, "Revealing the Potential and Challenges of High-Entropy Layered Cathodes for Sodium-Based Energy Storage", **Advanced Energy Materials**, 14, 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.

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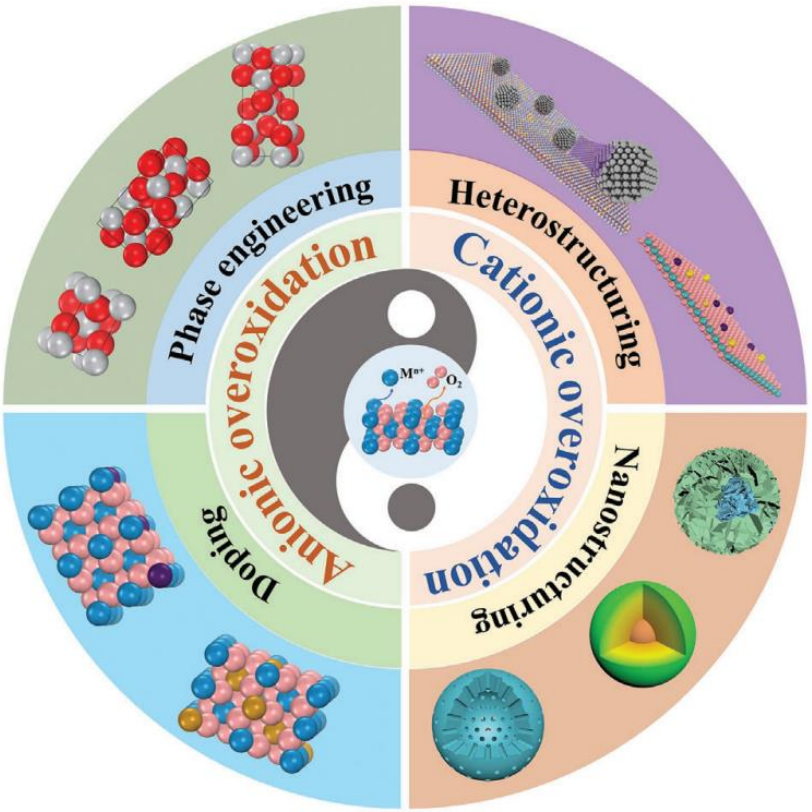
18. Xiaobo Zheng, Jiarui Yang, Xun Xu, Shixue Dou, Wenping Sun, Dingsheng Wang, **Guoxiu Wang***, “Deciphering Cationic and Anionic Overoxidation: Key Insights into the Intrinsic Structural Degradation of Catalysts”, **Advanced Energy Materials**, 14, 2401227, 2024. IF=27.8. DOI: 10.1002/aenm.202401227

**ABSTRACT:** Proton exchange membrane water electrolyzer (PEMWE) technology holds tremendous promise for large-scale green hydrogen production. However, its widespread application faces significant constraints due to the limited lifespan of the oxygen evolution reaction (OER) catalyst in highly acidic and oxidative operating environments. Therefore, a comprehensive understanding of the catalyst's structural degradation mechanism is imperative for the rational design of high-performance acidic catalysts. In this review, the essence of the structural degradation of catalysts: and irreversible cationic and anionic overoxidation is initially unveiled. This is followed by an in-depth exploration of their intricate relationship with the adsorbate evolution mechanism (AEM) and lattice oxygen oxidation mechanism (LOM). Then, state-of-the-art characterization techniques for cationic and anionic overoxidation analysis are introduced. Subsequently, 4 cutting-edge catalyst antioxidation strategies, including heterostructure engineering, doping strategy, nanostructuring, and phase engineering are systematically discussed, aiming to reveal their intrinsic factors for effectively inhibiting catalyst overoxidation. Finally, the remaining challenges and prospective insights into catalysts for PEMWE are delineated. The overarching goal of this review is to facilitate a fundamental understanding of catalyst structural degradation mechanisms and provide principal guidelines for the rational design of robust acidic OER catalysts.

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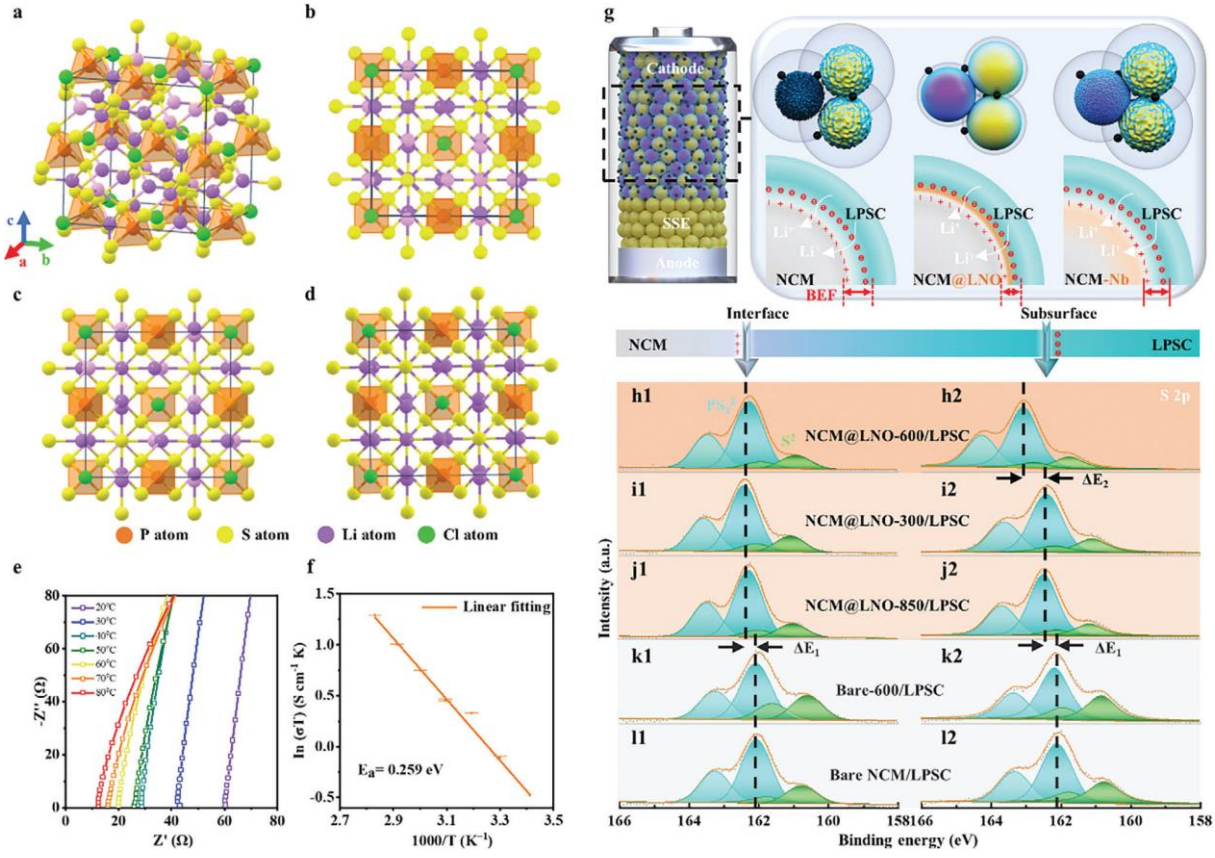
19. Ya Chen, Ling Huang, Deli Zhou, Xin Gao, Tengfei Hu, Zhiyuan Zhang, Zheng Zhen, Xiaodong Chen, Lifeng Cui, **Guoxiu Wang***, “Elucidating and Minimizing the Space-Charge Layer Effect between NCM Cathode and Li6PS5Cl for Sulfide-Based Solid-State Lithium Batteries”, **Advanced Energy Materials**, 14, 2304443, 2024. IF=27.8

