

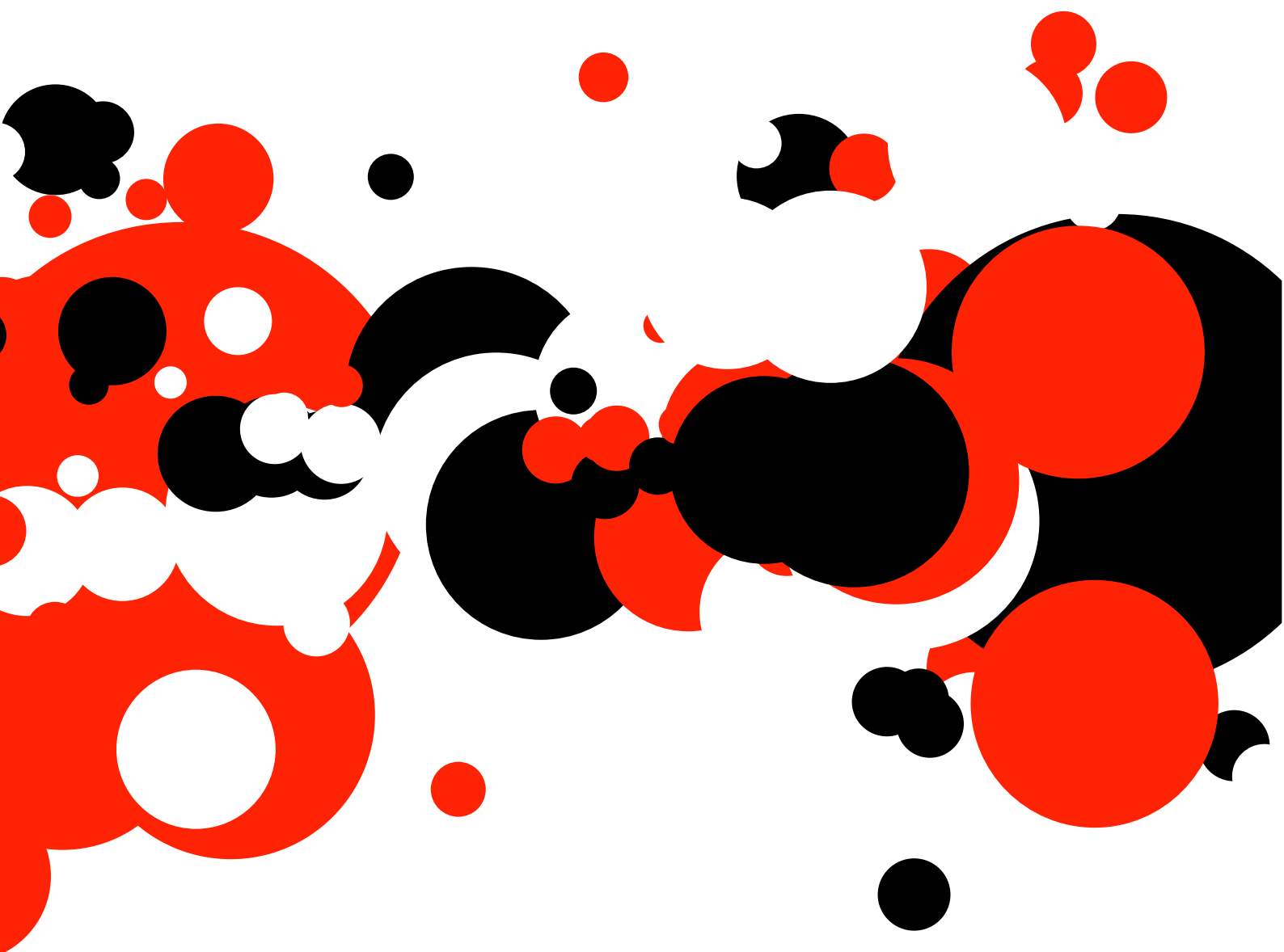


University of Technology Sydney

Centre for Clean Energy Technology

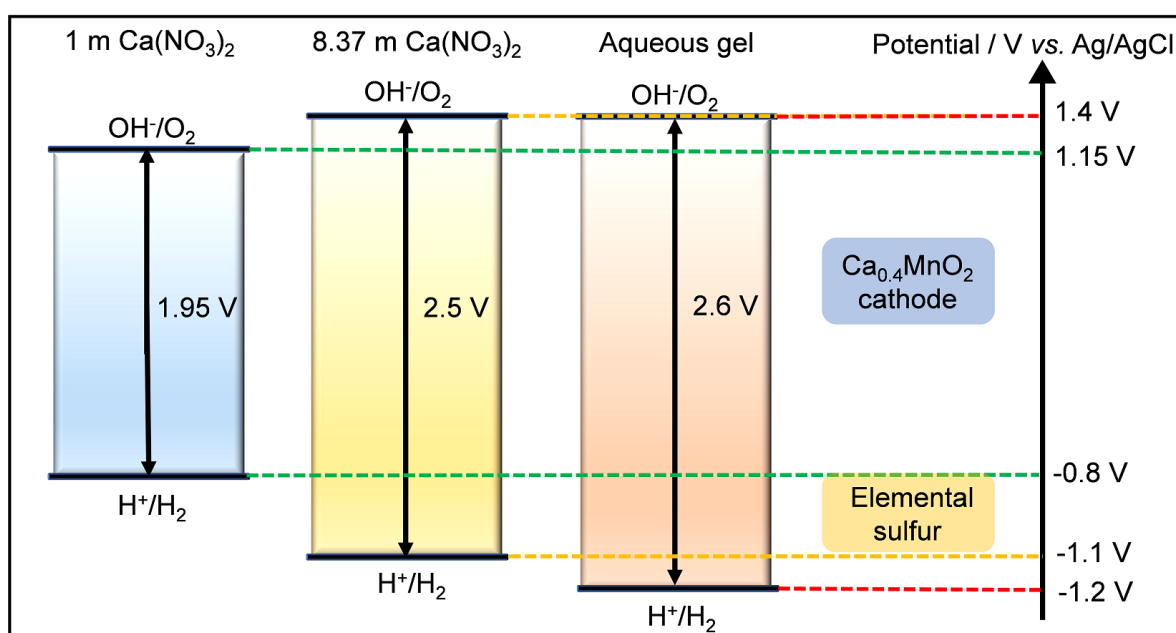
Research Highlights in 2021 (Jan-Dec)

In 2021, over 50 refereed journal papers, including Nature Communications (4), Journal of the American Chemical Society (1), Advanced Materials (1), Angewandte Chemie International Edition (2), Advanced Energy Materials (2), Advanced Functional Materials (3), Chem (1), Nano Letters (3) and Chemical Science (1) have been published in the Centre for Clean Energy Technology.



1. Xiao Tang, Dong Zhou*, Bao Zhang, Shijian Wang, Peng Li, Hao Liu, Xin Guo, Pauline Jaumaux, Xiaochun Gao, Yongzhu Fu, Chengyin Wang, Chunsheng Wang*, **Guoxiu Wang***, “A universal strategy towards high–energy aqueous multivalent–ion batteries”, **Nature Communications**, 2021, 12, 2857. IF=14.919. DOI: 10.1038/s41467-021-23209-6

