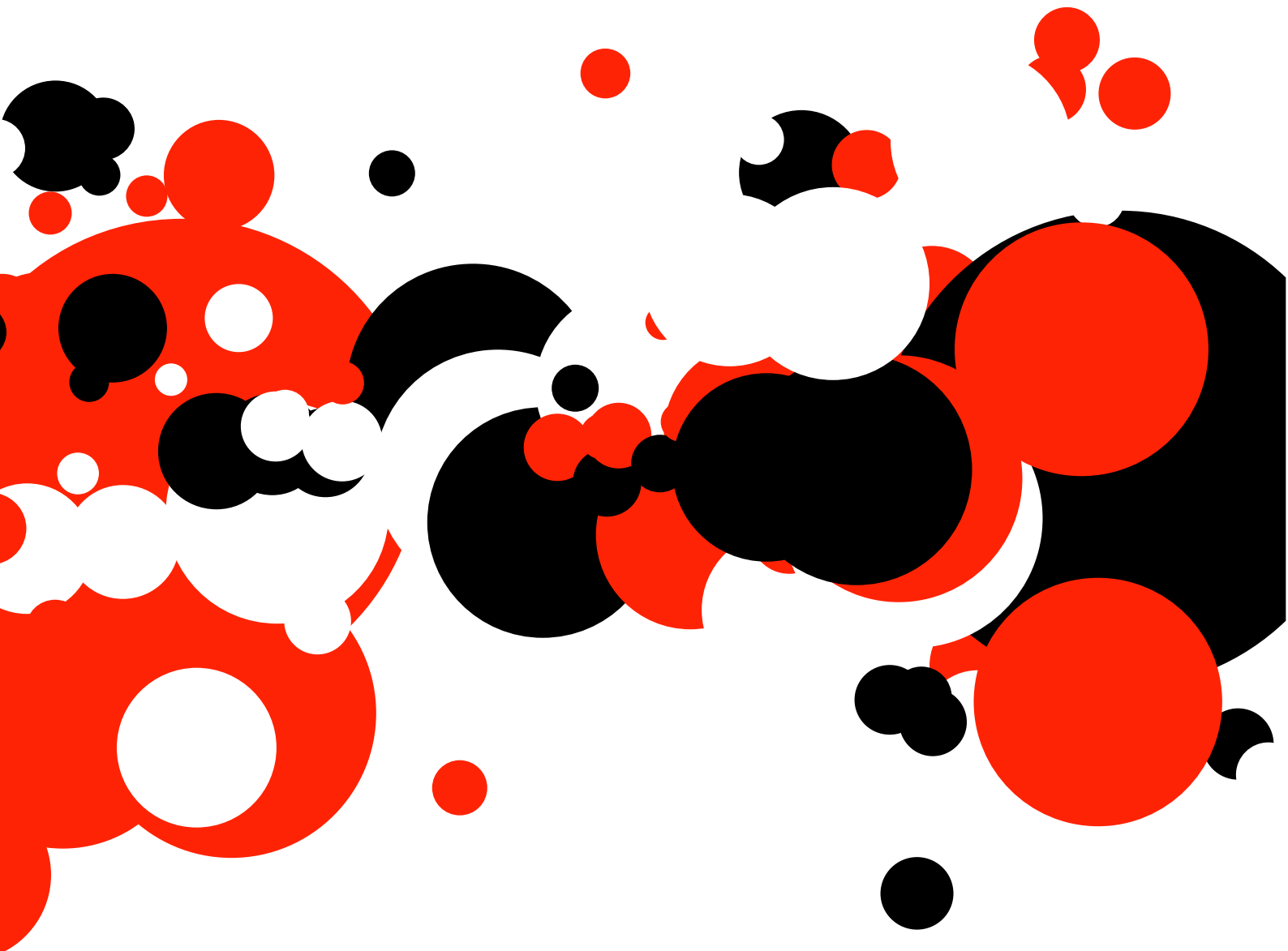




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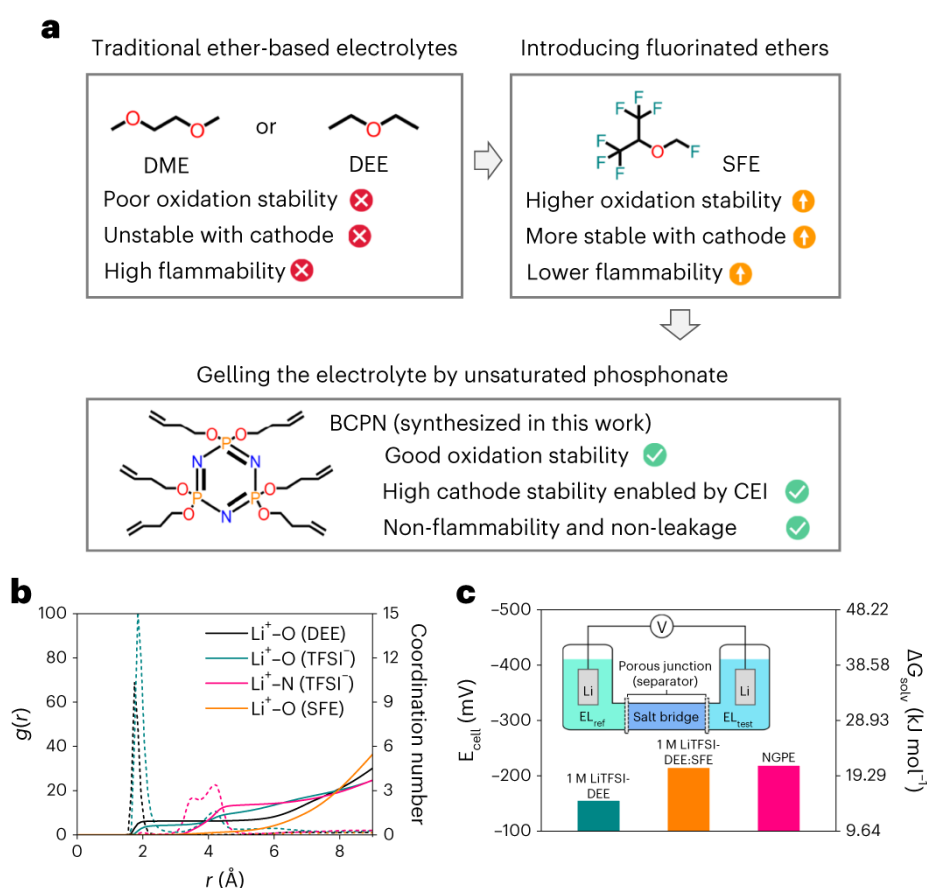
Centre for Clean Energy Technology

Research Highlights in 2023



1. Yuefeng Meng, Dong Zhou, Ruliang Liu, Yao Tian, Yifu Gao, Yao Wang, Bing Sun, Feiyu Kang, Michel Armand, Baohua Li, **Guoxiu Wang***, Doron Aurbach, **2023**, “Designing phosphazene-derivative electrolyte matrices to enable high-voltage lithium metal batteries for extreme working conditions”, **Nature Energy**, 2023. IF=67.439. DOI: 10.1038/s41560-023-01339-z

ABSTRACT: The current high-energy lithium metal batteries are limited by their safety and lifespan owing to the lack of suitable electrolyte solutions. Here we report a synergy of fluorinated co-solvent and gelation treatment by a butenoxycyclotriphosphazene (BCPN) monomer, which facilitates the use of ether-based electrolyte solutions for high-energy lithium metal batteries. We show that the safety risks of fire and electrolyte leakage are eliminated by the fluorinated co-solvent and fireproof polymeric matrices. The compatibility with high-energy cathodes is realized by a well-tailored Li^+ solvation sheath, along with BCPN-derived protective surface films developed on the cathodes. Our $\text{Li} \parallel \text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ cells reach high-capacity retention, superior low-temperature performance, good cyclability under high pressure and steady power supply under abusive conditions. Our electrolyte design concept provides a promising path for high energetic, durable, and safe rechargeable Li batteries.



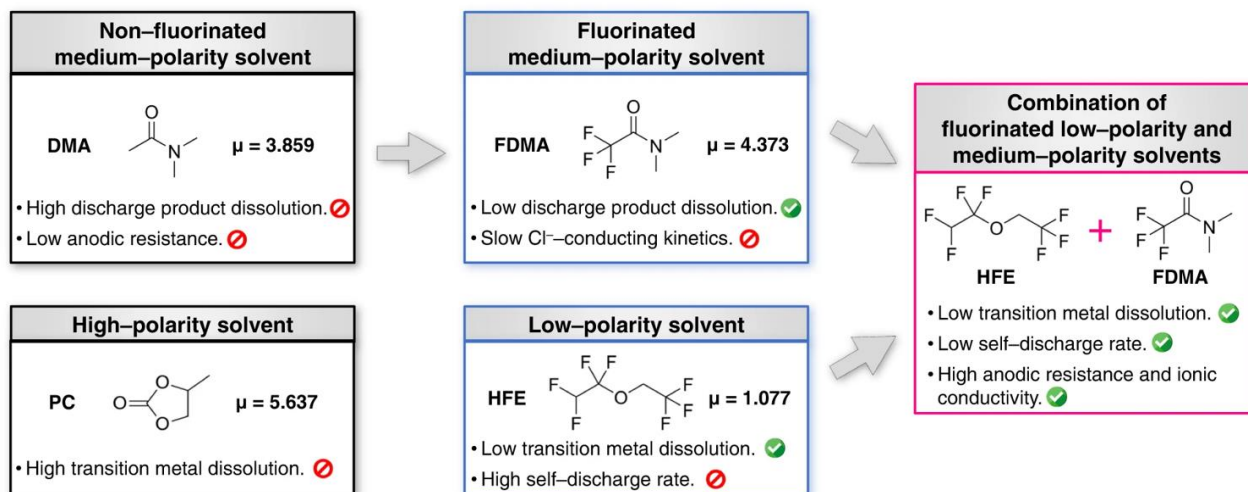
<https://www.nature.com/articles/s41560-023-01339-z>

<https://mp.weixin.qq.com/s/JKUucEPwKeNAc5xWweqcoQ>

2. Xu Yang, Xu Yang, Bao Zhang, Yao Tian, Yao Wang, Zhiqiang Fu, Dong Zhou, Hao Liu, Feiyu Kang, Baohua Li, Chunsheng Wang and **Guoxiu Wang***, “Electrolyte design principles for developing quasi-solid-state rechargeable halide-ion batteries”, **Nature Communications**, 14, 925, 2023. IF= 17.69. DOI: 10.1038/s41467-023-36622-w.

ABSTRACT: Rechargeable halide-ion batteries (HIBs) are good candidates for large-scale due to their appealing energy density, low cost, and dendrite-free features. However, state-of-the-art electrolytes limit the HIBs’ performance and cycle life. Here, via experimental measurements and modelling approach, we demonstrate that the dissolutions in the electrolyte of transition metal and elemental halogen from the positive electrode and discharge products from the negative electrode cause the HIBs failure. To circumvent these issues, we propose the combination of fluorinated low-polarity solvents with a gelation treatment to prevent dissolutions at the interphase, thus, improving the HIBs’ performance. Using this approach, we develop a quasi-solid-state Cl-ion-conducting gel polymer electrolyte. This electrolyte is tested in a single-layer pouch cell configuration with an iron oxychloride-based positive electrode and a lithium metal negative electrode at 25 °C and 125 mA g⁻¹. The pouch delivers an initial discharge capacity of 210 mAh g⁻¹ and a discharge capacity retention of almost 80% after 100 cycles. We also report assembly and testing of fluoride-ion and bromide-ion cells using quasi-solid-state halide-ion-conducting gel polymer electrolyte.