**ABSTRACT:** The electrochemical performance of all-solid-state lithium batteries (ASSLBs) can be significantly improved by addressing the challenges posed by space charge layer (SCL) effect, which plays a crucial role in determining $Li^+$ ions transport kinetic at cathodic interface. Therefore, it is critical to realize the in situ inspection and visualization of SCL behaviors for solving sluggish Li+ ions transport issues, despite remaining grant challenges. Therewith, the well-defined model of $LiNbO_3$-coated NCM (NCM@LNO) cathode is constructed and assembled for the representative $Li_6PS_5Cl$-based ASSLBs, which not only ensures excellent cathodic compatibility, but also preferably enables the better monitoring of Li+ ions transport kinetics. Combining ex situ analysis with DFT calculation, the formation and evolution mechanism of SCL are comprehensively understood, and the relationship between well-controlled SCL configuration and $Li^+$ electrochemical behavior has been also further illustrated and established through the operando Raman spectroscopy. On these grounds, the preferred NCM@LNO cathodes acquire the enhanced discharge capacity of 90.6% (144.8 mAh $g^{-1}$) after 100 cycles and it can still deliver the exceptional capacity of 136.2 mAh $g^{-1}$ after 800 cycles in ASSLBs. Hence, the research will pave up a new perspective for fundamental scientific insight of the SCL and reasonable tailoring of cathodic interface for high-efficiency ASSLBs.

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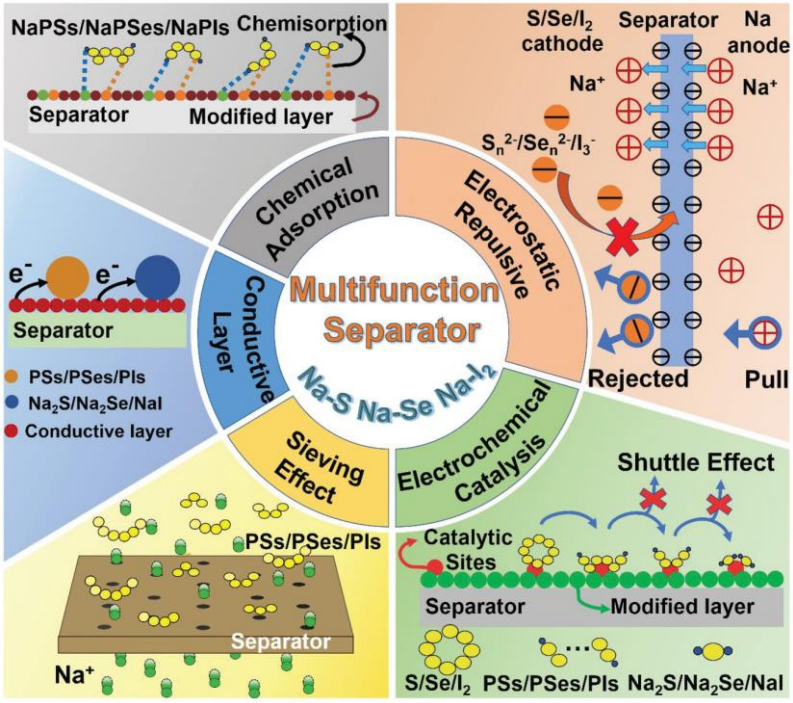
20. Jing Xu, Yashuang Qiu, Jianhao Yang, Haolin Li, Pingan Han, Yang Jin, Xiaoli Zhang, Hao Liu, Bing Sun, **Guoxiu Wang***, "Review of Separator Modification Strategies: Targeting Undesired Anion Transport in Room Temperature Sodium–Sulfur/Selenium/Iodine Batteries", **Advanced Functional Materials**,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

**ABSTRACT:** Rechargeable sodium–sulfur/selenium/iodine (Na–S/Se/$I_2$) 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/$I_2$ 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/$I_2$ 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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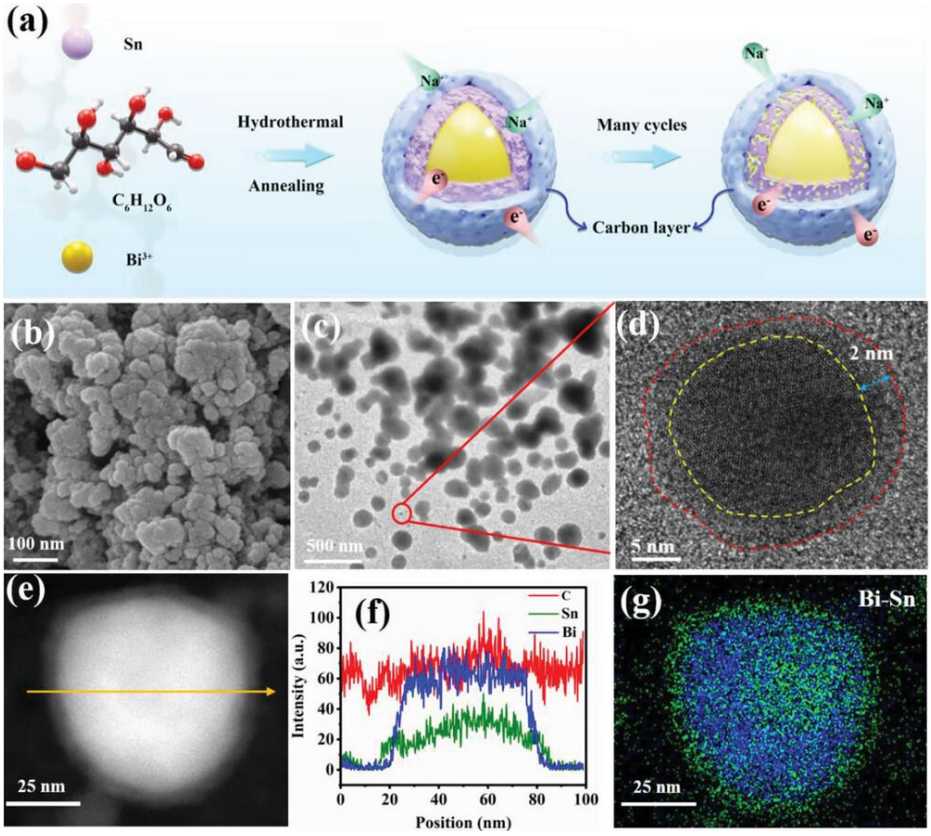
21. Jun Chen, Guilai Zhang, Jun Xiao, Jiayi Li, Yang Xiao, Dingyi Zhang, Hong Gao, Xin Guo, **Guoxiu Wang***, Hao Liu, "A Stress Self-Adaptive Bimetallic Stellar Nanosphere for High-Energy Sodium-Ion Batteries", **Advanced Functional Materials**, 33, 2307959, 2023. IF= 19. DOI: 10.1002/ adfm.202307959