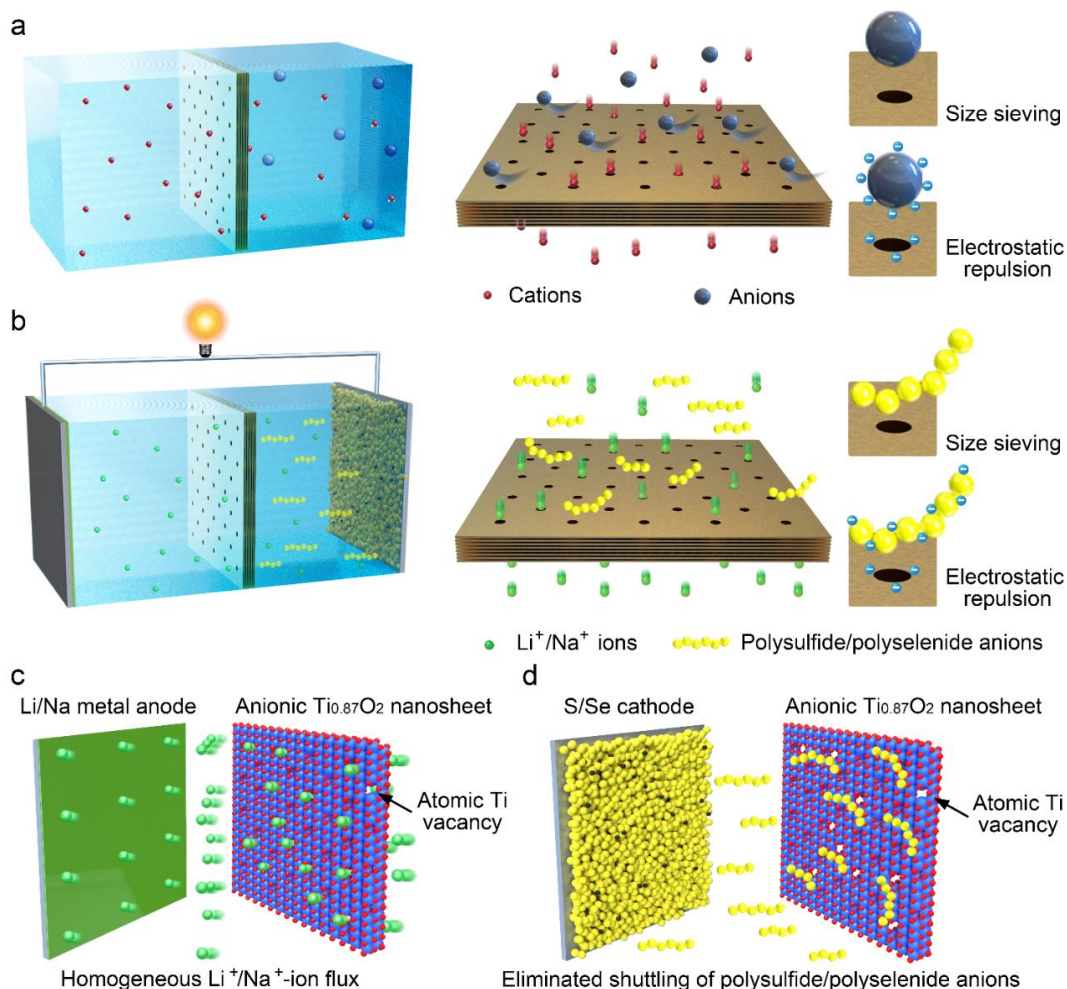
Abstract: Rechargeable multivalent metal (*e.g.*, Ca, Mg or, Al) batteries are ideal candidates for large–scale electrochemical energy storage due to their intrinsic low cost. However, their practical application is hampered by the low electrochemical reversibility, dendrite growth at the metal anodes, sluggish multivalent–ion kinetics in metal oxide cathodes and, poor electrode compatibility with non–aqueous organic–based electrolytes. To circumvent these issues, here we report various aqueous multivalent–ion batteries comprising of concentrated aqueous gel electrolytes, sulfur–containing anodes and, high–voltage metal oxide cathodes as alternative systems to the non–aqueous multivalent metal batteries. This rationally designed aqueous battery chemistry enables satisfactory specific energy, favourable reversibility and improved safety. As a demonstration model, we report a room–temperature calcium–ion/sulfur||metal oxide full cell with a specific energy of 110 Wh kg^{−1} and remarkable cycling stability. Molecular dynamics modelling and experimental investigations reveal that the side reactions could be significantly restrained through the suppressed water activity and formation of a protective inorganic solid electrolyte interphase. The unique redox chemistry of the multivalent–ion system is also demonstrated for aqueous magnesium–ion/sulfur||metal oxide and aluminum–ion/sulfur||metal oxide full cells.



<https://www.nature.com/articles/s41467-021-23209-6>

2. Pan Xiong, Fan Zhang, Xiuyun Zhang, Yifan Liu, Yunyan Wu, Shijian Wang, Javad Safaei, Bing Sun, Renzhi Ma, Zongwen Liu, Yoshio Bando, Takayoshi Sasaki, Xin Wang, Junwu Zhu*, **Guoxiu Wang***, “Atomic-scale regulation of anionic and cationic migration in alkali metal batteries”, **Nature Communications**, 2021, 12, 4184. IF= 14.919. DOI: 10.1038/s41467-021-24399-9

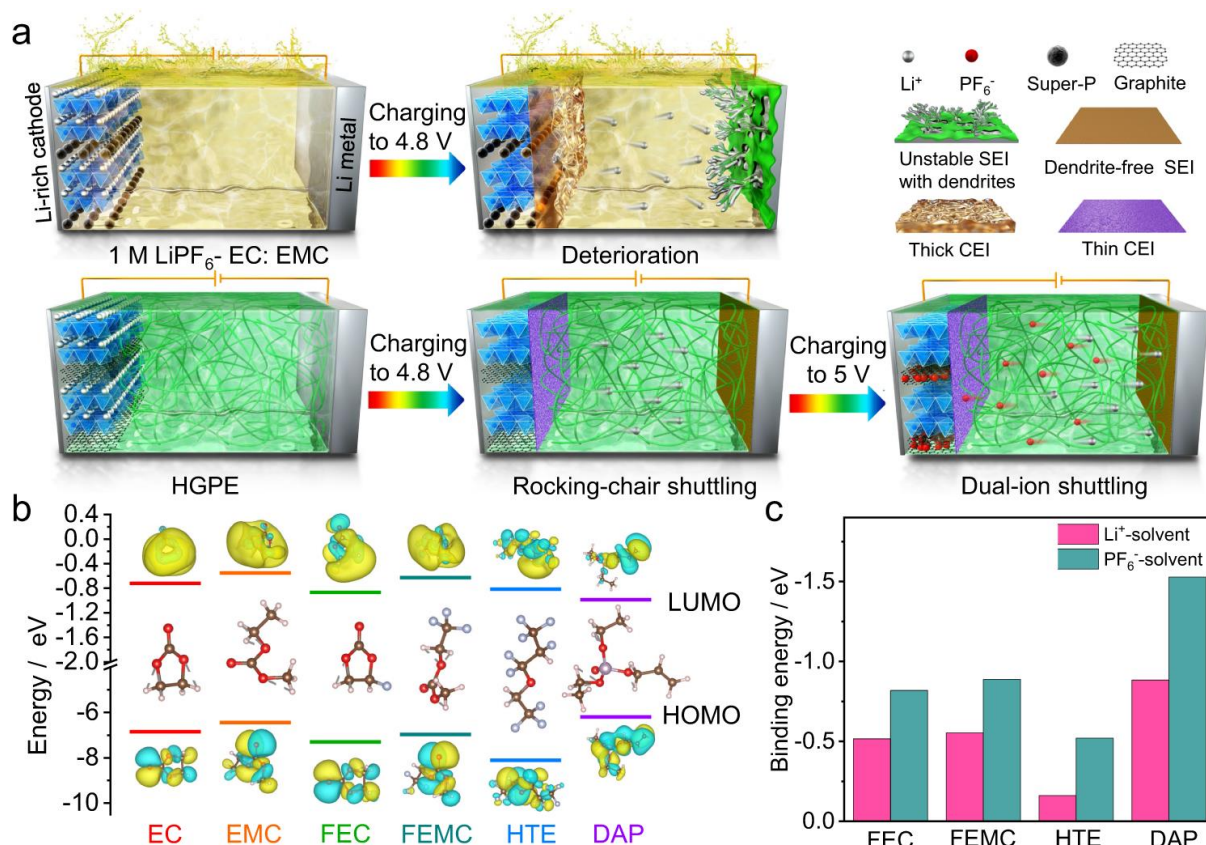
Abstract: The regulation of anions and cations at the atomic scale is of great significance in membrane-based separation technologies. Ionic transport regulation techniques could also play a crucial role in developing high-performance alkali metal batteries such as alkali metal-sulfur and alkali metal-selenium batteries, which suffer from the non-uniform transport of alkali metal ions (e.g., Li^+ or Na^+) and detrimental shuttling effect of polysulfide/polyselenide anions. These drawbacks could cause unfavourable growth of alkali metal depositions at the metal electrode and irreversible consumption of cathode active materials, leading to capacity decay and short cycling life. Herein, we propose the use of a polypropylene separator coated with negatively charged $\text{Ti}_{0.87}\text{O}_2$ nanosheets with Ti atomic vacancies to tackle these issues. In particular, we demonstrate that the electrostatic interactions between the negatively charged $\text{Ti}_{0.87}\text{O}_2$ nanosheets and polysulfide/polyselenide anions reduce the shuttling effect. Moreover, the $\text{Ti}_{0.87}\text{O}_2$ -coated separator regulates the migration of alkali ions ensuring a homogeneous ion flux and the Ti vacancies, acting as sub-nanometric pores, promote fast alkali-ion diffusion.