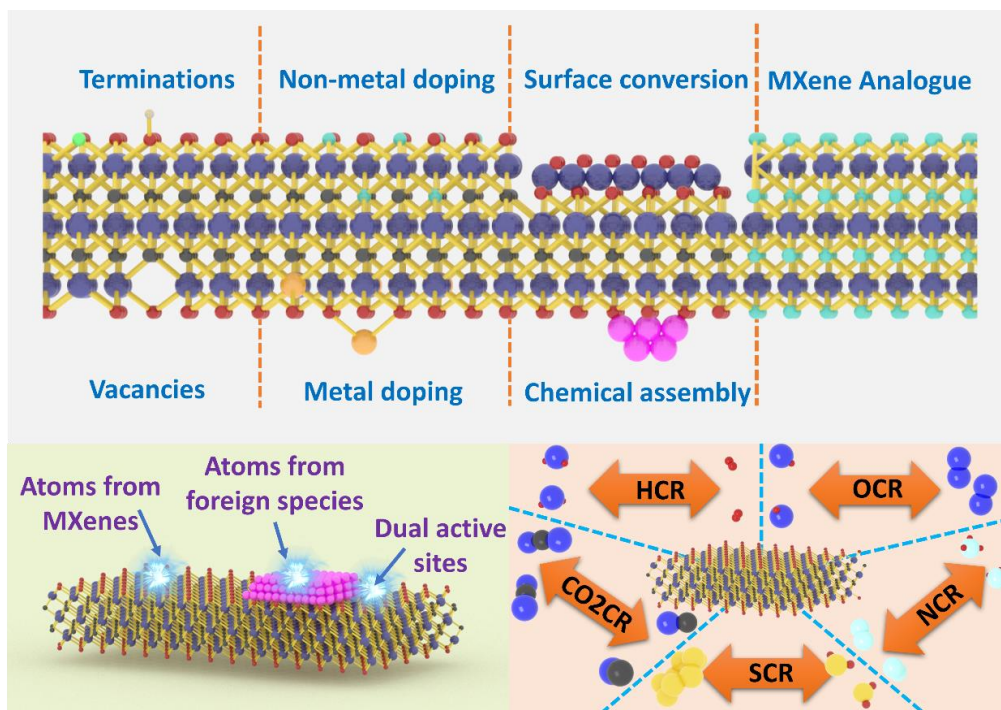
Electrolyte design flow



<https://www.nature.com/articles/s41467-023-36622-w>

3. Yufei Zhao, Jinqiang Zhang, Xin Guo, Xianjun Cao, Shijian Wang, Hao Liu, and **Guoxiu Wang***, “Engineering strategies and active site identification of MXene-based catalysts for electrochemical conversion reactions”, **Chemical Society Review**, 52, 3215-3264, 2023. IF=54.56. DOI: 10.1039/d2cs00698g

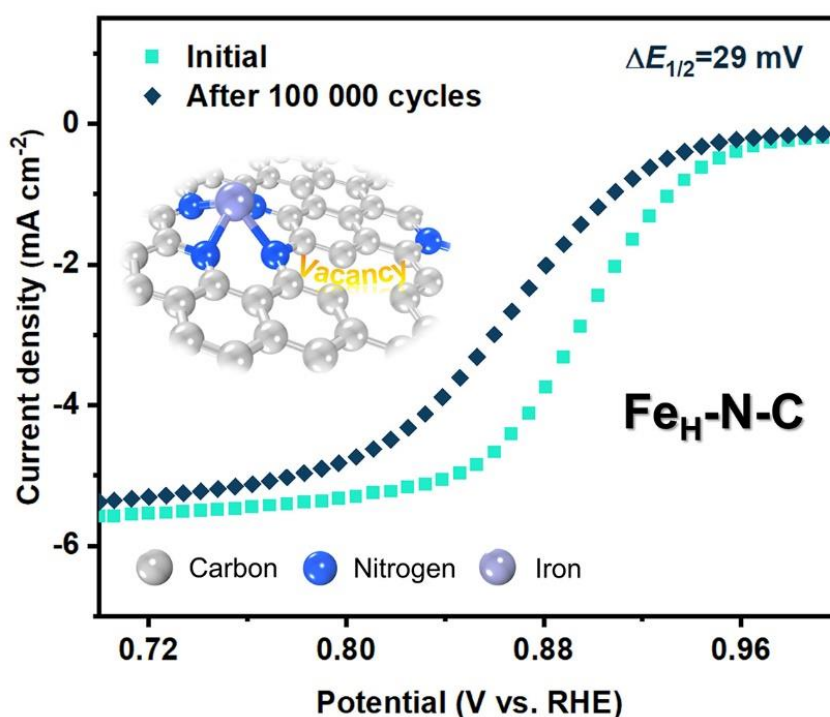
ABSTRACT: MXenes have been extensively studied due to their high metallic conductivity, hydrophilic properties, tunable layer structure and attractive surface chemistry, making them highly desirable for energy-related applications. However, slow catalytic reaction kinetics and limited active sites have severely impeded their further practical applications. Surface engineering of MXenes has been rationally designed and investigated to regulate their electronic structure, increase the density of active sites, optimize the binding energy, and thus boost the electrocatalytic performance. In this review, we comprehensively summarized the surface engineering strategies of MXenes nanostructures, including surface termination engineering, defect engineering, heteroatom doping engineering (metal or non-metal), secondary material engineering, and extension to MXene analogues. By identifying the roles of each component in the engineered MXenes at the atomic level, their intrinsic active sites have been discussed to establish the relationships between the atomic structures and catalytic activities. We highlighted the state-of-the-art progress of MXenes in electrochemical conversion reactions including hydrogen, oxygen, carbon dioxide, nitrogen and sulfur conversion reactions. The challenges and perspectives of MXenes-based catalysts for electrochemical conversion reactions are presented to inspire more efforts toward the understanding and development of MXene-based materials to meet the ever-growing demand for a sustainable future.



<https://pubs.rsc.org/en/content/articlelanding/2023/CS/D2CS00698G>

4. Hao Tian, Ailing Song, Peng Zhang, Kaian Sun, Jingjing Wang, Bing Sun, Qiaohui Fan, Guangjie Shao, Chen Chen, Hao Liu, Yadong Li, **Guoxiu Wang***, “High durability of Fe-N-C single atom catalysts with carbon vacancies towards oxygen reduction reaction in alkaline media”, **Advanced Materials**, 35, 2210714, 2023. IF= 30.85. DOI: 10.1002/adma.202210714.

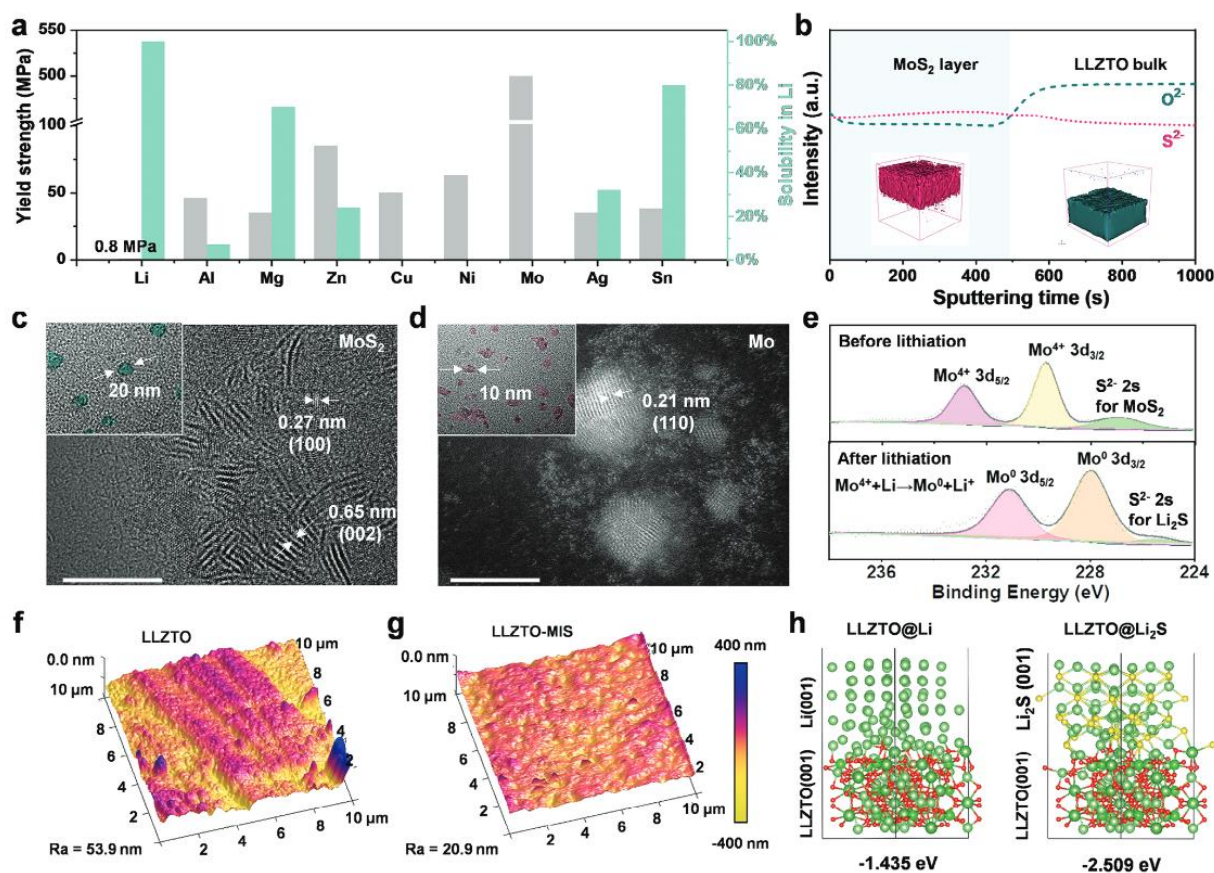
ABSTRACT: Single atom catalysts (SACs) have attracted extensive interest to catalyze the oxygen reduction reaction (ORR) in fuel cells and metal-air batteries. However, the development of SACs with high selectivity and long-term stability is a great challenge. In this work, we practically designed and synthesized carbon vacancy modified Fe-N-C SACs ($\text{Fe}_\text{H}\text{-N-C}$) through microenvironment modulation, achieving high-efficient utilization of active sites and optimization of electronic structures. The $\text{Fe}_\text{H}\text{-N-C}$ catalyst exhibited a half-wave potential ($E_{1/2}$) of 0.91 V and sufficient durability of 100,000 voltage cycles with 29 mV $E_{1/2}$ loss. Density functional theory calculations confirmed the vacancies around metal- N_4 sites can reduce the adsorption free energy of OH^* , and hinder the dissolution of metal centre, significantly enhancing the ORR kinetics and stability. Accordingly, $\text{Fe}_\text{H}\text{-N-C}$ SACs presented a high-power density and long-term stability over 1200 h in rechargeable zinc-air batteries. This work will not only guide for developing highly active and stable SACs through rational modulation of metal- N_4 sites, but also provide an insight into the optimization of the electronic structure to boost electro-catalytical performances.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.202210714>

5. 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.