**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) sneath (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 $Na_3V_2(PO_4)_3$ ‖ 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 $kg^{-1}$.

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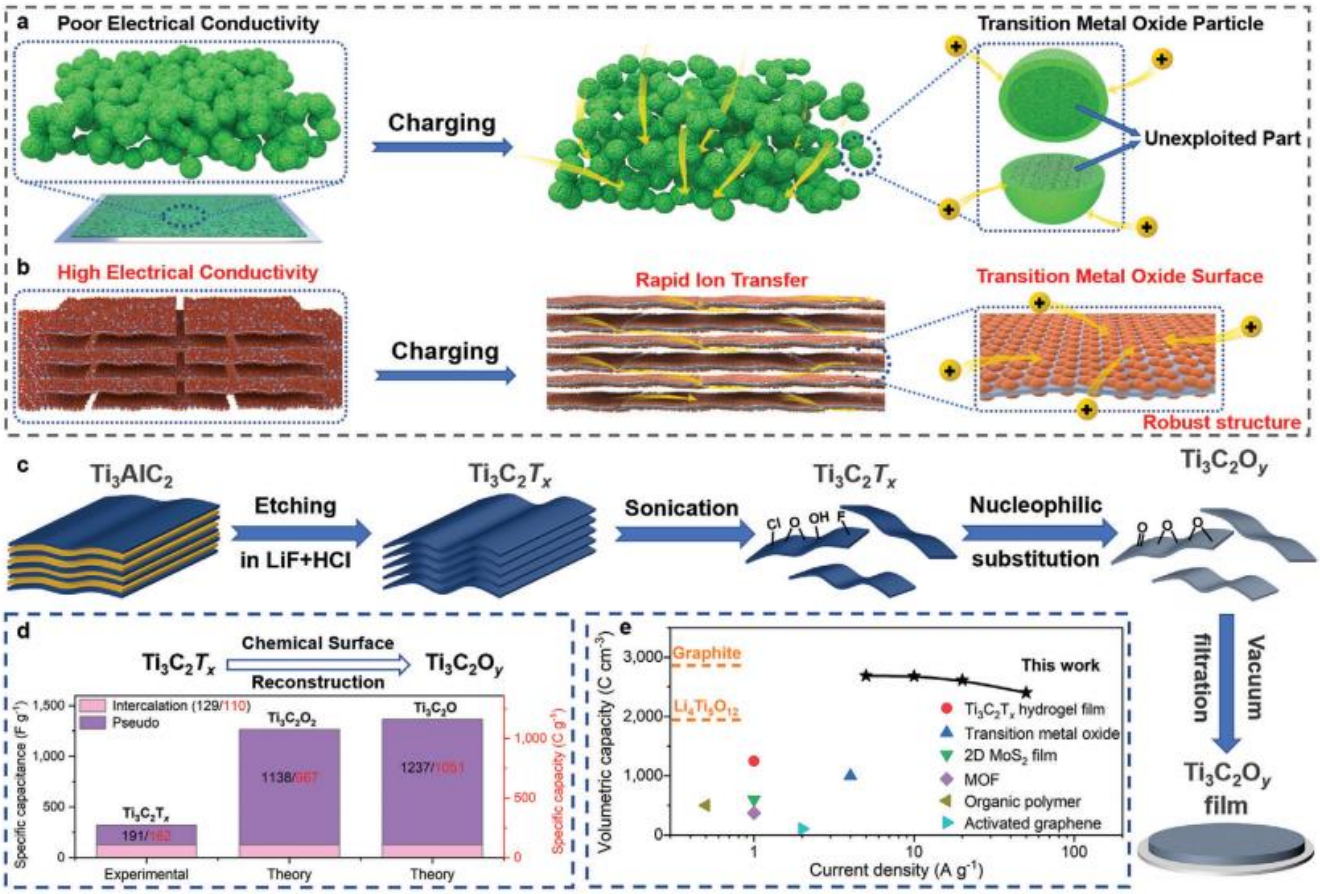
22. Jiang Xu, Ryan S Longchamps, Xi Wang, Bingqing Hu, Xude Li, Shijian Wang, Lvzhou Li, Yaokai Gu, Xiaoting Cao, Ningyi Yuan, Shanhai Ge, **Guoxiu Wang***, Jianning Ding, "Nucleophilic Substitution Enables MXene Maximum Capacitance and Improved Stability", **Advanced Functional Materials**, 34, 2408892, 2024. IF=18.5. DOI: 10.1002/adfm.202408892

**ABSTRACT:** Combining the merits of battery and supercapacitor into a single device represents a major scientific and technological challenge. From a design perspective, electrode material plays a key role in the device and the fundamental difficulty lies in incorporating a high density of active sites into a stable material with excellent charge transfer kinetics. Here, the synthesis is reported of a nearly full-oxygen-functionalized 2D conductive transition metal carbide ($Ti_3C_2O_y$) with ultrahigh density of Ti-O/-O redox-active sites by nucleophilic substitution and in situ oxidation under the presence of a proper electrophilic reagent ($K^+$). The fabricated electrode delivered exceptionally high gravimetric and volumetric capacitance (1,082 F $g^{-1}$ and 3,182 F $cm^{-3}$ in a potential window of 0.85 V, approximating the theoretical capacity of many transition metal oxides), fast charging/discharging in tens of seconds across a wide range of temperature (−70 to 60 °C), and excellent structural and chemical stability. These promising results provide avenues for the development of high-energy, high-power storage devices as well as electromagnetic shielding, and electronic and optoelectronic devices.