<https://www.nature.com/articles/s41467-021-24399-9>

3. Junru Wu, Xianshu Wang, Qi Liu, Shuwei Wang, Dong Zhou*, Feiyu Kang, Devaraj Shanmukaraj, Michel Armand, Teofilo Rojo, Baohua Li*, **Guoxiu Wang***, “A synergistic exploitation to produce high-voltage quasi-solid-state lithium metal batteries”, **Nature Communications**, 2021, 12, 5746. IF= 14.919. DOI: 10.1038/s41467-021-26073-6.

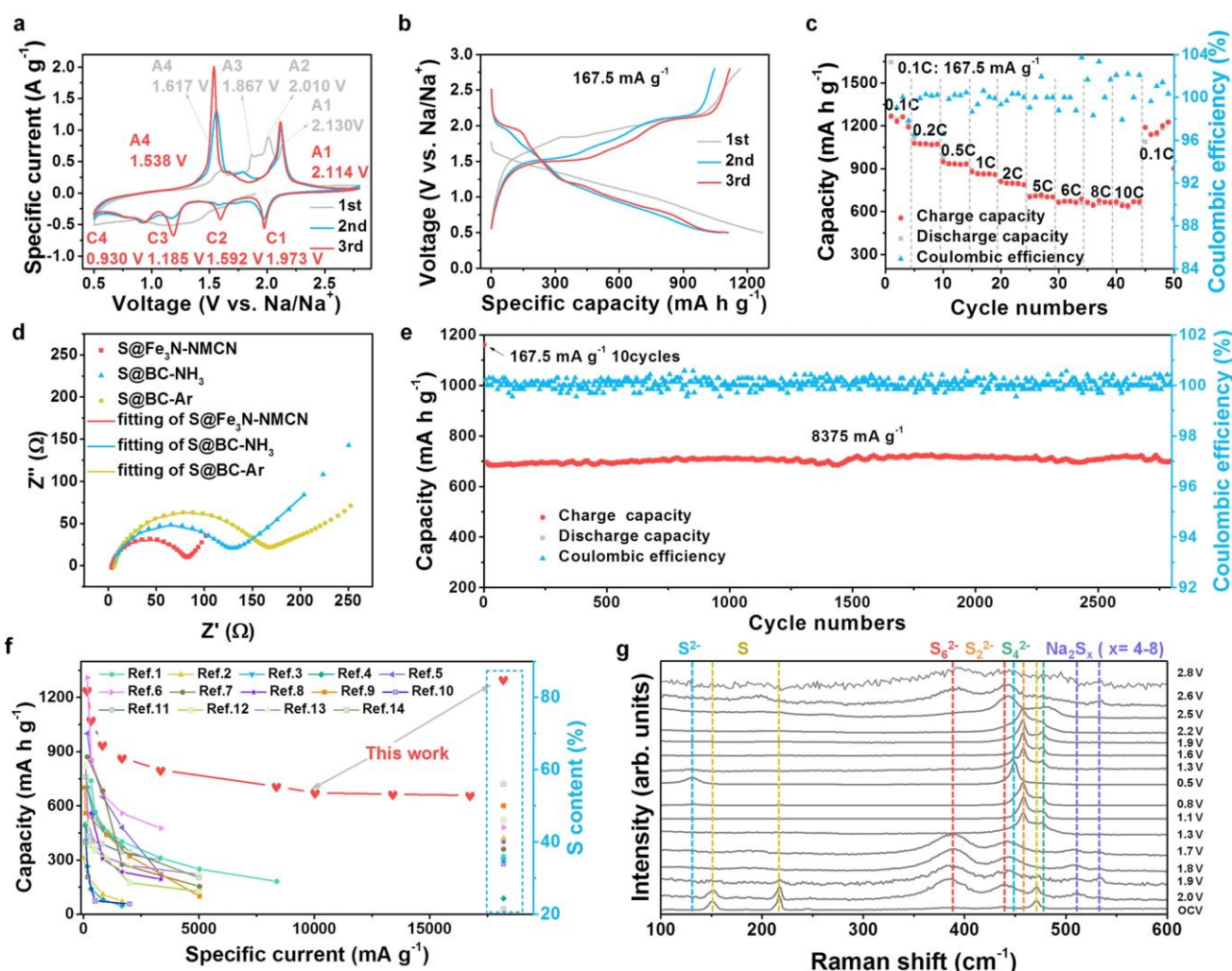
Abstract: The current Li-based battery technology is limited in terms of energy contents. Therefore, several approaches are considered to improve the energy density of these energy storage devices. Here, we report the combination of a heteroatom-based gel polymer electrolyte with a hybrid cathode comprising of a Li-rich oxide active material and graphite conductive agent to produce a high-energy “shuttle-relay” Li metal battery, where additional capacity is generated from the electrolyte’s anion shuttling at high voltages. The gel polymer electrolyte, prepared via in situ polymerization in an all-fluorinated electrolyte, shows adequate ionic conductivity (around 2 mS cm^{-1} at $25 \text{ }^\circ\text{C}$), oxidation stability (up to 5.5 V vs Li/Li^+), compatibility with Li metal and safety aspects (i.e., non-flammability). The polymeric electrolyte allows for a reversible insertion of hexafluorophosphate anions into the conductive graphite (i.e., dual-ion mechanism) after the removal of Li ions from Li-rich oxide (i.e., rocking-chair mechanism).



<https://www.nature.com/articles/s41467-021-26073-6>

4. Yuruo Qi, Qing-Jie Li, Yuanke Wu, Shu-juan Bao, Changming Li, Yuming Chen, **Guoxiu Wang***, Maowen Xu, "A Fe₃N/carbon composite electrocatalyst for effective polysulfides regulation in roomtemperature Na-S batteries", **Nature Communications**, 2021, 12, 6347. IF= 14.919. DOI: 10.1038/s41467-021-26631-y