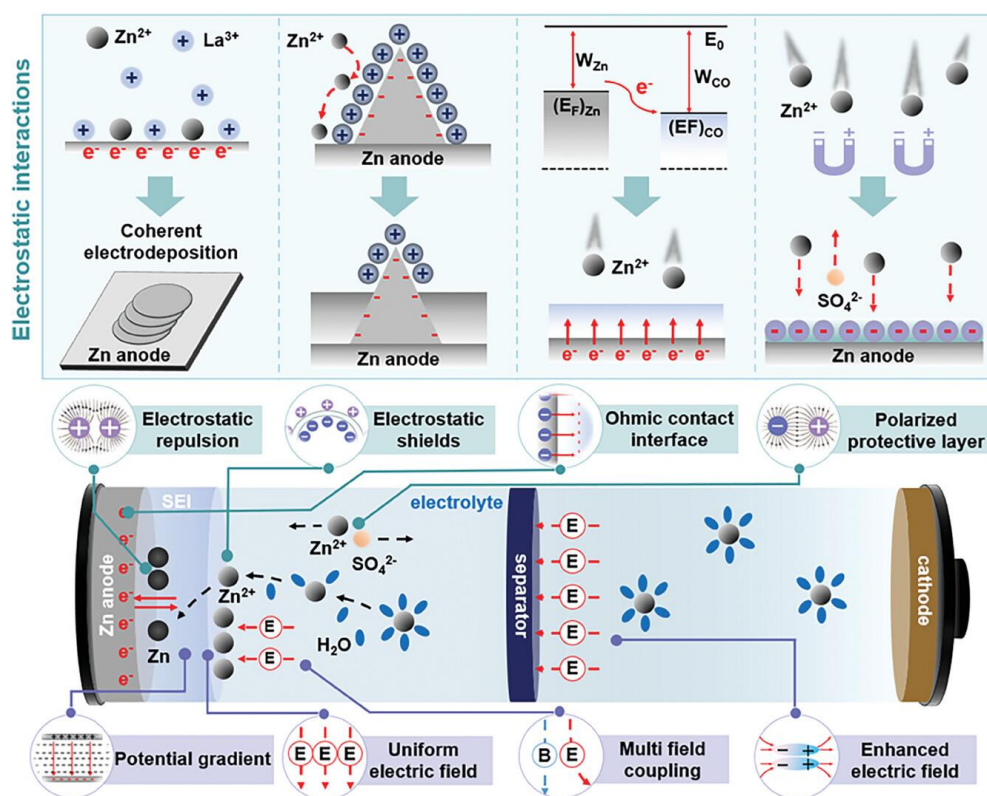
ABSTRACT: Lithium garnets are considered as promising solid-state electrolytes for next-generation solid-state Li metal batteries (SSLBs). However, the Li intrusion driven by external stack pressure triggers premature of Li metal batteries. Herein, for the first time, an in situ constructed interfacial 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>

6. Jing Xu, Haolin Li, Yang Jin, Dong Zhou, Bing Sun, Michel Armand, **Guoxiu Wang***, “Comprehensive Understanding the Roles of Electrical Mechanisms in Aqueous Zinc-ion Batteries: from Electrostatic Interactions to Electric Fields and Potentials”, **Advanced Materials**, 35, 2309726, 2023. IF= 30.85. 10.1002/ adma.202309726.

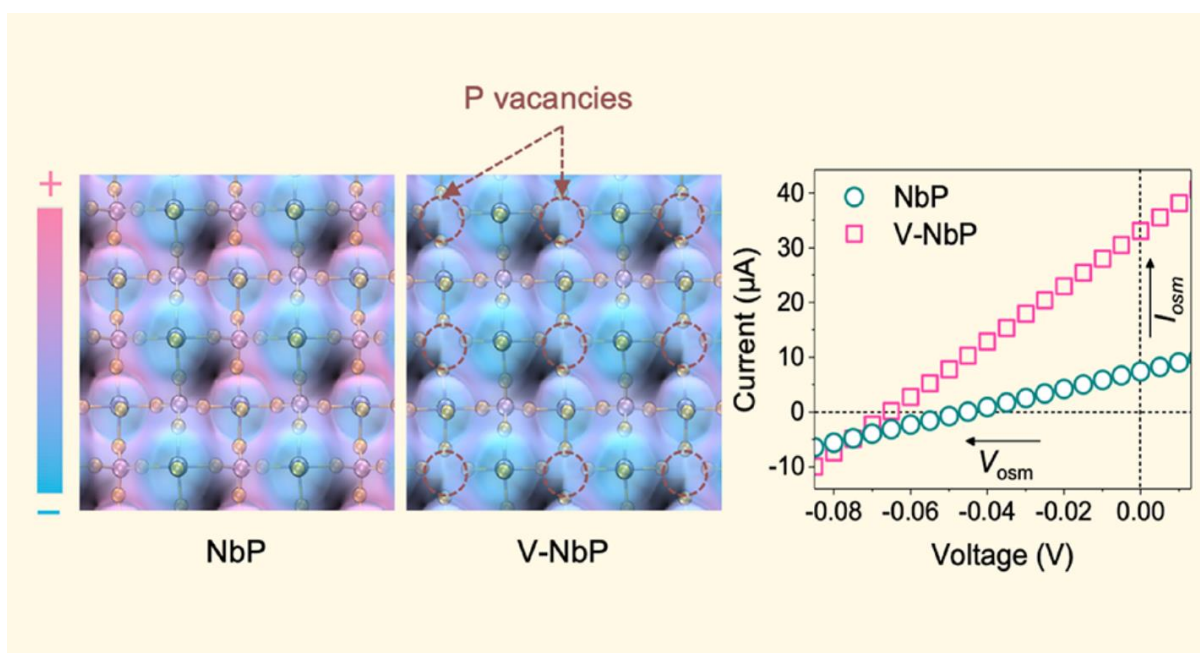
ABSTRACT: Aqueous Zn metal batteries are considered as competitive candidates for next-generation energy storage systems due to their excellent safety, low cost, and environmental friendliness. However, the inevitable dendrite growth, severe hydrogen evolution, surface passivation, and sluggish reaction kinetics of Zn metal anodes hinder the practical application of Zn metal batteries. Detailed summaries and prospects have been reported focusing on the research progress and challenges of Zn metal anodes, including electrolyte engineering, electrode structure design, and surface modification. However, the essential electrical mechanisms that significantly influence Zn^{2+} ions migration and deposition behaviors have not been reviewed yet. Herein, in this review, the regulation mechanisms of electrical-related electrostatic repulsive/attractive interactions on Zn^{2+} ions migration, desolvation, and deposition behaviors are systematically discussed. Meanwhile, electric field regulation strategies to promote the Zn^{2+} ions diffusion and uniform Zn deposition are comprehensively reviewed, including enhancing and homogenizing electric field intensity inside the batteries and adding external magnetic/pressure/thermal field to couple with the electric field. Finally, future perspectives on the research directions of the electrical-related strategies for building better Zn metal batteries in practical applications are offered.



<https://onlinelibrary.wiley.com/doi/10.1002/adma.202309726>

7. Javad Safaei, Yifu Gao, Mostafa Hosseinpour, Xiuyun Zhang, Yi Sun, Xiao Tang, Zhijia Zhang, Shijian Wang, Xin Guo, Yao Wang, Zhen Chen, Dong Zhou, Feiyu Kang, Lei Jiang, **Guoxiu Wang***, “Vacancy Engineering for High-Efficiency Nanofluidic Osmotic Energy Generation”, **Journal of the American Chemical Society**, 145, 2669, 2023. IF=16.383. DOI: 10.1021/jacs.2c12936.

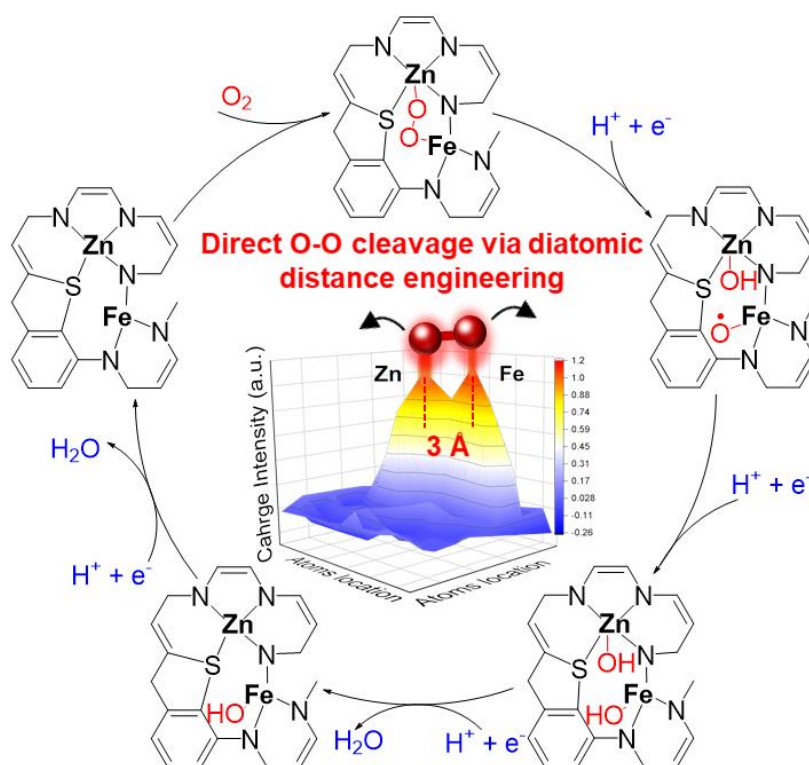
ABSTRACT: Two-dimensional (2D) nanofluidic membranes have shown great promise in harvesting osmotic energy from the salinity difference between seawater and fresh water. However, the output power densities are strongly hampered by insufficient membrane permselectivity. Herein, we demonstrate that vacancy engineering is an effective strategy to enhance the permselectivity of 2D nanofluidic membranes to achieve high-efficiency osmotic energy generation. Phosphorus vacancies were facilely created on NbOPO₄ (NbP) nanosheets, which remarkably enlarged their negative surface charge. As verified by both experimental and theoretical investigations, the vacancy-introduced NbP (V-NbP) exhibited fast transmembrane ion migration and high ionic selectivity originating from the improved electrostatic affinity of cations. When applied in a natural river water|seawater osmotic power generator, the macroscopic-scale V-NbP membrane delivered a record-high power density of 10.7 W m⁻², far exceeding the commercial benchmark of 5.0 W m⁻². This work endows the remarkable potential of vacancy engineering for 2D materials in nanofluidic energy devices.