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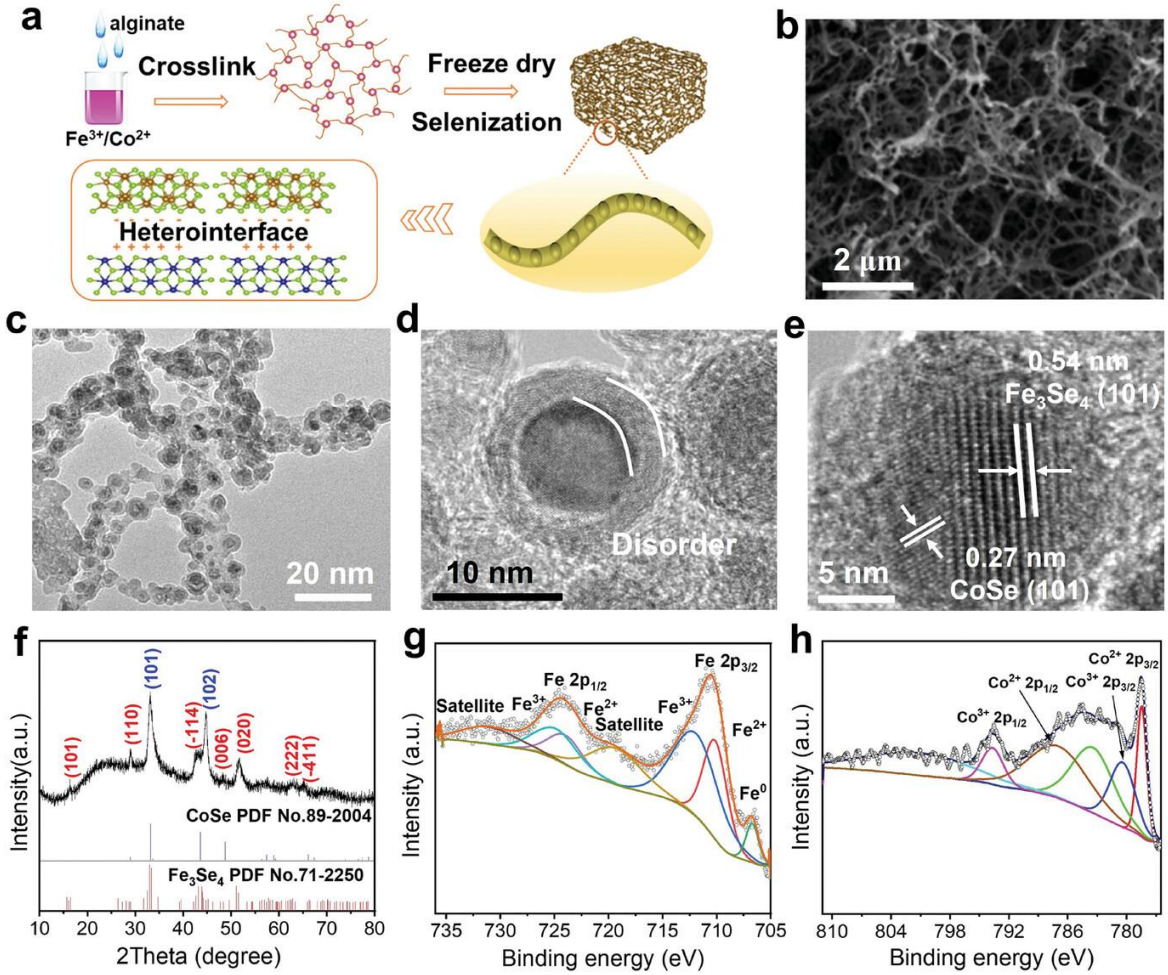
23. Chunrong Ma, Xiao Tang, Haoxi Ben, Wei Jiang, Xinyu Shao, **Guoxiu Wang***, Bing Sun, "Promoting Reaction Kinetics and Boosting Sodium Storage Capability via Constructing Stable Heterostructures for Sodium-Ion Batteries", **Advanced Functional Materials**, 34, 2412879, 2024. IF=18.5. DOI: 10.1002/adfm.202412879

**ABSTRACT:** Constructing heterostructures containing multiple active components is proven to be an efficient strategy for enhancing the sodium storage capability of anode materials in sodium-ion batteries (SIBs). However, performance enhancement is often attributed to the unclear synergistic effects among the active components. A comprehensive understanding of the reaction mechanisms on the interfaces at the atomic level remains elusive. Herein, the carbon-coated $Fe_3Se_4$/CoSe (Fe3Se4/CoSe-C) anode material as a model featuring atomic-scale contact interfaces is synthesized. This unique heterogeneous architecture offers an adjustable electronic structure, which facilitates rapid reaction kinetics and enhances structural integrity. In situ microscopic and ex situ spectral characterization techniques, along with theoretical simulations, confirm that the heterointerface with strong electric fields promotes $Na^+$ ion migration. Based on solid-state nuclear magnetic resonance (NMR) analysis, an interface charge storage mechanism is revealed, resulting in the enhanced specific capacity of the anode materials. When employed as an anode in SIBs, the Fe3Se4/CoSe-C electrode demonstrates excellent rate capabilities (218 mAh $g^{-1}$ at 7 A $g^{-1}$) and prolonged cycling stability (258 mAh $g^{-1}$ at 5 A $g^{-1}$ after 1000 cycles). This work highlights the significance of heterointerface engineering in electrode material design for rechargeable batteries.