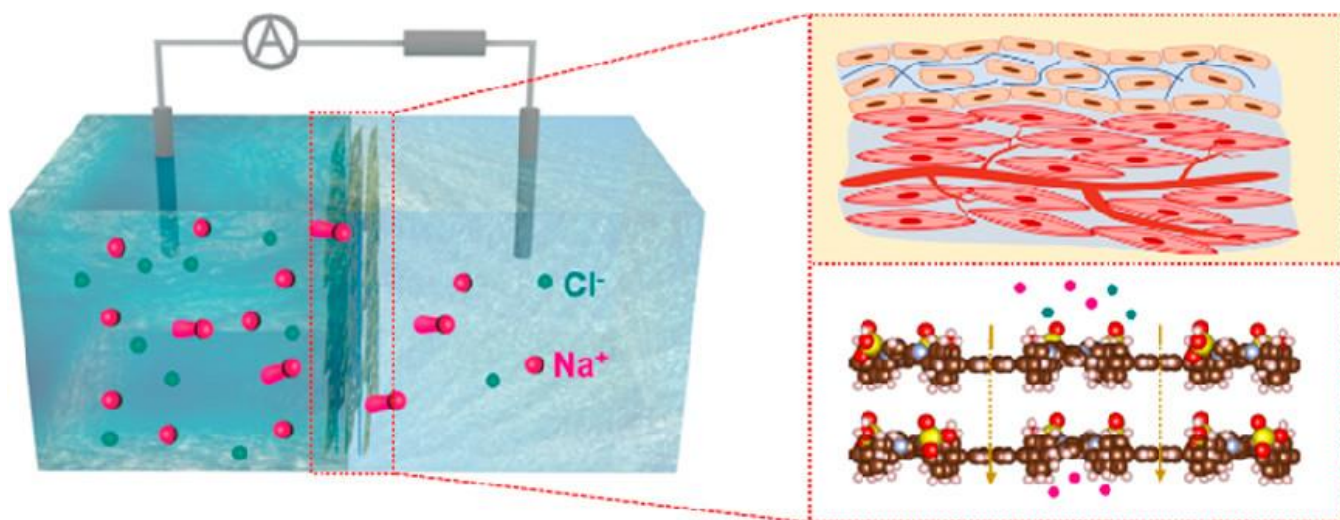
Abstract: The practical application of room-temperature Na-S batteries is hindered by the low sulfur utilization, inadequate rate capability and poor cycling performance. To circumvent these issues, here, we propose an electrocatalyst composite material comprising of N-doped nanocarbon and Fe₃N. The multilayered porous network of the carbon accommodates large amounts of sulfur, decreases the detrimental effect of volume expansion, and stabilizes the electrodes structure during cycling. Experimental and theoretical results testify the Fe₃N affinity to sodium polysulfides via Na-N and Fe-S bonds, leading to strong adsorption and fast dissociation of sodium polysulfides. With a sulfur content of 85 wt.%, the positive electrode tested at room-temperature in non-aqueous Na metal coin cell configuration delivers a reversible capacity of about 1165 mA h g⁻¹ at 167.5 mA g⁻¹, satisfactory rate capability and stable capacity of about 696 mA h g⁻¹ for 2800 cycles at 8375 mA g⁻¹.



<https://www.nature.com/articles/s41467-021-26631-y>

5. Zengming Man, Javad Safaei, Zhen Zhang, Yizhou Wang, Dong Zhou*, Peng Li, Xiaogang Zhang, Lei Jiang, **Guoxiu Wang***, “Serosa-Mimetic Nanoarchitecture Membranes for Highly Efficient Osmotic Energy Generation”, **Journal of the American Chemical Society**, 2021, 143, 16206–16216. IF = 15.419. DOI: 10.1021/jacs.1c07392.

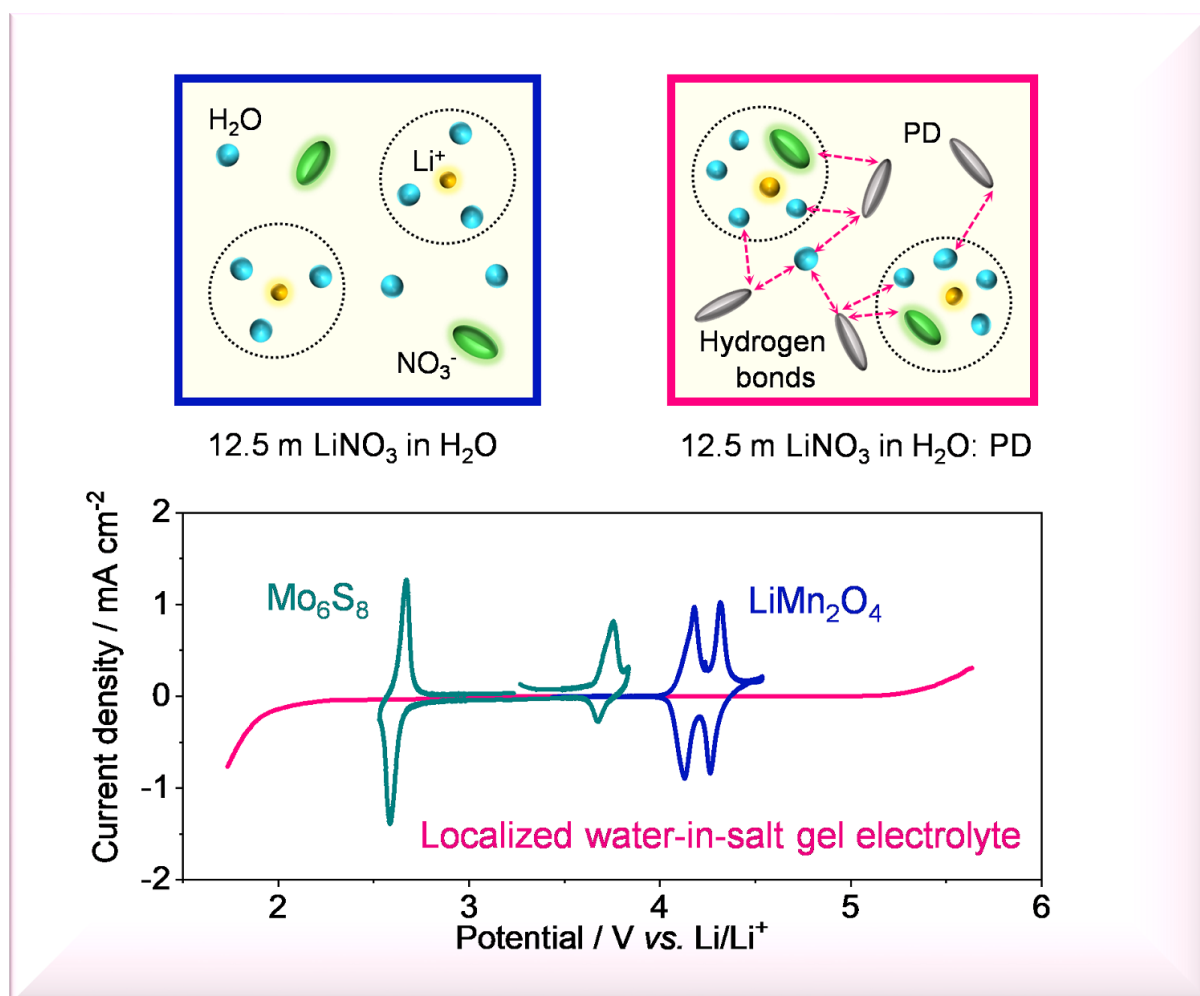
Abstract: Osmotic energy stored between seawater and freshwater is a clean and renewable energy source. However, developing high-efficiency and durable permselective membranes for harvesting osmotic energy remains a longstanding bottleneck. Herein, we report that a nanocomposite membrane with a biological serosa-mimetic structure can achieve high-performance osmotic energy generation through the coupling of two-dimensional (2D) sulfonated covalent organic framework (COF) nanosheets and anion-grafted aramid nanofibers (ANFs). As verified by theoretical calculations and experimental investigations, the 2D COF nanosheets not only provide abundant one-dimensional (1D)/2D nanofluidic channels to synergistically benefit an ultrafast ion migration but also enable high cation permselectivity via the covalently tethered anions. The grafted ANFs increase the mechanical strength of the membrane and further improve the ion diffusion/rectification. When it was applied in an osmotic power generator, the biomimetic membrane delivered a power density of 9.6 W m^{-2} , far surpassing the commercial benchmark of 5.0 W m^{-2} . This work could boost the viability of osmotic energy conversion toward a sustainable future.



<https://pubs.acs.org/doi/abs/10.1021/jacs.1c07392>

6. Pauline Jaumaux, Xu Yang, Bao Zhang, Javad Safaei, Xiao Tang, Dong Zhou*, Chunsheng Wang*, **Guoxiu Wang***. ““Localized Water-In-Salt” Electrolyte for Aqueous Lithium-Ion Batteries”, **Angewandte Chemie International Edition**, 2021, 60, 19965. IF= 15.336. DOI:10.1002/anie.202107389.