<https://pubs.acs.org/doi/abs/10.1021/jacs.2c12936>

8. Yuhan Xie, Xin Chen, Kaian Sun, Jinqiang Zhang, Wei-Hong Lai, Hao Liu, **Guoxiu Wang***. “Direct Oxygen-Oxygen Cleavage through Optimizing Interatomic Distances in Dual Single-atom Electrocatalysts for Efficient Oxygen Reduction Reaction”, **Angewandte Chemie International Edition**, 62, e202301833, 2023. IF = 16.823. DOI: 10.1002/anie.202301833

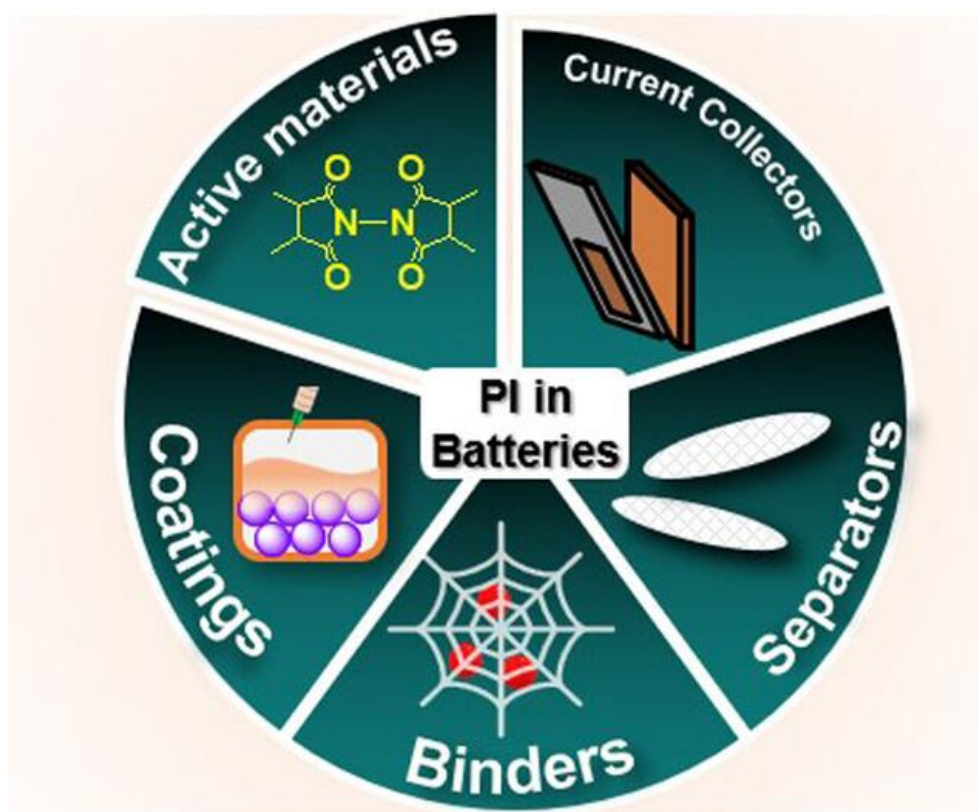
ABSTRACT: The oxygen reduction reaction (ORR) on transition single-atom catalysts (SACs) is sustainable in energy-conversion devices. However, the atomically controllable fabrication of single-atom sites and the sluggish kinetics of ORR have remained challenging. Here, we accelerate the kinetics of acid ORR through a direct O–O cleavage pathway through using a bi-functional ligand-assisted strategy to pre-control the distance of hetero-metal atoms. Concretely, the as-synthesized Fe–Zn diatomic pairs on carbon substrates exhibited an outstanding ORR performance with the ultrahigh half-wave potential of 0.86 V vs. RHE in acid electrolyte. Experimental evidence and density functional theory calculations confirmed that the Fe–Zn diatomic pairs with a specific distance range of around 3 Å, which is the key to their ultrahigh activity, average the interaction between hetero-diatomic active sites and oxygen molecules. This work offers new insight into atomically controllable SACs synthesis and addresses the limitations of the ORR dissociative mechanism.



<https://onlinelibrary.wiley.com/doi/full/10.1002/anie.202301833>

9. Amey Nimkar, Gil Bergman, Elad Ballas, Nophar Tubul, Noam Levi, Fyodor Malchik, Idan Kukurayeva, Munseok S Chae, Daniel Sharon, Mikhael Levi, Netanel Shpigel, **Guoxiu Wang***, Doron Aurbach, “Polyimide Compounds for Post-Lithium Energy Storage Applications”, **Angewandte Chemie International Edition**, 62, e202306904, 2023. IF = 16.823. DOI: 10.1002/anie. 202306904

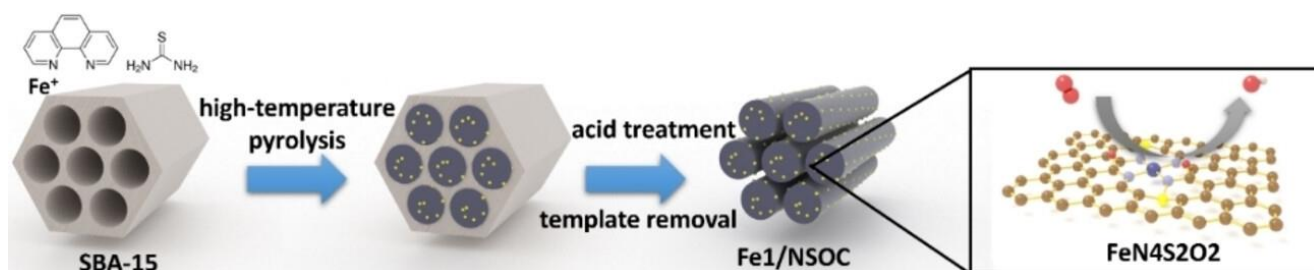
ABSTRACT: The exploration of cathode and anode materials that enable reversible storage of mono and multivalent cations has driven extensive research on organic compounds. In this regard, polyimide (PI)-based electrodes have emerged as a promising avenue for the development of post-lithium energy storage systems. This review article provides a comprehensive summary of the syntheses, characterizations, and applications of PI compounds as electrode materials capable of hosting a wide range of cations. Furthermore, the review also delves into the advancements in PI based solid state batteries, PI-based separators, current collectors, and their effectiveness as polymeric binders. By highlighting the key findings in these areas, this review aims at contributing to the understanding and advancement of PI-based structures paving the way for the next generation of energy storage systems.



<https://onlinelibrary.wiley.com/doi/10.1002/anie.202306904>

10. Yufei Zhao, Ziyang Shen, Juanjuan Huo, Xianjun Cao, Pengfei Ou, Junpeng Qu, Xinming Nie, Jinqiang Zhang, Minghong Wu, **Guoxiu Wang***, Hao Liu, “Epoxy-rich Fe Single Atom Sites Boost Oxygen Reduction Electrocatalysis”, **Angewandte Chemie International Edition**, 62, e202308349, 2023. IF = 16.823. DOI: 10.1002/anie.202308349

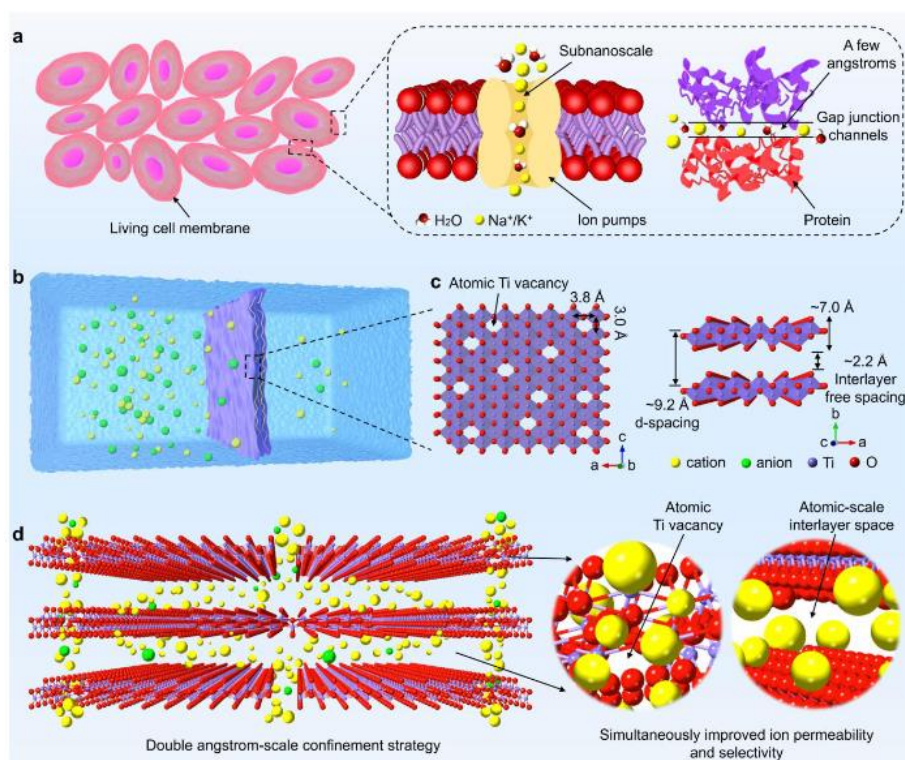
ABSTRACT: Electrocatalysts for highly efficient oxygen reduction reaction (ORR) are crucial for energy conversion and storage devices. Single-atom catalysts with maximized metal utilization and altered electronic structure are the most promising alternatives to replace current benchmark precious metals. However, the atomic level understanding of the functional role for each species at the anchoring sites is still unclear and poorly elucidated. Herein, we report Fe single atom catalysts with the sulfur and oxygen functional groups near the atomically dispersed metal centers (Fe1/NSOC) for highly efficient ORR. The Fe1/NSOC delivers a half-wave potential of 0.92 V vs. RHE, which is much better than those of commercial Pt/C (0.88 V), Fe single atoms on N-doped carbon (Fe1/NC, 0.89 V) and most reported nonprecious metal catalysts. The spectroscopic measurements reveal that the presence of sulfur group induces the formation of epoxy groups near the FeN₄S₂ centers, which not only modulate the electronic structure of Fe single atoms but also participate the catalytic process to improve the kinetics. The density functional theory calculations demonstrate the existence of sulfur and epoxy group engineer the charges of Fe reactive center and facilitate the reductive release of OH* (rate-limiting step), thus boosting the overall oxygen reduction efficiency.



<https://onlinelibrary.wiley.com/doi/full/10.1002/anie.202308349>

11. Chao Liu, Caichao Ye, Tianning Zhang, Jiheng Tang, Kunpeng Mao, Long Chen, Liang Xue, Jingweng Sun, Wenqing Zhang, Xin Wang, Pan Xiong, **Guoxiu Wang***, Junwu Zhu, “Bio-inspired Double Angstrom-Scale Confinement in Ti-deficient Ti_{0.87}O₂ Nanosheet Membranes for Ultrahigh-performance Osmotic Power Generation”, **Angewandte Chemie International Edition**, 62, e202315947, 2023. IF = 16.823. DOI: 10.1002/anie.202315947

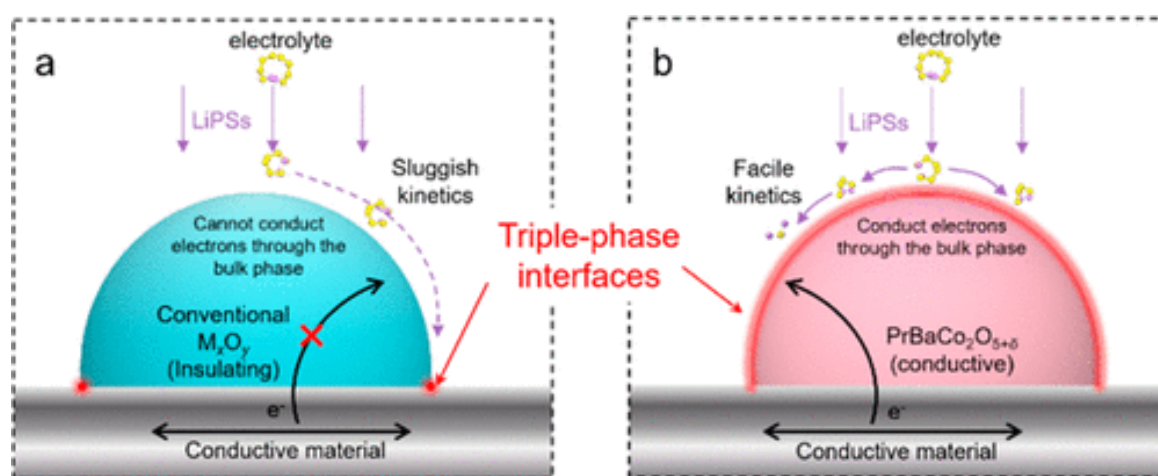
ABSTRACT: Osmotic power, a clean energy source, can be harvested from the salinity difference between seawater and river water. However, the output power densities are hampered by the tradeoff between ion selectivity and ion permeability. Here we propose an effective strategy of double angstrom-scale confinement (DAC) to design ion-permeable channels with enhanced ion selectivity and permeability simultaneously. The fabricated DAC-Ti_{0.87}O₂ membranes possess both Ti atomic vacancies and an interlayer free spacing of ~2.2 Å, which not only generates a profitable confinement effect for Na⁺ ions to enable high ion selectivity but also induces a strong interaction with Na⁺ ions to benefit high ion permeability. Consequently, when applied to osmotic power generation, the DAC-Ti_{0.87}O₂ membranes achieved an ultrahigh power density of 17.8 W m⁻² by mixing 0.5/0.01M NaCl solution and up to 114.2 W m⁻² with a 500-fold salinity gradient, far exceeding all the reported macroscopic-scale membranes. This work highlights the potential of the construction of DAC ion-permeable channels for two-dimensional materials in high-performance nanofluidic energy systems.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/anie.202315947>