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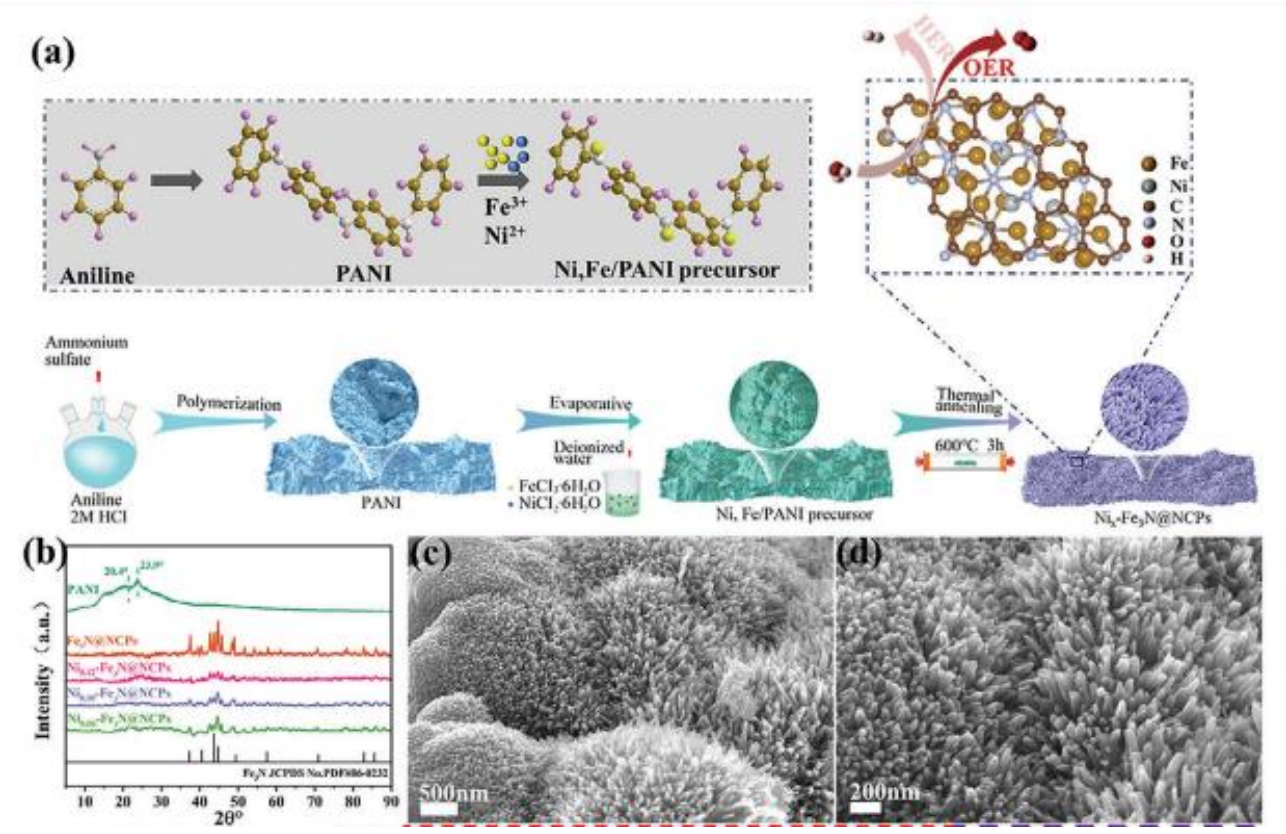
24. Gang Wang, Wenshuai Tang, Ya Chen, Peiyi Ji, Mingxia Lu, Hongliang Wei, Lifeng Cui, Xiaodong Chen, Guoxiu Wang, "The Ni Heteroatom-Induced Electronic Structure Tailoring of Ultrastable $Fe_3N$@ NCPs Nanosheets Electrocatalyst for Boosting Alkaline Seawater Electrolysis", **Advanced Functional Materials**, 34, 2404470, 2024. IF=18.5. DOI: 10.1002/adfm.202404470

**ABSTRACT:** The exploration of high-efficiency electrocatalysts is becoming indispensable for the production of H2 from electrochemcial seawater splitting. Herein, a feasible strategy is developed to realize the in situ encapsulation of Ni-decorated Fe3N into porous N-doped carbon nanolayer, abbreviated as Nix-$Fe_3N$@NCPs, by an association of wet-impregnation treatment and thermal annealing. More concretely, the doping of Ni can initiate the synergistic effect to optimize the internal electronic structure, which strengthen the adsorption of intermediates and interface charge transfer. Meantime, the architecture of NCPs encapsulation nanolayer not only enhances the conductivity and the structural stability, but also effectively prevents $Cl^-$ ions transport from poisoning of electrocatalytic active sites. On these grounds, the preferred $Ni_{0.10}$-$Fe_3N$@NCPs electrocatalyst delivers the exceptional bifunctional electrocatalytic performance in electrolytic seawater. The corresponding HER and OER overpotentials to attain the current densities of 10, 100, and 500 mA cm−2 are only required 47, 147, and 291 mV as well as 152, 249, and 312 mV, respectively. In addition, the Ni0.10-$Fe_3N$@NCPs electrocatalysts achieves an ultra-low voltage of 1.80 V at 500 mA $cm^{-2}$ with ultralong lifespan of 1200 h in zero-gap alkaline electrolyzer. This work provides a cutting-edge scientific research exploration for the development of electrocatalyst for seawater electrolysis.

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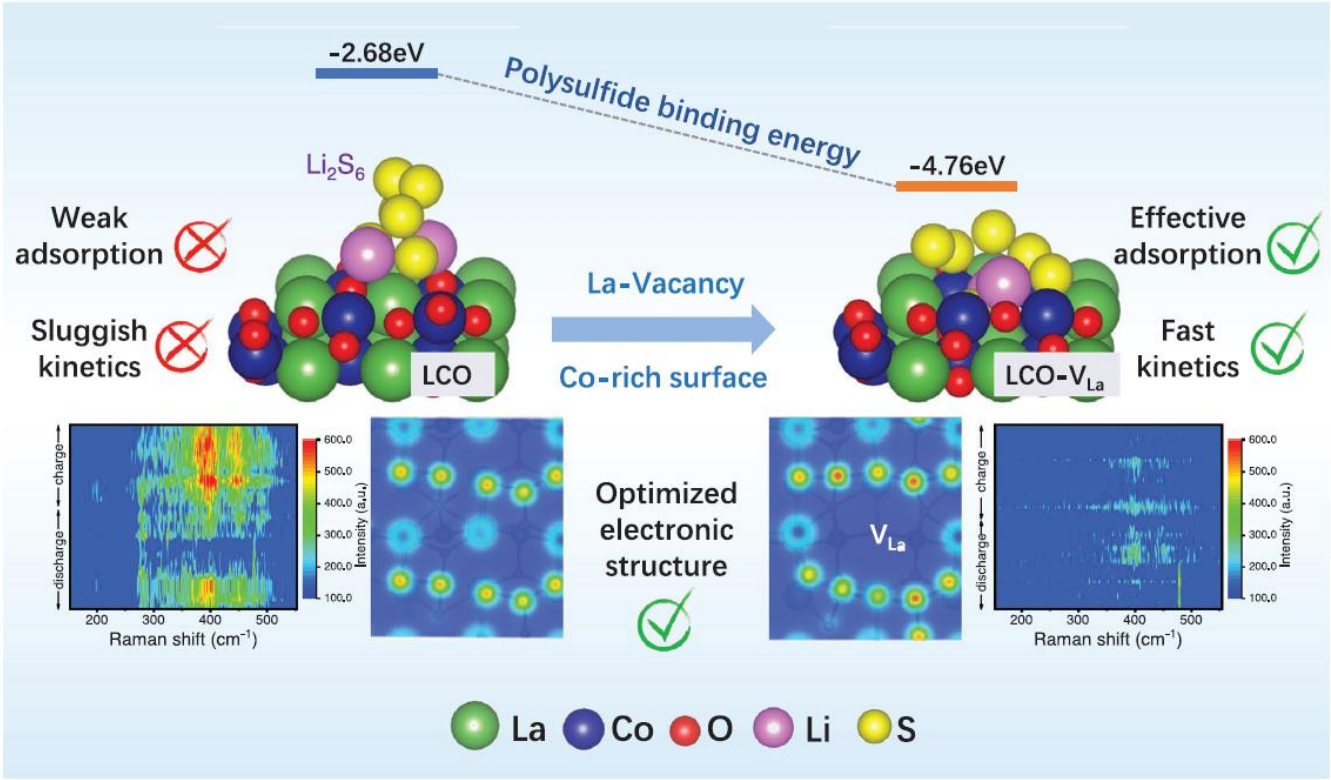
25. Zhe Bai, Zhenhua Wang, Tan Wang, Zeyu Wu, Xiaotian Gao, Yu Bai, **Guoxiu Wang***, Kening Sun, “Cation-Vacancy Engineering Modulated Perovskite Oxide for Boosting Electrocatalytic Conversion of Polysulfides”, **Advanced Functional Materials**, 34, 2419105, 2024. IF=18.5. DOI: 10.1002/ adfm.202419105.