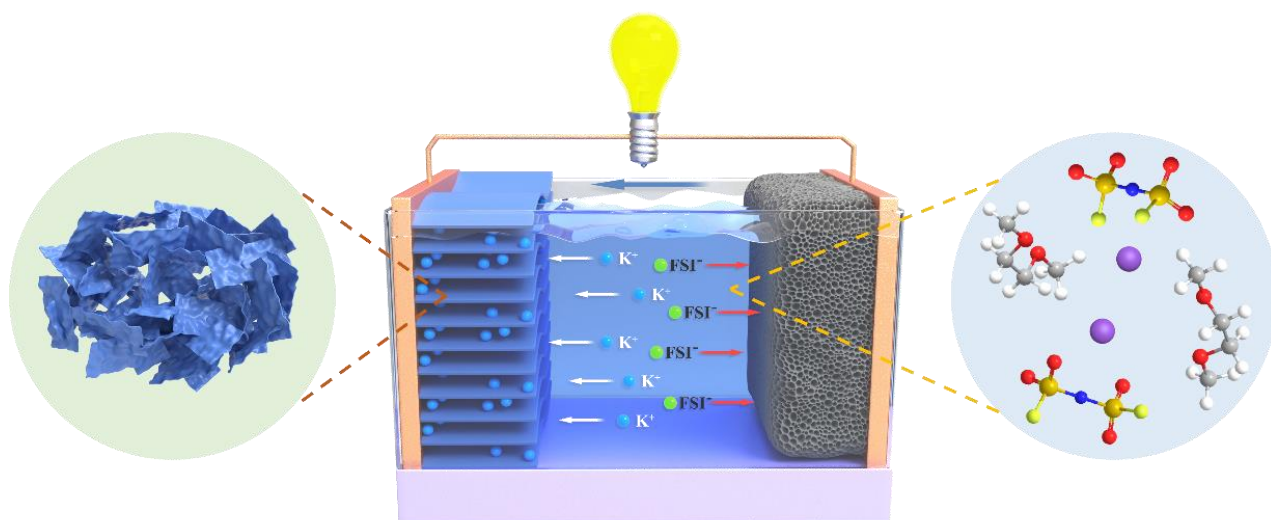
Abstract: “Water-in-salt” (WIS) electrolytes using super-concentrated organic lithium (Li) salts are attracting tremendous interest for high energy aqueous Li-ion batteries, owing to their wide electrochemical stability window that enables the application of high-energy electrode couples. However, the high salt cost, high viscosity, poor wettability and environmental hazards remain a great challenge. Herein, we present a “localized water-in-salt” (LWIS) electrolyte based on low-cost lithium nitrate (LiNO_3) salt and 1,5-pentanediol (PD) as inert diluent. The addition of PD not only maintains the solvation structure of the WIS electrolyte and improves the electrolyte stability via hydrogen-bonding interactions with water and NO_3^- molecules, but also dramatically reduces the total salt concentration. Furthermore, by *in-situ* gelling the LWIS electrolyte with tetraethylene glycol diacrylate (TEGDA) monomer, the electrolyte stability window can be further expanded to 3.0 V. The as-developed Mo_6S_8 |LWIS gel electrolyte| LiMn_2O_4 (LMO) batteries delivered outstanding cycling performance with an average Coulombic efficiency of 98.53 % after 250 cycles at 1 C.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/anie.202107389>

7. Shuoqing Zhao, Zhichao Liu, Guanshun Xie, Xin Guo, Ziqi Guo, Fei Song, Guohao Li, Chi Chen, Xiuqiang Xie*, Nan Zhang, Bing Sun*, Shaojun Guo and **Guoxiu Wang***. “Achieving High-Performance 3D K⁺-Pre-intercalated Ti₃C₂T_x MXene for Potassium-Ion Hybrid Capacitors via Regulating Electrolyte Solvation Structure”, **Angewandte Chemie International Edition**, 2021, 60, 26246–26253. IF= 15.336. DOI: 10.1002/anie.202112090.

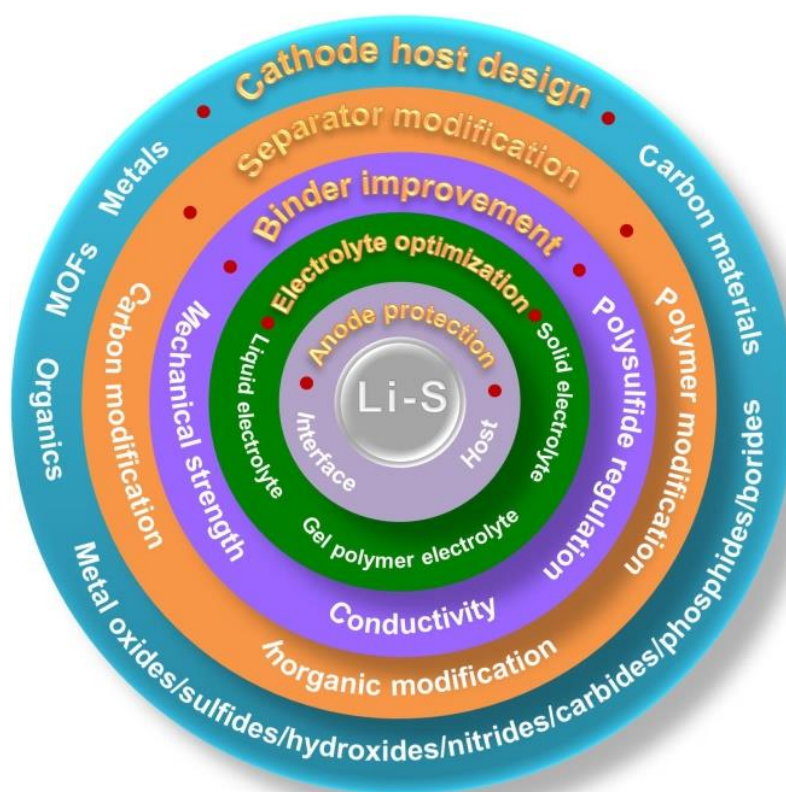
Abstract: The development of high-performance anode materials for potassium-based energy storage devices with long-term cyclability requires combined innovations from rational material design to electrolyte optimization. In this work, three-dimensional K⁺-pre-intercalated Ti₃C₂T_x MXene with enlarged interlayer distance has been constructed for efficient electrochemical potassium-ion storage. We found that the optimized solvation structure of the concentrated ether-based electrolyte leads to the formation of a thin and inorganic-rich solid electrolyte interphase (SEI) on the K⁺-pre-intercalated Ti₃C₂T_x electrode, which is beneficial for interfacial stability and reaction kinetics. As a proof of concept, 3D K⁺-Ti₃C₂T_x//activated carbon (AC) potassium-ion hybrid capacitors (PIHCs) have been assembled and exhibited promising electrochemical performances. These results highlight the significant roles of both rational structure design and electrolyte optimization for highly reactive MXene-based anode materials in energy storage devices.



<https://onlinelibrary.wiley.com/doi/10.1002/anie.202112090>

8. Yi Chen, Tianyi Wang, Huajun Tian, Dawei Su*, Qiang Zhang* and **Guoxiu Wang***, “Advances in Lithium-Sulfur Batteries: From Academic Research to Commercial Viability”, **Advanced Materials**, 2021, 2003666. IF=30.849. DOI: 10.1002/adma.202003666.