12.Zhe Bai, Zhenhua Wang*, Ruilong Li, Zeyu Wu, Pingli Feng, Lina Zhao, Tan Wang, Wenshuo Hou, Yu Bai, **Guoxiu Wang***, and Kening Sun, “Engineering Triple-Phase Interfaces Enabled by Layered Double Perovskite Oxide for Boosting Polysulfide Redox Conversion”, **Nano Letters**, 23, 4908–4915, 2023. IF= 10.8. DOI: 10.1021/acs.nanolett.3c00566

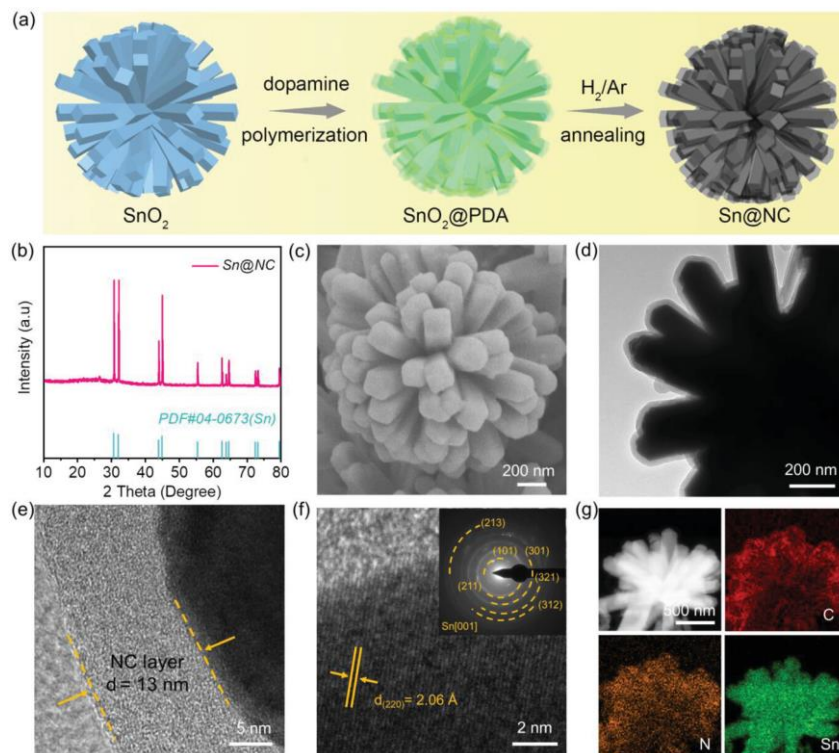
ABSTRACT: The electrocatalytic conversion of polysulfides is crucial to lithium–sulfur batteries and mainly occurs at triple-phase interfaces (TPIs). However, the poor electrical conductivity of conventional transition metal oxides results in limited TPIs and inferior electrocatalytic performance. Herein, a TPI engineering approach comprising superior electrically conductive layered double perovskite $\text{PrBaCo}_2\text{O}_{5+\delta}$ (PBCO) is proposed as an electrocatalyst to boost the conversion of polysulfides. PBCO has superior electrical conductivity and enriched oxygen vacancies, effectively expanding the TPI to its entire surface. DFT calculation and in situ Raman spectroscopy manifest the electrocatalytic effect of PBCO, proving the critical role of enhanced electrical conductivity of this electrocatalyst. PBCO-based Li–S batteries exhibit an impressive reversible capacity of 612 mAh g^{-1} after 500 cycles at 1.0 C with a capacity fading rate of 0.067% per cycle. This work reveals the mechanism of the enriched TPI approach and provides novel insight into designing new catalysts for high-performance Li–S batteries.



<https://pubs.acs.org/doi/10.1021/acs.nanolett.3c00566>

13. Jian Yang, Xin Guo, Hong Gao, Tianyi Wang, Zhigang Liu, Qing Yang, Hang Yao, Jiabao Li, Chengyin Wang, **Guoxiu Wang***, “A High-Performance Alloy-Based Anode Enabled by Surface and Interface Engineering for Wide-temperature Sodium-Ion Batteries”, **Advanced Energy Materials**, 2300351, 2023. IF= 27.8. DOI: 10.1002/aenm.202300351.

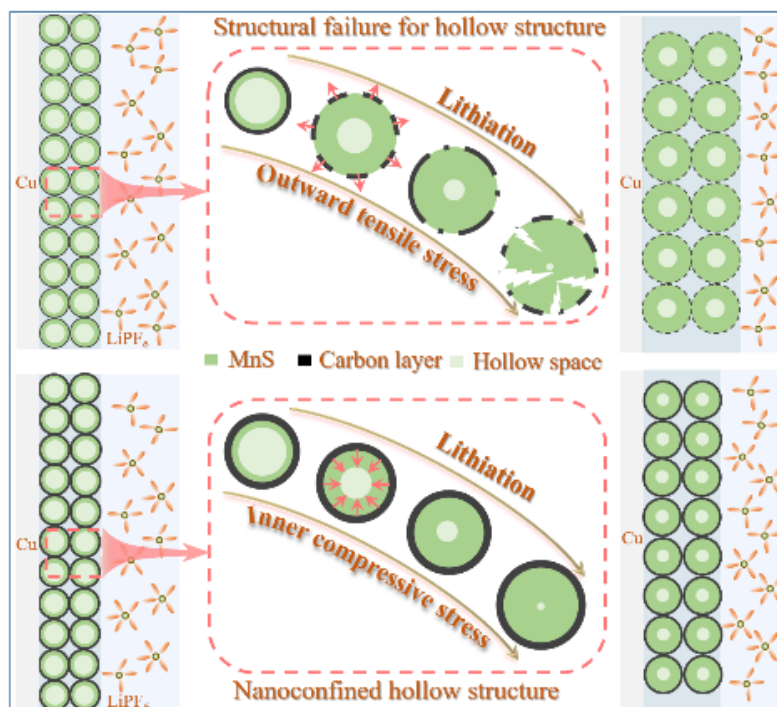
ABSTRACT: Alloy-based anodes have shown great potential to be applied in sodium-ion batteries (SIBs) due to their high theoretical capacities, suitable working potential, and abundant earth reserves. However, their practical applications are severely impeded by large volume expansion, unstable solid-electrolyte interfaces (SEI), and sluggish reaction kinetics during cycling. Herein, a surface engineering of tin nanorods via N-doped carbon layers (Sn@NC) and an interface engineering strategy to improve the electrochemical performance in SIBs are reported. In particular, the authors demonstrate that uniform surface modification can effectively facilitate electron and sodium transport kinetics, confine alloy pulverization, and simultaneously synergize interactions with the ether-based electrolyte to form a robust organic-inorganic SEI. Moreover, it is discovered that the diethylene glycol dimethyl ether electrolyte with strong stability and an optimized Na⁺ solvation structure can co-embed the carbon layer to achieve fast reaction kinetics. Consequently, Sn@NC anodes deliver extra-long cycling stability of more than 10 000 cycles. The full cell of Na₃V₂(PO₄)₃ || Sn@NC exhibits high energy density (215 Wh kg⁻¹), excellent high-rate capability (reaches 80% capacity in 2 min), and long cycle life over a wide temperature range of -20 to 50 °C.



<https://onlinelibrary.wiley.com/doi/full/10.1002/aenm.202300351>

14. Zhipeng Ma, Ailing Song, Zhan Liu, Yaqian Guo, Xin Yang, Qing Li, Yuqian Fan, Lei Dai, Hao Tian, Xiujuan Qin, Hao Liu, Guangjie Shao and **Guoxiu Wang***, “Nanoconfined expansion behavior of hollow MnS@Carbon anode with extended lithiation cyclic stability”, **Advanced Functional Materials**, 33, 2301112, 2023. IF= 19.92. DOI: 10.1002/adfm.202301112.

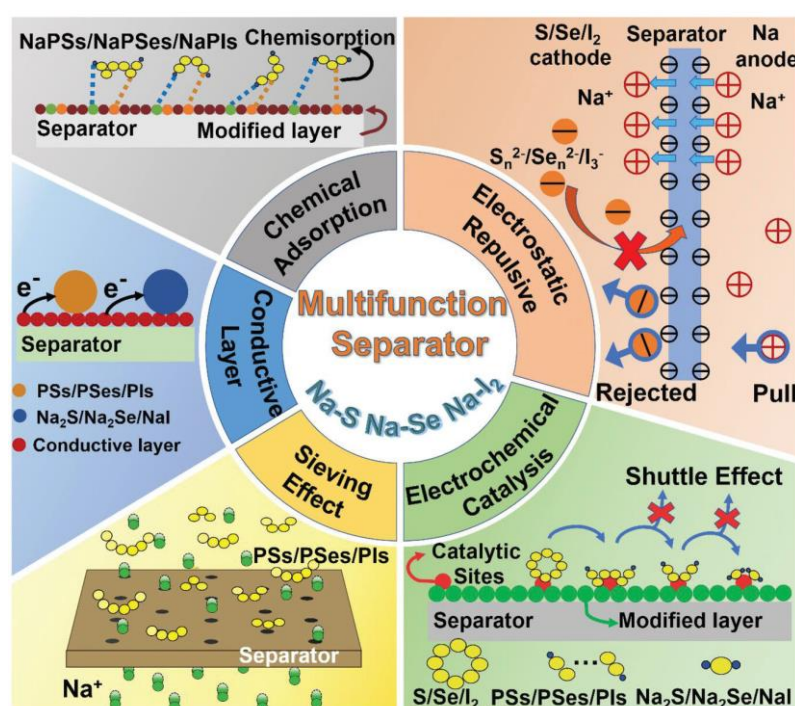
ABSTRACT: The construction of hollow nanostructure by compositing with carbonaceous materials is generally considered an effective strategy to mitigate the drastic volume expansion of transition metal sulfides (TMSs) with high theoretical specific capacity in the process of lithium storage. However, designing well-controlled architectures with extended lithiation cyclic stability, and eased the expansion of the electroactive materials into the reserved hollow spaces still needs to be developed. Here, using MnS as an example, the hollow double-shell carbon-coated TMSs architecture is designed to achieve the controllable operation of shell thickness to regulate interfacial stress. The functional architecture enables the high-capacity MnS to reach reversible capacities and extended lithiation cycling stability at high current densities. In-situ TEM, optical observation characterizations and finite elements are used to analyze the nanoconfined expansion behavior of hollow MnS@C anodes. The as-designed hollow structure with a carbon shell thickness about 12.5 nm can effectively restrict the drastic expansion of MnS nanoshell into inner voids with compressive stress. This work demonstrates a general strategy to design functional carbon coating metal sulfides with tailored interfacial stress.