**ABSTRACT:** Lithium-sulfur batteries face challenges like polysulfide shuttle and slow conversion kinetics, hindering their practical applications in renewable energy storage and electric vehicles. Herein, a solution to solve this issue is reported by using a cation vacancy engineering strategy with rational synthesis of La-deficient LaCoO3 (LCO-VLa). The introduction of cation vacancies in LCO-VLa modifies the geometric structure of coordinating atoms, exposing Co-rich surface with more catalytically active surfaces. Meanwhile, the d-band center of LCO-VLa shifts toward the Fermi level, enhancing polysulfide adsorption. Furthermore, multivalent cobalt ions ($Co^{3+}$/$Co^{4+}$) induced by charge compensation enhance the electrical conductivity of LCO-VLa, accelerating electron transfer processes and improving catalytic performance. Theoretical calculations and experimental characterizations demonstrate that La-deficient LCO-VLa effectively suppresses the polysulfide shuttle, reduces the energy barrier for polysulfide conversion, and accelerates redox reaction kinetics. LCO-VLa-based batteries demonstrate exceptional rate performance and cycling stability, retaining 70% capacity after nearly 500 cycles at 1.0 C, with a minimal decay rate of 0.055% per cycle. These findings highlight the significance of cation vacancy engineering for exploring precise structure-activity relationships during polysulfides conversion, facilitating the rational design of catalysts at the atomic level for lithium-sulfur batteries.

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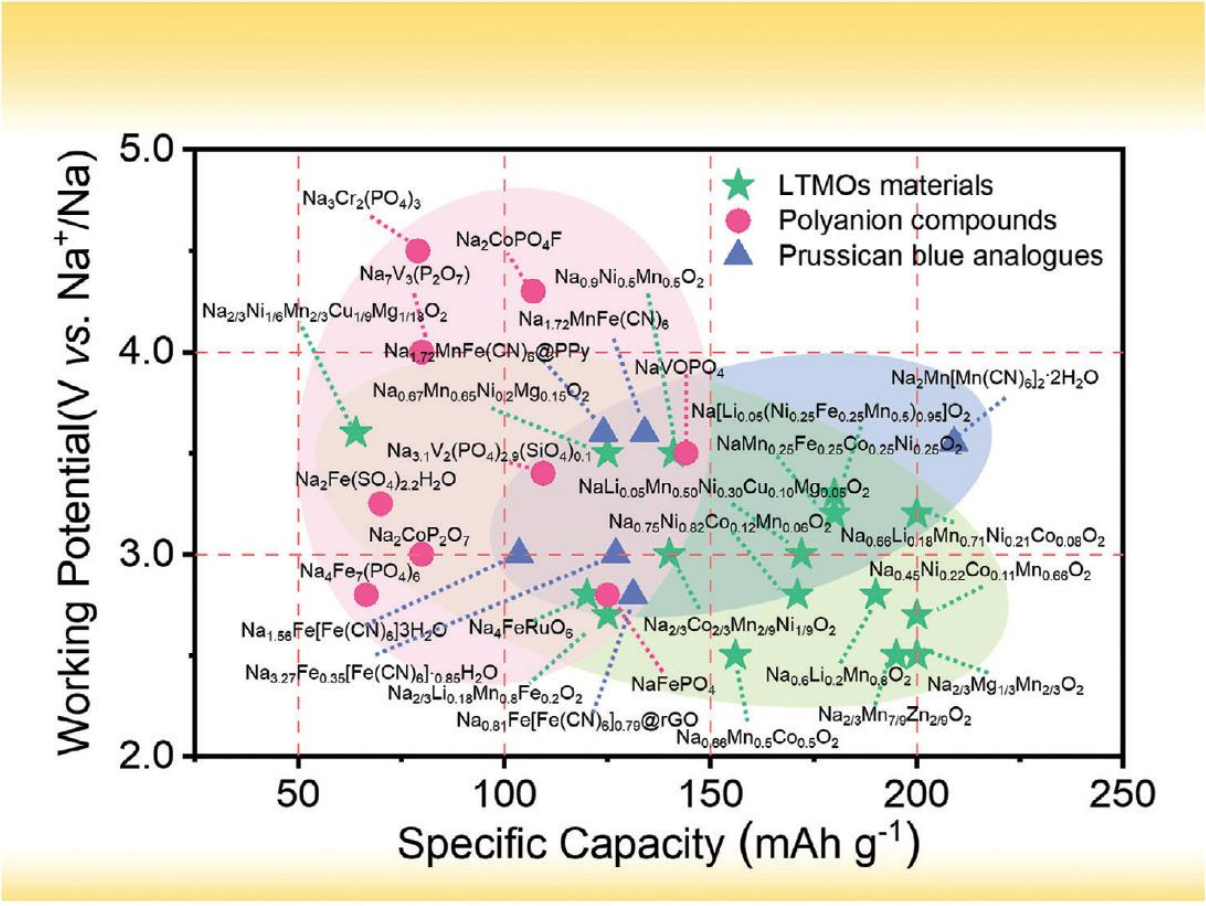
26. Lei Wang, Leilei Wang, Haichao Wang, Hanghang Dong, Weiwei Sun, Li-Ping Lv, Chao Yang, Yao Xiao, Feixiang Wu, Yong Wang, Shulei Chou, Bing Sun, **Guoxiu Wang***, Shuangqiang Chen, "Progress and Perspective of High-Entropy Strategy Applied in Layered Transition Metal Oxide Cathode Materials for High-Energy and Long Cycle Life Sodium-Ion Batteries", **Advanced Functional Materials**, 34, 2405122, 2024. IF=18.5. DOI: 10.1002/adfm.202405122.