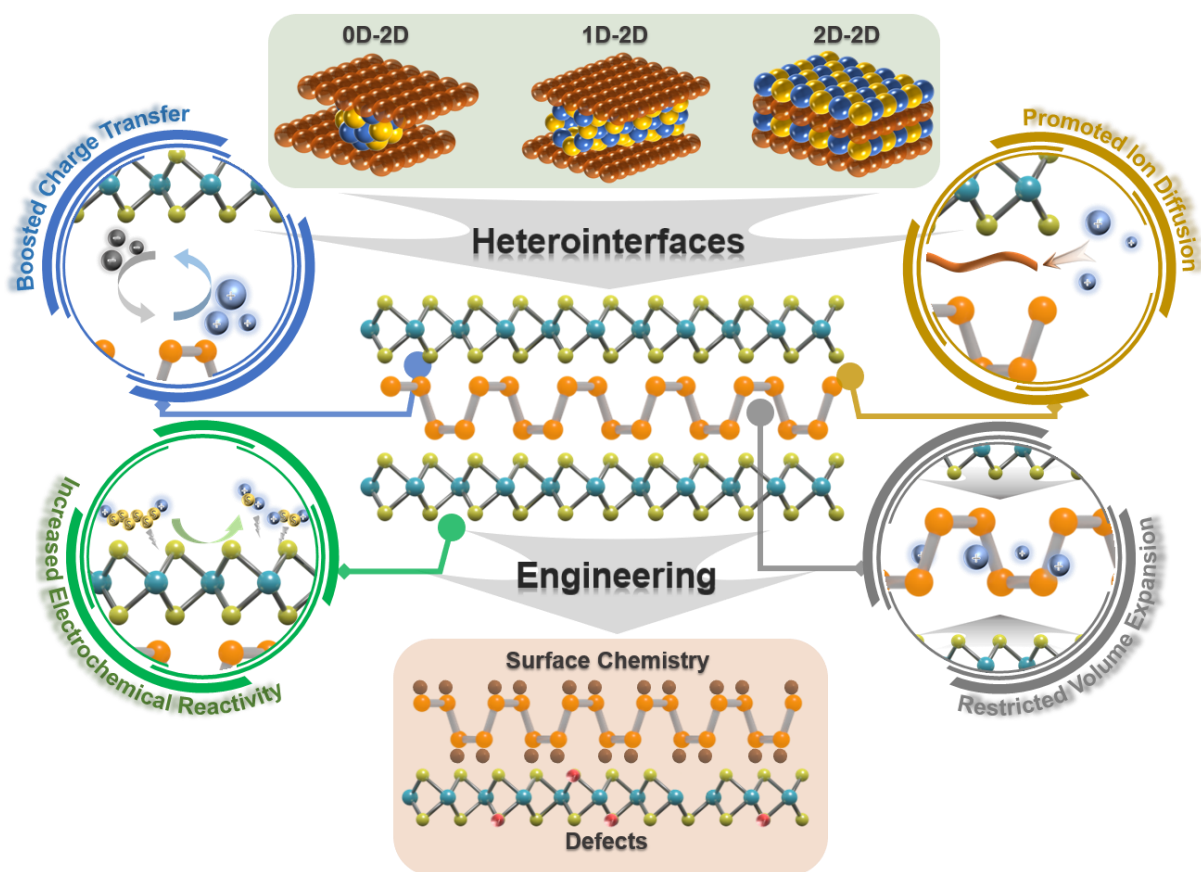
Abstract: Lithium-ion batteries, which revolutionize the portable electronics over the past three decades, have been eventually recognized with the 2019 Nobel Prize in chemistry. As the energy density of current lithium-ion batteries is approaching its limit, developing new battery technology beyond lithium-ion chemistry is significant for next-generation high energy storage. Lithium-sulfur (Li-S) batteries, which rely on the reversible redox reactions between lithium and sulfur, appears to be a promising energy storage system to take over from the conventional lithium-ion batteries for next-generation energy storage owing to their overwhelming energy density compared to the existing lithium-ion batteries today. Over the past 60 years, especially the past decade, significant academic and commercial progress has been made on Li-S batteries. From the concept of sulfur cathode first proposed in the 1960s to the current some commercial Li-S batteries used in unmanned aircrafts, the story of Li-S batteries is full of breakthroughs and back tracing steps. This review will summarize the development and advancement of Li-S batteries in terms of sulfur-based composite cathode design, separator modification, binder improvement, electrolyte optimization and lithium metal protection. It will also offer an outlook on the future directions and prospects for Li-S batteries.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.202003666>

9. Shijian Wang, Sai Zhao, Xin Guo*, **Guoxiu Wang***, “2D Material-Based Heterostructures for Rechargeable Batteries”, **Advanced Energy Materials**, 2021, 2100864. IF= 29.368. DOI:10.1002/aenm.202100864.

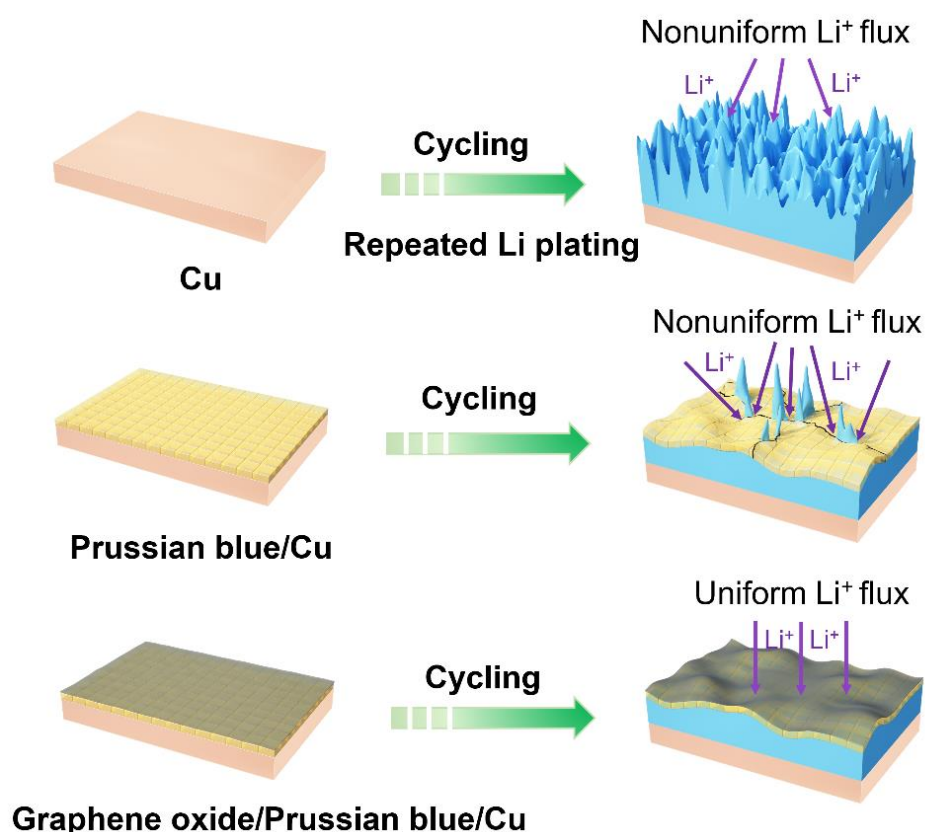
Abstract: 2D materials are regarded as promising electrode materials for rechargeable batteries because of their advantages in providing ample active sites and improving electrochemical reaction kinetics. However, it remains a great challenge for 2D materials to fulfill all requirements for high-performance energy storage devices in terms of electronic conductivity, the number of accessible active sites, structural stability, and mass production capability. Recent advances in constructing 2D material-based heterostructures offer opportunities for utilizing synergistic effects between the individual blocks to achieve optimized properties and enhanced performance. In this perspective, the latest advances of 2D material-based heterostructures are summarized, with particular emphasis on their multifunctional roles in high-performance rechargeable batteries. Synthetic strategies, structural features in mixed dimensionalities, structure engineering strategies, and distinct functionalities of the 2D material-based heterostructures in various electrochemical applications are systematically introduced. Finally, challenges and perspectives are presented to highlight future opportunities for developing 2D material-based heterostructures for practical energy storage.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202100864>

10. Lishuang Fan, Bing Sun*, Kang Yan, Pan Xiong, Xin Guo, Zhikun Guo, Naiqing Zhang*, Yujie Feng, Kening Sun, **Guoxiu Wang***. "A Dual-Protective Artificial Interface for Stable Lithium Metal Anodes", **Advanced Energy Materials**, 2021, accepted. IF=29.368. DOI: 10.1002/aenm.202102242.