<https://onlinelibrary.wiley.com/doi/10.1002/adfm.202301112>

15. 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

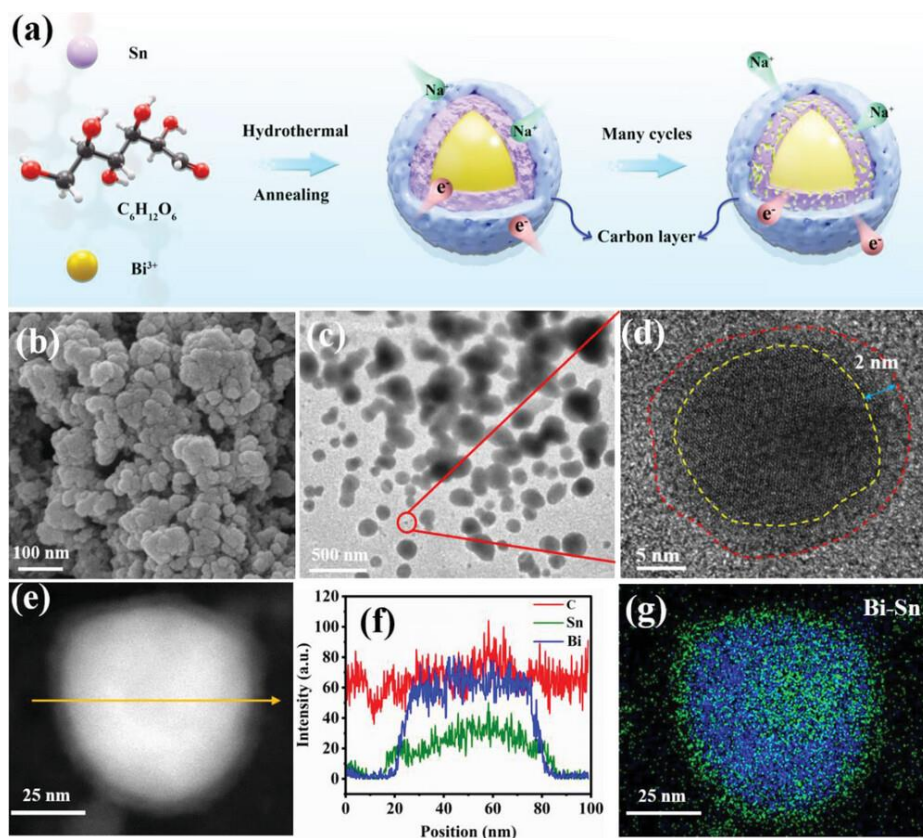
ABSTRACT: Rechargeable sodium–sulfur/selenium/iodine (Na–S/Se/I₂) batteries are regarded as promising candidates for large-scale energy storage systems, with the advantages of high energy density, low cost, and environmental friendliness. However, the electrochemical performances of Na–S/Se/I₂ batteries 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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16. 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

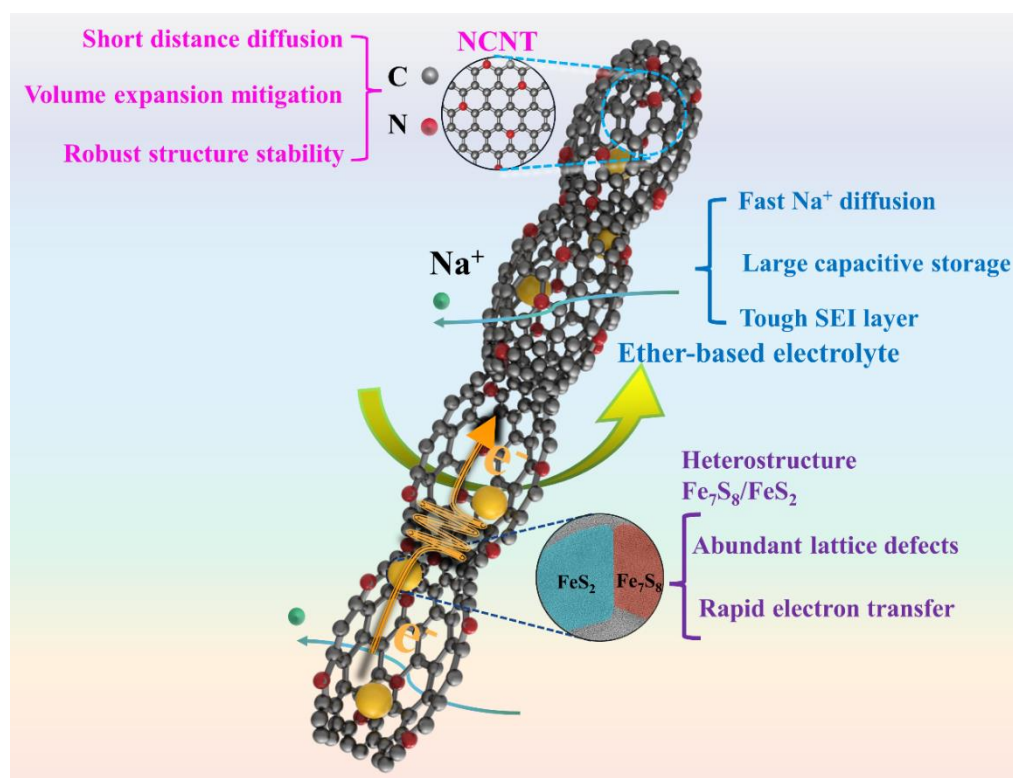
ABSTRACT: Bimetallic composites exhibit great potential as anode materials in advanced energy storage systems owing to their inherent tunability, cost-effectiveness, and simultaneous achievement of high specific capacity and low reaction potential. However, simple biphase mixing often fails to achieve 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>

17. Penghao Song, Jian Yang, Chengyin Wang, Tianyi Wang, Hong Gao, **Guoxiu Wang***, Jiabao Li, “Interface Engineering of Fe₇S₈/FeS₂ Heterostructure *in-situ* Encapsulated into Nitrogen-Doped Carbon Nanotubes for High Power Sodium-Ion Batteries”, **Nano-Micro Letters**, 15, 118, 2023. IF= 26.6. DOI:10.1007/s40820-023-01082-w

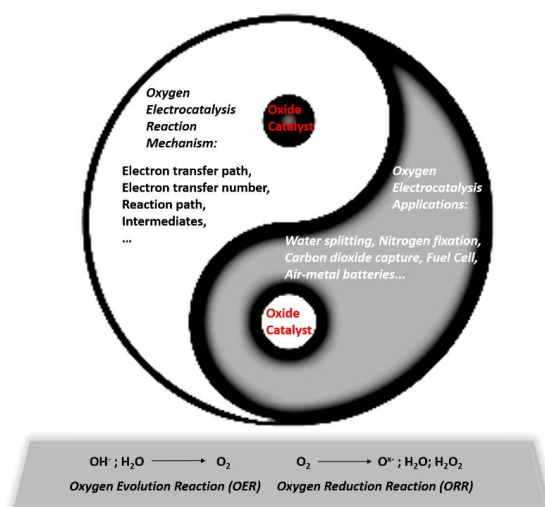
ABSTRACT: Heterostructure engineering combined with carbonaceous materials shows great promise toward promoting sluggish kinetics, improving electronic conductivity, and mitigating the huge expansion of transition metal sulfide electrodes for high-performance sodium storage. Herein, the iron sulfide-based heterostructures *in-situ* hybridized with nitrogen-doped carbon nanotubes (Fe₇S₈/FeS₂/NCNT) have been prepared through a successive pyrolysis and sulfidation approach. The Fe₇S₈/FeS₂/NCNT heterostructure delivered a high reversible capacity of 403.2 mAh g⁻¹ up to 100 cycles at 1.0 A g⁻¹ and superior rate capability (273.4 mAh g⁻¹ at 20.0 A g⁻¹) in ester-based electrolyte. Meanwhile, the electrodes also demonstrated long-term cycling stability (466.7 mAh g⁻¹ after 1000 cycles at 5.0 A g⁻¹) and outstanding rate capability (536.5 mAh g⁻¹ at 20.0 A g⁻¹) in ether-based electrolyte. This outstanding performance could be mainly attributed to the fast sodium-ion diffusion kinetics, high capacitive contribution, and convenient interfacial dynamics in ether-based electrolyte.



<https://link.springer.com/article/10.1007/s40820-023-01082-w>

18. Ning Han, Wei Zhang, Wei Guo, Xuan Zhang, Hui Pan, Lingbao Xing, Hao Tian, **Guoxiu Wang***, Jan Fransaer, **2023**, “Designing oxide catalysts for oxygen electrocatalysis: Insights from mechanism to application”, **Nano-Micro Letter**, 15, 185, 2023. IF=26.6. DOI: 10.1007/s40820-023-01152-z.

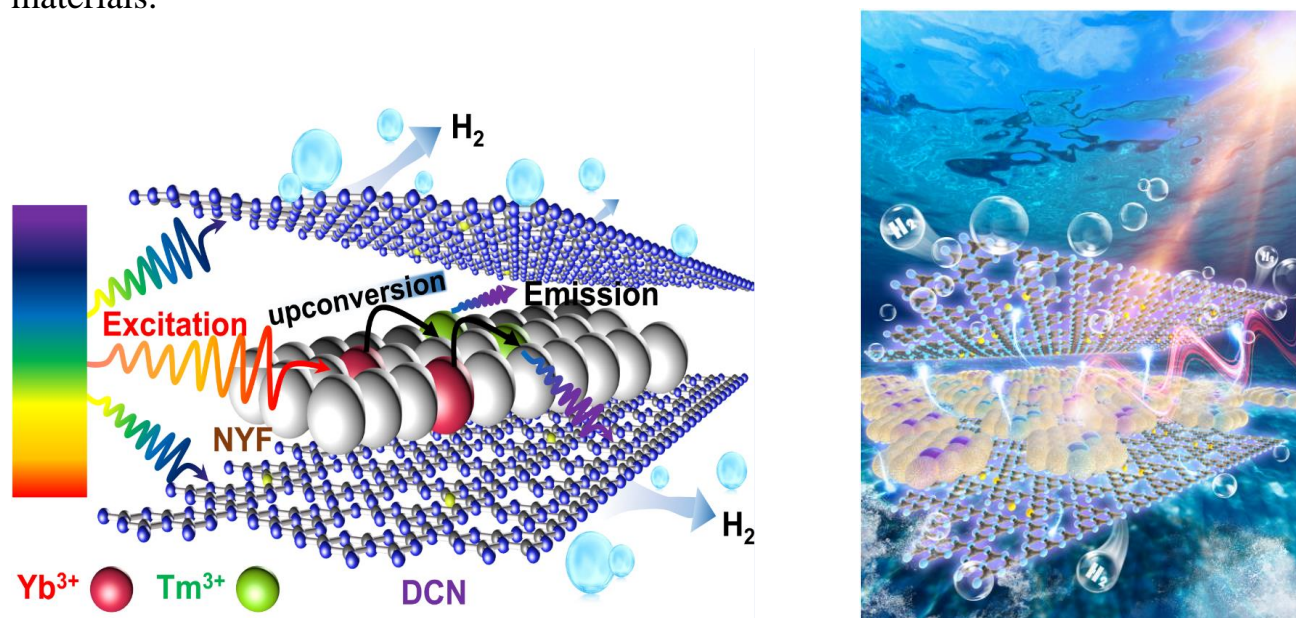
ABSTRACT: The electrochemical oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) are fundamental processes in a range of energy conversion devices such as fuel cells and metal-air batteries. ORR and OER both have significant activation barriers, which severely limit the overall performance of energy conversion devices that utilize ORR/OER. Meanwhile, ORR is another very important electrochemical reaction involving oxygen that has been widely investigated. ORR occurs in aqueous solutions via two pathways: the direct 4-electron reduction or 2-electron reduction pathways from O_2 to water (H_2O) or from O_2 to hydrogen peroxide (H_2O_2). Noble metal electrocatalysts are often used to catalyze OER and ORR, despite the fact that noble metal electrocatalysts have certain intrinsic limitations, such as low storage. Thus, it is urgent to develop more active and stable low-cost electrocatalysts, especially for severe environments (e.g. acidic media). Theoretically, an ideal oxygen electrocatalyst should provide adequate binding to oxygen species. Transition metals not belonging to the Platinum group metals based oxides are a low-cost substance that could give a d orbital for oxygen species binding. As a result, transition metal oxides are regarded as a substitute for typical precious metal oxygen electrocatalysts. However, the development of oxide catalysts for oxygen reduction and oxygen evolution reactions still faces significant challenges, e.g. catalytic activity, stability, cost, and reaction mechanism. We discuss the fundamental principles underlying the design of oxide catalysts, including the influence of crystal structure, and electronic structure on their performance. We also discuss the challenges associated with developing oxide catalysts and the potential strategies to overcome these challenges.