**ABSTRACT:** Layered transition metal oxide (LTMO) cathode materials of sodium-ion batteries (SIBs) have shown great potential in large-scale energy storage applications owing to their distinctive periodic layered structure and 2D ion diffusion channels. However, several challenges have hindered their widespread application, including phase transition complexities, interface instability, and susceptibility to air exposure. Fortunately, an impactful solution has emerged in the form of a high-entropy doping strategy employed in energy storage research. Through the implementation of high-entropy doping, LTMOs can overcome the aforementioned limitations, thereby elevating LTMO materials to a highly competitive and attractive option for next-generation cathodes of SIBs. Thus, a comprehensive overview of the origins, definition, and characteristics of high-entropy doping is provided. Additionally, the challenges associated with LTMOs in SIBs are explored, and discussed various modification methods to address these challenges. This review places significant emphasis on conducting a thorough analysis of the research advancements about high-entropy LTMOs utilized in SIBs. Furthermore, a meticulous assessment of the future development trajectory is undertaken, heralding valuable research insights for the design and synthesis of advanced energy storage materials.

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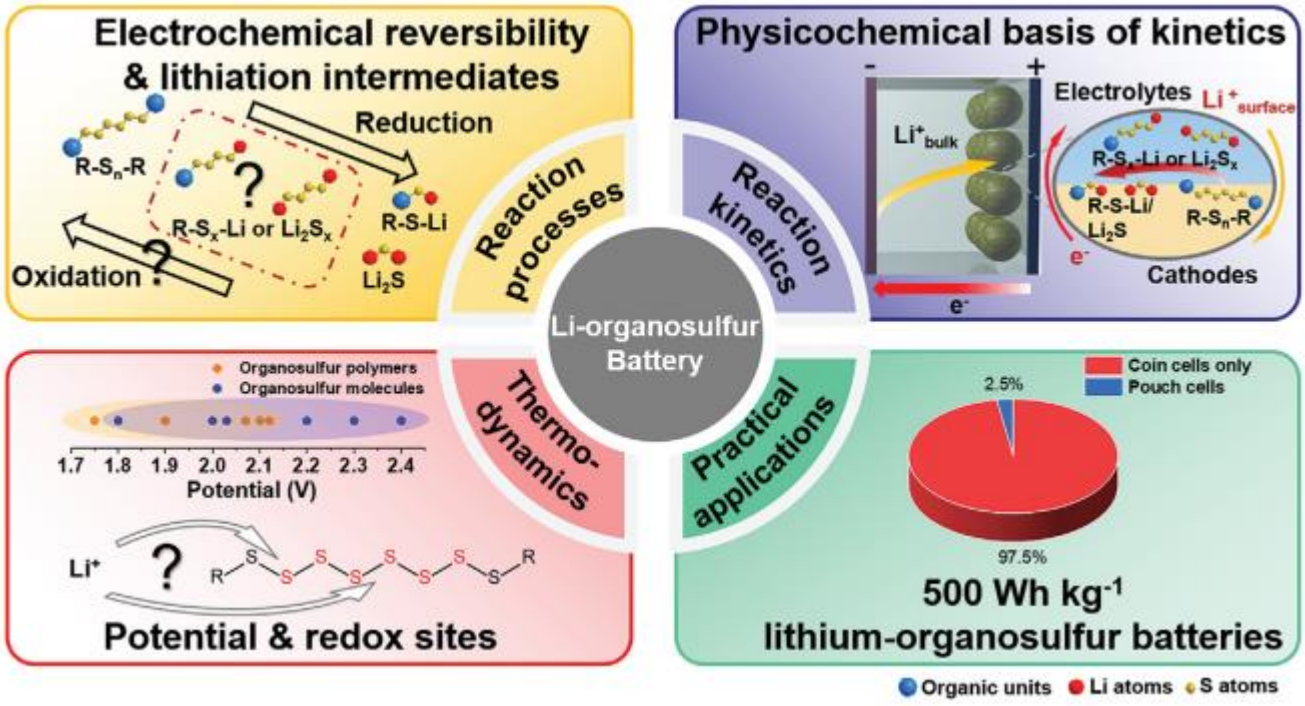
27. Xiaoyin Zhang, Tong Yu, Shuaiyi Yang, Zhuoyan Qu, Ru Xiao, **Guoxiu Wang*,** Zhenhua Sun, Feng Li, "Organosulfur Cathodes in Lithium Metal Batteries: Bridging the Gap between Fundamentals and Practical Applications", **Advanced Functional Materials**, 34, 2405122, 2024. IF=18.5. DOI: 10.1002/adfm.202405122.

**ABSTRACT:** High-specific energy sulfur-based cathodes have attracted considerable interest in lithium batteries. Organosulfur cathodes offer inherent advantages of high element abundance and an extended cycling life, aligning with the evolving requirements of future energy storage devices. Over the past decade, research efforts have been devoted to optimizing electrochemical performance through the rich and tunable molecular structures of organosulfur compounds. To further advance the fundamental research and practical application of lithium-organosulfur batteries, a systematical analysis of the correlation between the molecular structures and electrochemical mechanisms of organosulfur cathodes is imperative. This involves deriving the key parameters at the cell level and investigating the feasibility. In this review, the thermodynamics, reaction processes, and electrochemical kinetics of organosulfur cathodes, grounded in fundamental theories of electrochemistry and materials science are discussed. Expanding the insights, comparisons among elemental sulfur, organosulfur, and n-type organic cathodes (e.g., carbonyl cathodes) are drawn. The gap between fundamentals and practical applications targeting 500 Wh $kg^{-1}$ lithium organosulfur batteries is highlighted through energy density calculations and identification of key factors affecting pouch cells. Finally, potential strategies and prospects for the overall design of advanced lithium-organosulfur batteries are proposed, considering both theoretical foundations and practical implementations.