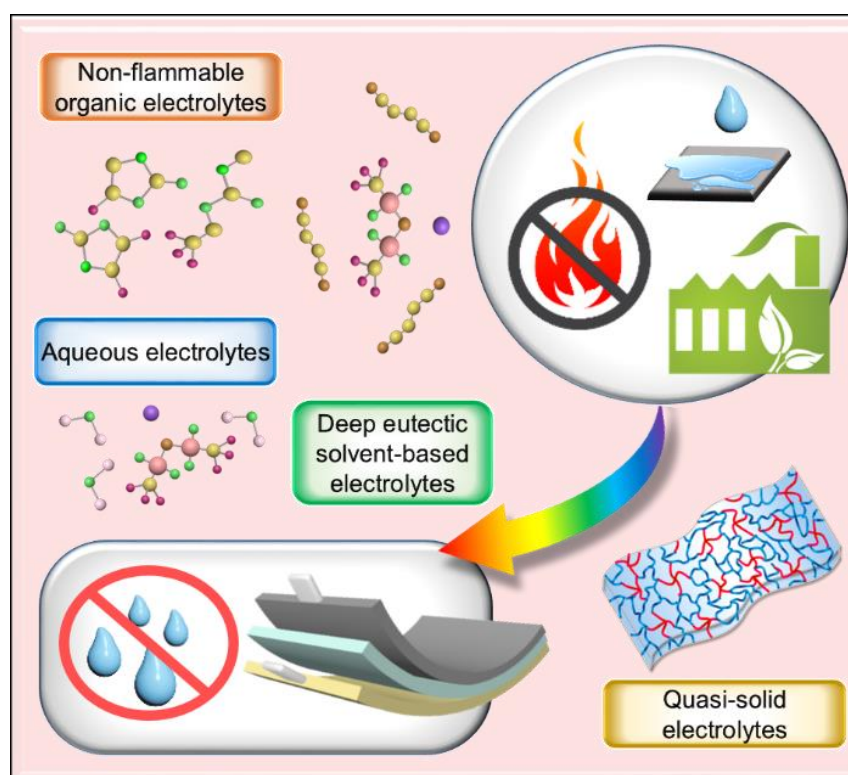
Abstract: Constructing an advanced artificial solid electrolyte interphase (SEI) on lithium metal anodes is a promising strategy to protect Li anodes and enable them to maintain long-term cycling stability and safety. Herein, the development of a dual-protective interface as an artificial SEI with high ionic conductivity and appropriate mechanical strength to protect Li anodes from parasitic reactions and dendrite formation is reported. The dual-protective interface consists of a Prussian blue (PB) inner layer and a reduced graphene oxide (rGO) outer layer. The compact and uniform PB layer with abundant Li-ion diffusion channels facilitates fast and uniform Li-ion flux to or from the surface of the Li metal anode, guiding uniform Li deposition without dendrite formation. In addition, the flexible rGO layer on the top of the PB layer enhances the structural integrity of the PB film against severe volume change during repeated Li plating and stripping. As a result, the Li metal anodes with the dual-protective interfaces show significantly improved cycling stability with high Coulombic efficiency and dendrite-free morphology. This work provides a new strategy to enhance the stability and safety of Li metal anodes for lithium metal batteries.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202102242>

11. Pauline Jaumaux, Junru Wu, Devaraj Shanmukaraj, Yizhou Wang, Dong Zhou*, Bing Sun, Feiyu Kang, Baohua Li*, Michel Armand*, **Guoxiu Wang*** “Non-Flammable Liquid and Quasi-Solid Electrolytes Towards Highly-Safe Alkali Metal-Based Batteries”, **Advanced Functional Materials**, 2021, 31, 2008644. IF=18.808. DOI: 10.1002/adfm.202008644.

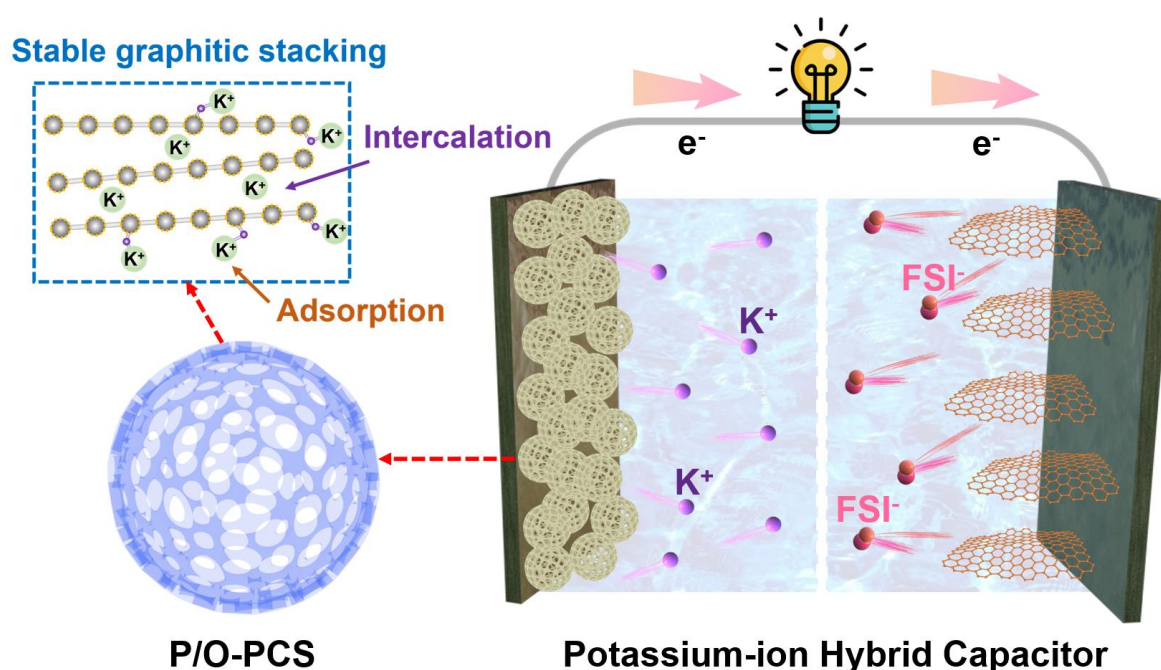
Abstract: Rechargeable alkali metal (i.e., lithium, sodium, potassium)-based batteries have been considered as vital energy storage technologies in modern society. However, the traditional liquid electrolytes applied in alkali metal-based batteries mainly consist of thermally unstable salts and highly flammable organic solvents, which triggers numerous accidents related to fire, explosion and leakage of toxic chemicals. Therefore, exploring non-flammable electrolytes is of paramount importance for achieving safe batteries. Although replacing traditional liquid electrolytes with all-solid-state electrolytes is the ultimate way to solve the above safety issues, developing non-flammable liquid electrolytes can more directly fulfil the current needs considering the low ionic conductivities and inferior interfacial properties of existing all-solid-state electrolytes. Moreover, the electrolyte leakage concern can be further resolved by gelling non-flammable liquid electrolytes to obtain quasi-solid electrolytes. Herein, we provide a comprehensive review on the latest progress of emerging non-flammable liquid electrolytes, including non-flammable organic liquid electrolytes, aqueous electrolytes and deep eutectic solvent-based electrolytes, and systematically introduce their flame-retardant mechanisms and electrochemical behaviors in alkali metal-based batteries. Then, we also summarized the gelation techniques for preparing quasi-solid electrolytes. Finally, the remaining challenges and future perspectives are presented. We anticipate that this review will promote a safety improvement of alkali metal-based batteries.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adfm.202008644>

12. Shuoqing Zhao, Kang Yan, Jiayu Liang, Qinghong Yuan, Jinqiang Zhang, Bing Sun*, Paul Munroe and **Guoxiu Wang***, “Phosphorus and Oxygen Dual-doped Porous Carbon Spheres with Enhanced Reaction Kinetics as Anode Materials for High-performance Potassium-ion Hybrid Capacitors”, **Advanced Functional Materials**, 2021, 31, 2102060. IF=18.808. DOI: 10.1002/adfm.202102060.