<https://link.springer.com/article/10.1007/s40820-023-01152-z>

19. Xiaochun Gao, Nailiang Yang, Jin Feng, Jiayan Liao, Shaoqi Hou, Xiaoguang Ma, Dawei Su, Xingxing Yu, Ziqing Yang, Javad Safaei, Dan Wang, **Guoxiu Wang***, “Defect and Interface Control on Graphitic Carbon Nitrides/Upconversion Nanocrystals for Enhanced Solar Hydrogen Production”, **National Science Open**, 2, 20220037, 2023, DOI: 10.1360/nso/20220037.

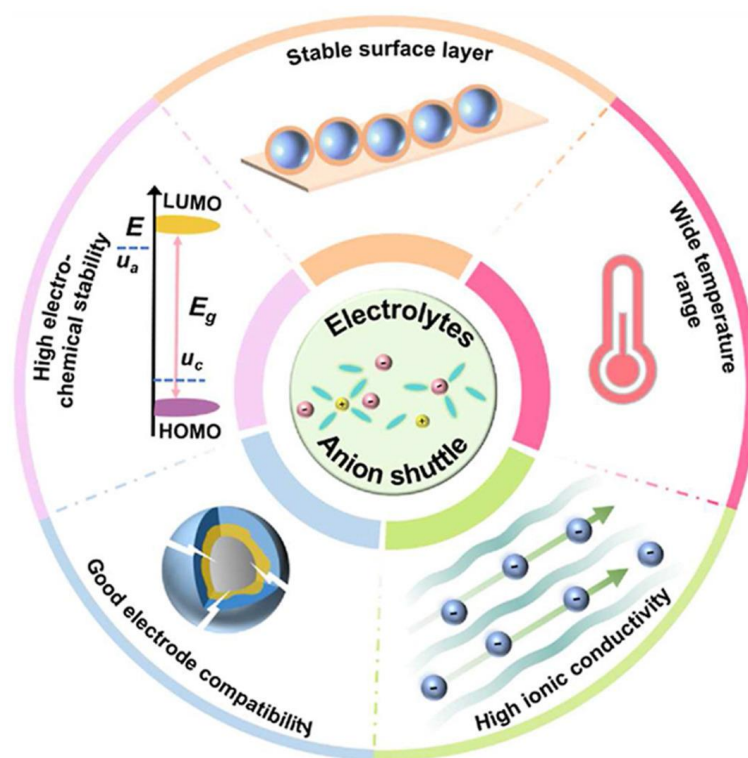
ABSTRACT: The effective utilization of solar energy for hydrogen production requires an abundant supply of thermodynamically active photo-electrons; however, the photocatalysts are generally impeded by insufficient light absorption and fast photocarrier recombination. Here, we report a multiple-regulated strategy to capture photons and boost photocarrier dynamics by developing a broadband photocatalyst composed of defect engineered g-C₃N₄ (DCN) and upconversion NaYF₄:Yb³⁺,Tm³⁺ (NYF) nanocrystals. Through a precise defect engineering, the S dopants and C vacancies jointly render DCN with defect states to effectively extend the visible light absorption to 590 nm and boost photocarrier separation via a moderate electron-trapping ability, thus facilitating the subsequent re-absorption and utilization of upconverted photons/electrons. Importantly, we found a promoted interfacial charge polarization between DCN and NYF has also been achieved mainly due to Y-N interaction, which further favors the upconverted excited energy transfer from NYF onto DCN as verified both theoretically and experimentally. With a 3D architecture, the NYF@DCN catalyst exhibits a superior solar H₂ evolution rate among the reported upconversionbased system, which is 19.3 and 1.5 fold higher than bulk material and DCN, respectively. This work provides an innovative strategy to boost solar utilization by using defect engineering and building up interaction between hetero-materials.



https://www.nso-journal.org/articles/nso/full_html/2023/02/NSO20220037/NSO20220037.html

20. Yao Wang, Xu Yang, Zhijia Zhang, Xia Hu, Yuefeng Meng, Xia Wang, Dong Zhou, Hao Liu, Baohua Li, **Guoxiu Wang***, “Electrolyte design for rechargeable anion shuttle batteries”, *eScience*, 2, 573-590, 2023. DOI: 10.1016/j.esci.2022.10.003.

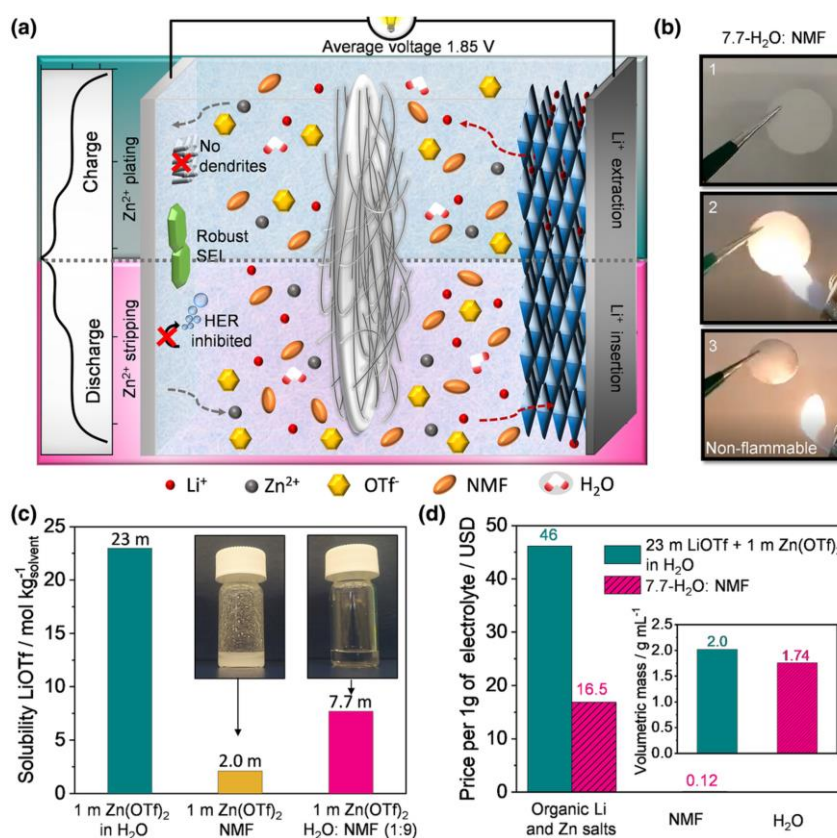
Abstract: As an emerging new type of battery chemistry, the anion shuttle battery (ASB), based on the shuttling and storage of anions, is considered a sustainable alternative to gigawatt-scale energy storage due to the associated resource abundance, low cost, high safety, and high energy density. Although significant progress has been achieved, practical applications of ASBs are still hindered by tough challenges, such as short lifetime, limited reversible capacity, and low Coulombic efficiency. Therefore, it is very necessary to design and explore new electrolyte systems with high electrochemical/chemical stability, sufficient compatibility towards electrodes, and excellent kinetics/reversibility for anion electrochemical reactions. Here, we review the recent achievements and main challenges in developing electrolytes for ASBs, which include solid, non-aqueous, and aqueous electrolytes. We mainly focus on the unique properties and basic principles of designing these electrolytes, and their various performance parameters. Perspectives on design strategies for ASB electrolytes are also presented, which could facilitate the development of advanced ASBs for grid-scale energy storage.



<https://www.sciencedirect.com/science/article/pii/S2667141722001161>

21. Pauline Jaumaux, Shijian Wang, Shuoqing Zhao, Bing Sun, **Guoxiu Wang***, “Electrolyte Solvation Structure Design for High Voltage Zinc-Based Hybrid Batteries”, **Energy & Environmental Materials**, e12578, 2023. **IF=15.0**. DOI:10.1002/eem2.12578.

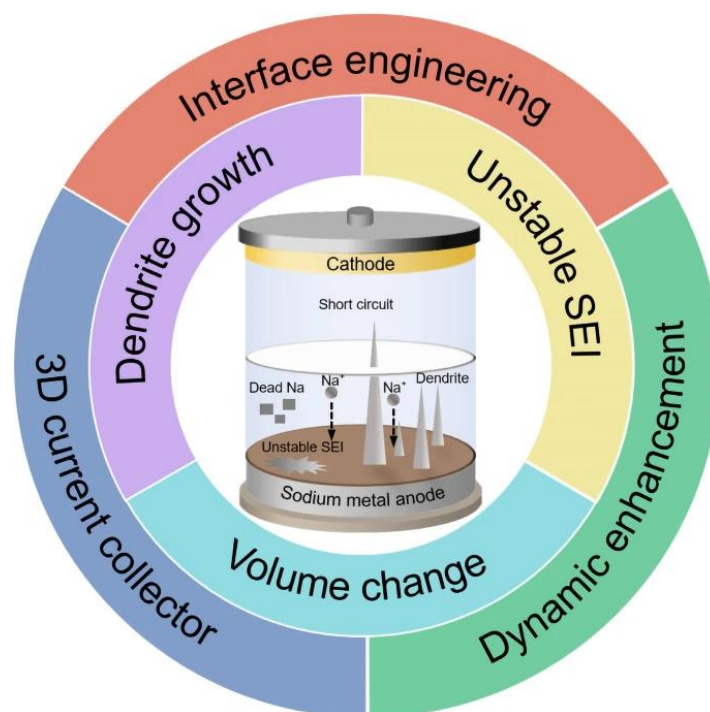
ABSTRACT: Zinc (Zn) metal anodes have enticed substantial curiosity for large-scale energy storage owing to inherent safety, high specific and volumetric energy capacities of Zn metal anodes. However, the aqueous electrolyte traditionally employed in Zn batteries suffers severe decomposition due to the narrow voltage stability window. Herein, we introduce N-methylformamide (NMF) as an organic solvent and modulate the solvation structure to obtain a stable organic/aqueous hybrid electrolyte for high-voltage Zn batteries. NMF is not only extremely stable against Zn metal anodes but also reduces the free water molecule availability by creating numerous hydrogen bonds, thereby accommodating high-voltage Zn || LiMn₂O₄ batteries. The introduction of NMF prevented hydrogen evolution reaction and promoted the creation of an F-rich solid electrolyte interphase, which in turn hampered dendrite growth on Zn anodes. The Zn || LiMn₂O₄ full cells delivered a high average Coulombic efficiency of 99.7% over 400 cycles.



<https://onlinelibrary.wiley.com/doi/full/10.1002/eem2.12578>

22. Jing Xu, Jianhao Yang, Yashuang Qiu, Tianyi Wang, Bing Sun, **Guoxiu Wang***, “Achieving High-performance Sodium Metal Anodes: from Structural Design to Reaction Kinetic Improvement”, **Nano Research**, 2023, accepted. IF= 9.9. DOI: 10.1007/s12274-023-5889-2.