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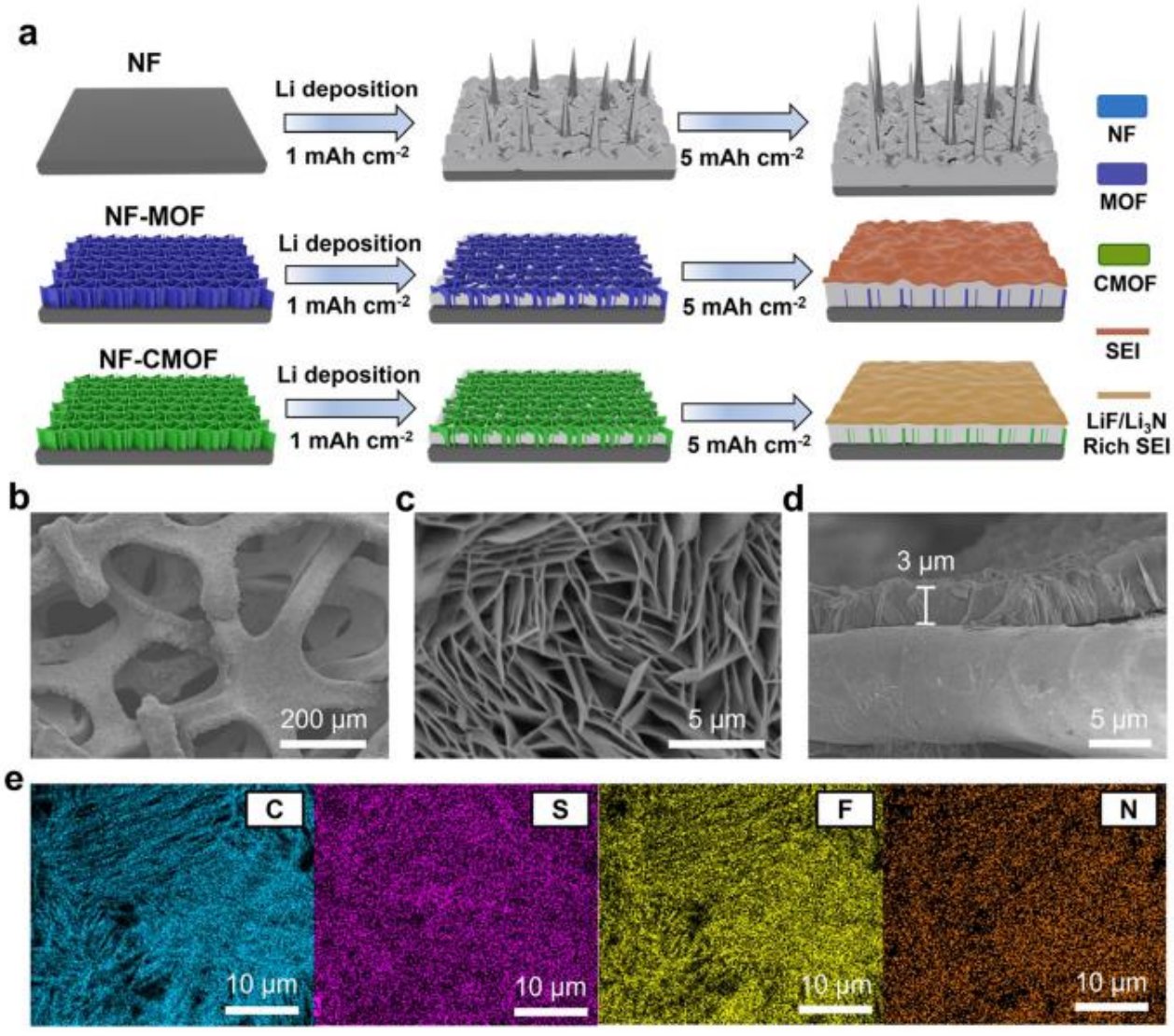
28. Le Pang, Jiahui Lu, Yongyue Yu, Dongfang Li, Yong Chen, Shijian Wang, Yan Wang, Bing Sun, Hongxia Wang, **Guoxiu Wang*, 2024**, "Cationic Metal–Organic Framework Arrays to Enable Dendrite-Free Lithium Metal Anodes", **ACS Energy Letters**, **9** (8), 3746-3753, 2024. IF=19.3. DOI: 10.1021/acsenergylett.4c01345

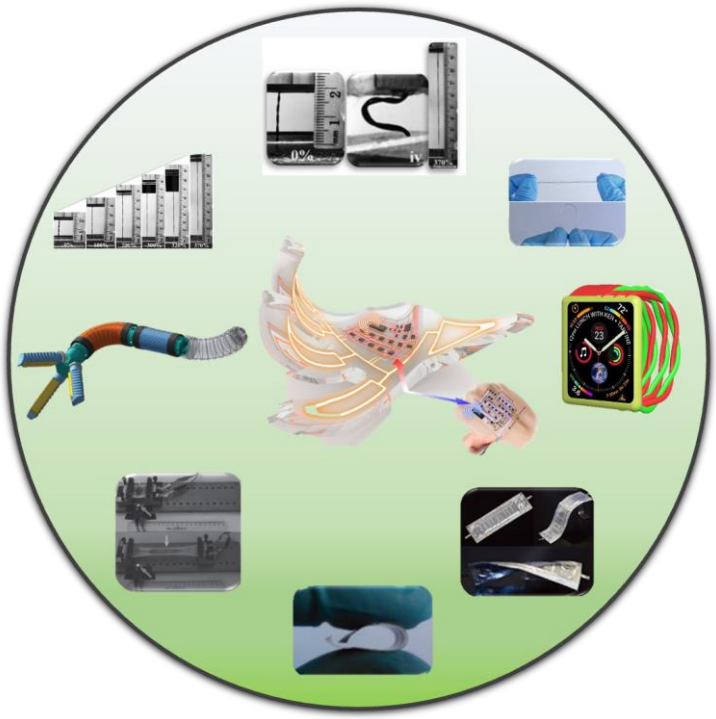
**ABSTRACT:** The severe safety issues caused by uncontrolled lithium (Li) dendrite growth hinder the practical application of Li metal anodes. Herein, we designed cationic metal–organic framework arrays on nickel foam (NF–CMOF) as a multifunctional current collector for dendrite-free Li metal anodes. The cationic MOF (CMOF) was synthesized by grafting bis(trifluoromethane)sulfonimidate (TFSI) functional groups on the MOF nanosheets via a facile hydrothermal method. The high surface area, good lithiophilicity, and positively charged properties of CMOF nanosheets effectively facilitate uniform Li deposition. In addition, the porous array structure acts as the host to accommodate deposited Li at a low areal capacity. Furthermore, the CMOF with grafted TFSI groups induces the formation of a LiF/$Li_3N$-rich solid electrolyte interphase, which further guarantees uniform Li deposition at high areal capacity. The full cells with Li predeposited NF–CMOF (Li@NF–CMOF) anodes maintained a high-capacity retention over 1000 cycles and achieved an enhanced rate capability at 10 C.




https://pubs.acs.org/doi/10.1021/acsenergylett.4c01345

29. 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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