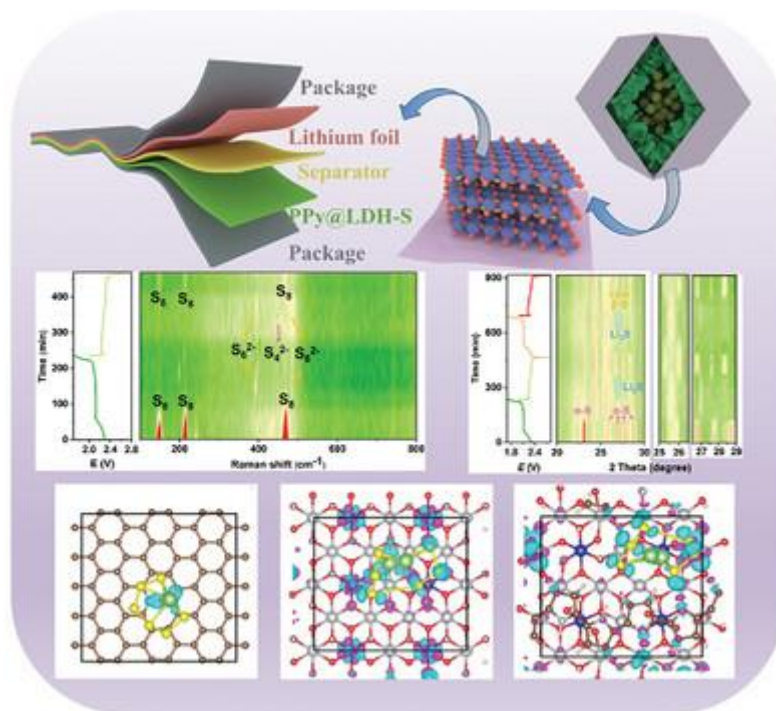
ABSTRACT: Sodium metal is one of the ideal anodes for high-performance rechargeable batteries because of its high specific capacity ($\sim 1166 \text{ mAh}\cdot\text{g}^{-1}$), low reduction potential (-2.71 V compared to standard hydrogen electrodes), and low cost. However, the unstable solid electrolyte interphase, uncontrolled dendrite growth, and inevitable volume expansion hinder the practical application of sodium metal anodes. At present, many strategies have been developed to achieve stable sodium metal anodes. Here, we systematically summarize the latest strategies adopted in interface engineering, current collector design, and the emerging methods to improve the reaction kinetics of sodium deposition processes. First, the strategies of constructing protective layers are reviewed, including inorganic, organic, and mixed protective layers through electrolyte additives or pretreatments. Then, the classification of metal-based, carbon-based, and composite porous frames is discussed, including their function in reducing local deposition current density and the effect of introducing sodiophilic sites. Third, the recent progress of alloys, nanoparticles, and single atoms in improving Na deposition kinetics is systematically reviewed. Finally, the future research direction and the prospect of high-performance sodium metal batteries are proposed.



<https://link.springer.com/article/10.1007/s12274-023-5889-2>

23. Hanghang Dong, Shuo Qi, Lei Wang, Xianfei Chen, Yao Xiao, Yong Wang, Bing Sun*, **Guoxiu Wang**, Shuangqiang Chen, “Conductive Polymer Coated Layered Double Hydroxide as a Novel Sulfur Reservoir for Flexible Lithium-Sulfur Batteries”, **Small**, 2300843, 2023. IF= 15.15. DOI: 10.1002/sml.202300843.

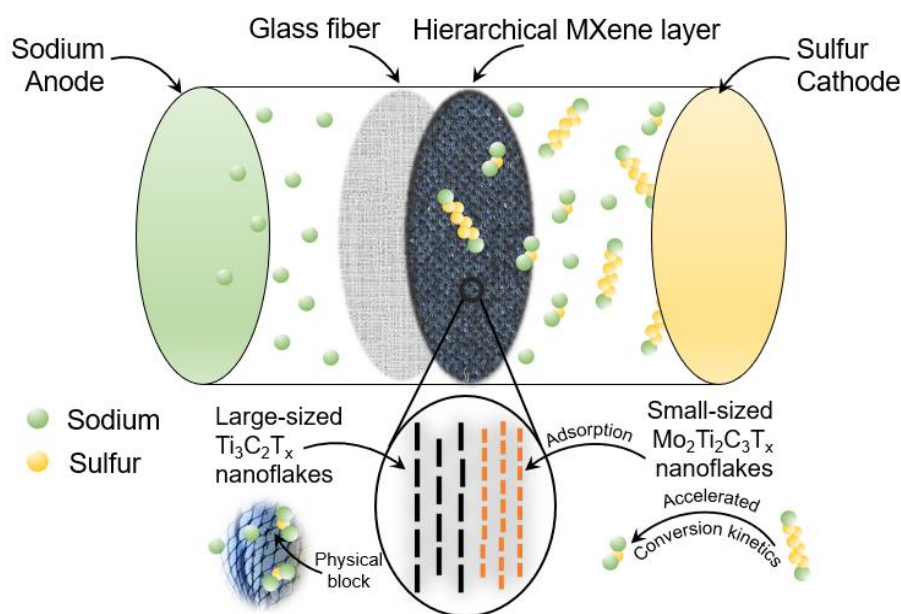
ABSTRACT: Lithium-sulfur battery (LSB) is widely regarded as the most promising next-generation energy storage system owing to its high theoretical capacity and low cost. However, the practical application of LSBs is mainly hampered by the low electronic conductivity of the sulfur cathode and the notorious “shuttle effect”, which lead to high voltage polarization, severe over-charge behavior, and rapid capacity decay. To address these issues, a novel sulfur reservoir is synthesized by coating polypyrrole (PPy) thin film on hollow layered double hydroxide (LDH) (PPy@LDH). After compositing with sulfur, such PPy@LDH-S cathode shows a multi-functional effect to reserve lithium polysulfides (LiPSs). In addition, the unique architecture provides sufficient inner space to encapsulate the volume expansion and enhances the reaction kinetics of sulfur-based redox chemistry. Theoretical calculations have illustrated that the PPy@LDH has shown stronger chemical adsorption capability for LiPSs than those of porous carbon and LDH, preventing the shuttling of LiPSs and enhancing the nucleation affinity of liquid-solid conversion. As a result, the PPy@LDH-S electrode delivers a stable cycling performance and a superior rate capability. Flexible battery has demonstrated this PPy@LDH-S electrode can work properly with treatments of bending, folding, and even twisting, paving the way for wearable devices and flexible electronics.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/sml.202300843>

24. Zefu Huang, Shijian Wang, Xin Guo*, Javad Safaei, Yaojie Lei, Wei-Hong Lai, Xiuyun Zhang, Bing Sun*, Devaraj Shanmukaraj, Michel Armand, Teofilo Rojo, **Guoxiu Wang***, “A hierarchical hybrid MXenes interlayer with triple function for room-temperature sodium-sulfur batteries”, **Advanced Materials Technologies**, 8, 2202147, 2023. IF= 8.856. DOI: 10.1002/admt.202202147

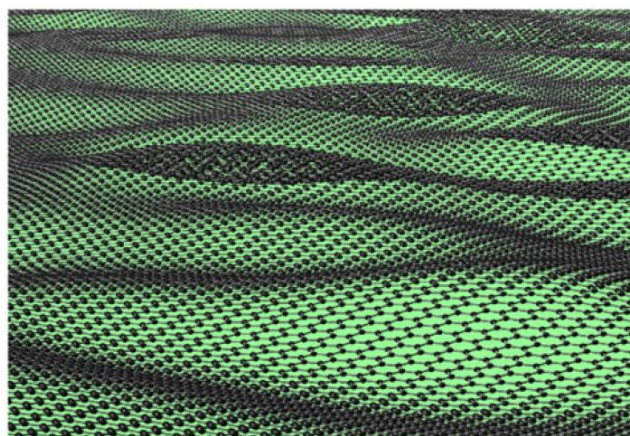
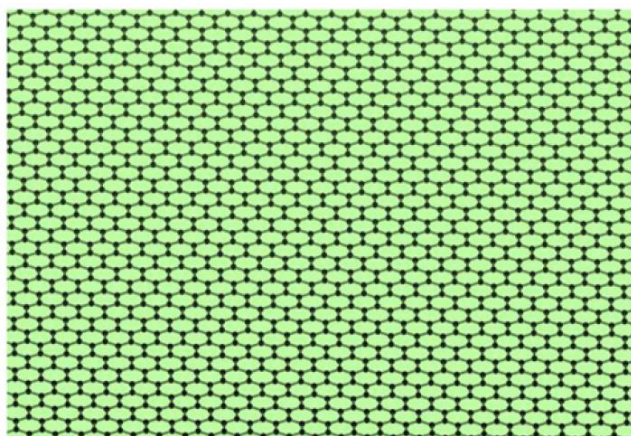
ABSTRACT: Room temperature sodium sulfur (RT Na-S) batteries with high theoretical energy density and low cost have recently gained extensive attention for potential large-scale energy storage applications. However, the shuttle effect of sodium polysulfides is still the main challenge that leads to poor cycling stability, which hinders the practical application of RT Na-S batteries. Herein, a multifunctional hybrid MXene interlayer is designed to stabilize the cycling performance of RT Na-S batteries. The hybrid MXene interlayer comprises a large-sized $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets inner layer followed by a small-sized $\text{Mo}_2\text{Ti}_2\text{C}_3\text{T}_x$ nanoflake outer layer on the surface of the glass fiber (GF) separator. The large-sized $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheet inner layer provides an effective physical block and chemical confinement for the soluble polysulfides. The small-sized $\text{Mo}_2\text{Ti}_2\text{C}_3\text{T}_x$ outer layer offers an excellent polysulfide trapping capability and accelerates the reaction kinetics of polysulfide conversion, due to its superior electronic conductivity, large specific surface area, and Mo-rich catalytic surfaces. As a result, RT Na-S batteries with this hybrid MXene interlayer modified glass fiber separator deliver a stable cycling performance over 200 cycles at 1 C with an enhanced capacity retention of 71%. This unique structure design provides a novel strategy to develop 2D material-based functional interlayer for high-performance metal-sulfur batteries.



<https://onlinelibrary.wiley.com/doi/full/10.1002/admt.202202147>

25. **Guoxiu Wang***, “Graphene nanoripples enable unexpected catalytic reactivity”, **Proceedings of the National Academy of Sciences (PNAS)**, 120, e2303353120, 2023. IF=12.78. DOI: 10.1073/pnas.2303353120

Abstract: Since the discovery of graphene in 2004 (1), graphene and graphene-based materials have been extensively explored for many diverse applications including electronics, optoelectronics, photonics, medicine, energy storage devices, and catalysis. It is well recognized that monolayer graphene is chemically inert and highly stable under ambient conditions. Because of its superior electronically conductive properties and large specific surface area, graphene has been commonly considered for use as a support for catalytically active species, such as nanoparticles, nanoclusters, and single atoms (2). Furthermore, graphene itself can be made catalytically active by creating heteroatom dopants, vacancies, edges, and other atomic-scale defects with the ultimate goal being to break its symmetry and hence, introduce local charge densities (3). In PNAS, Sun et al. (4) report that in addition to the defects described above, the ubiquitous nanoscale ripples in graphene exhibit enhanced catalytic activity, which are able to dissociate molecular hydrogen into protons as strongly as the best-known catalysts.



<https://www.pnas.org/doi/10.1073/pnas.2303353120>

See companion article, “Unexpected catalytic activity of nanorippled graphene,” 10.1073/pnas.2300481120.