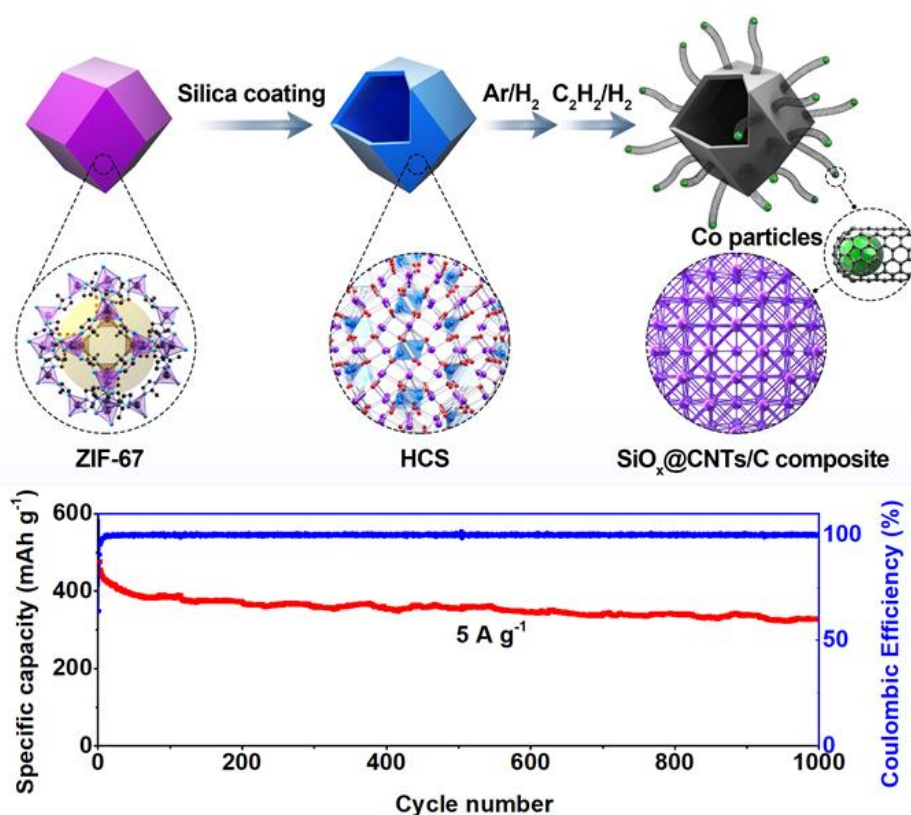
Abstract: Hard carbons with low cost and high specific capacity hold great potential as anode materials for potassium-based energy storage. However, their sluggish reaction kinetics and inevitable volume expansion degrade their electrochemical performances. Through rational nanostructure design and the heteroatom doping strategy, herein, we report the synthesis of phosphorus/oxygen dual-doped porous carbon spheres, which possess expanded interlayer distances, abundant redox active sites and oxygen-rich defects. The as-developed battery-type anode material shows high discharge capacity (401 mAh g^{-1} at 0.1 A g^{-1}), outstanding rate capability, and ultralong cycling stability (89.8% after 10000 cycles). In situ Raman spectroscopy and density functional theory calculations further confirm that the formation of P-C and P-O/P-OH bonds not only improves structural stability, but also contributes to a rapid surface-controlled potassium adsorption process. As a proof of concept, a potassium-ion hybrid capacitor was assembled by a dual-doped porous carbon sphere anode and an activated carbon cathode. It shows superior electrochemical performances, which opens a new avenue for innovative potassium-based energy storage technology.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adfm.202102060>

13. Hao Tian, Huajun Tian, Wu Yang, Fan Zhang, Wang Yang, Qiaobao Zhang*, Yong Wang, Jian Liu*, S. Ravi P. Silva, Hao Liu* and **Guoxiu Wang*** “Stable hollow-structured silicon suboxide-based anodes toward high-performance lithium-ion batteries”, **Advanced Functional Materials**, 2021, 31, 2101796. IF=18.808. DOI: 10.1002/adfm.202101796.

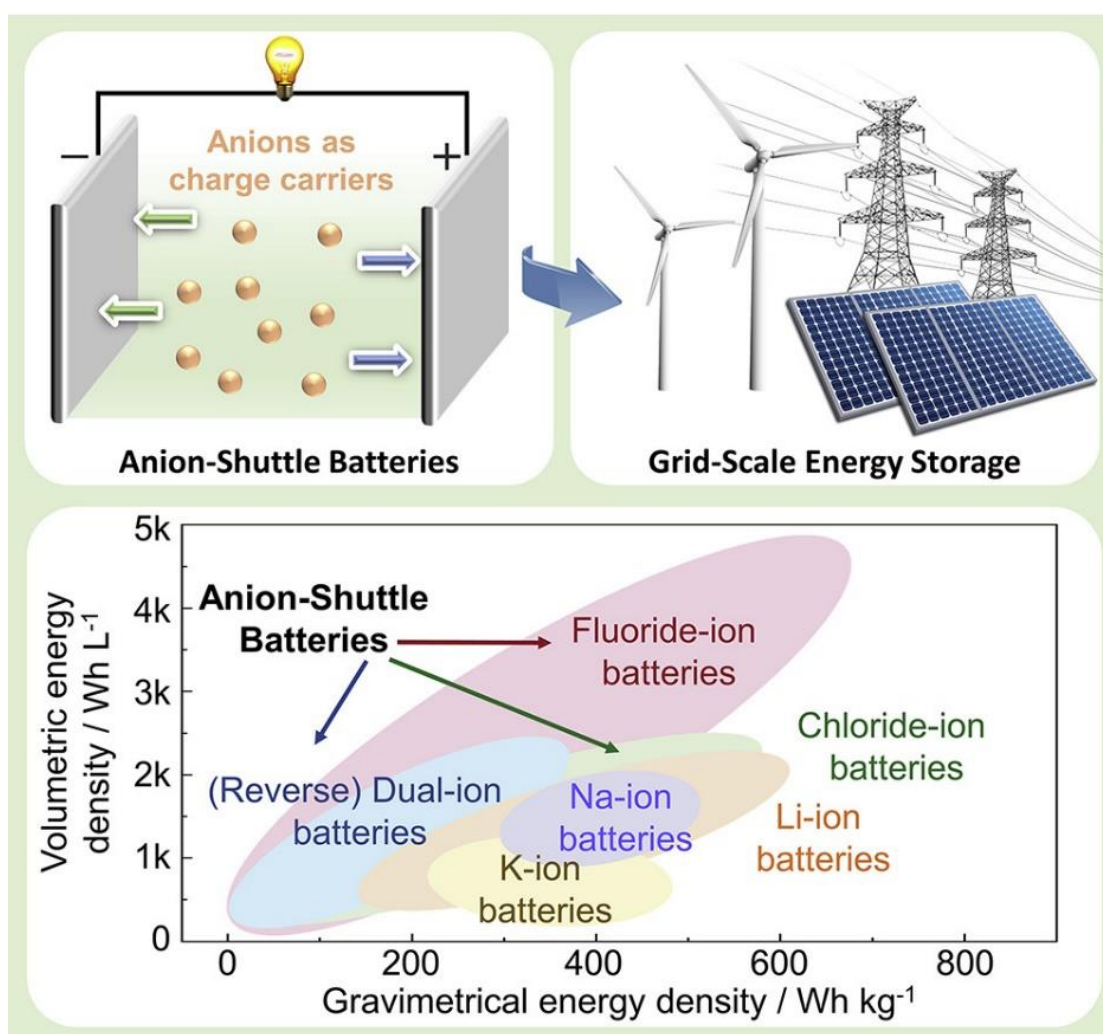
Abstract: Silicon (Si) has been regarded as an attractive high-capacity anode material for next-generation lithium-ion batteries (LIBs). However, Si anodes suffer from huge volume variation during cycling, which poses a critical challenge for stable battery operation. Compared with Si, Si suboxide (SiO_x) is one of the most promising candidates for high-energy-density LIBs because of its alleviated swelling and highly stable cycling performance. Whereas, the poor electronic conductivity and low (initial) Coulombic efficiency of SiO_x anodes severely hinder practical applications for LIBs. Herein, for the first time, we successfully solve these issues through rationally designing hollow-structured SiO_x @carbon nanotubes (CNTs)/C architectures with graphitic carbon coatings and in-situ growth of CNTs. When applied as anodes in LIBs, the SiO_x @CNTs/C anodes exhibit high reversible capacity, high initial Coulombic efficiency (88%), outstanding cycling performance and extraordinary mechanical strength during the calendaring process (200 MPa). This work paves the way for developing SiO_x -based anode materials for high-energy-density LIBs.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/adfm.202101796>

14. Qi Liu, Yizhou Wang, Xu Yang, Dong Zhou*, Xianshu Wang, Pauline Jaumaux, Feiyu Kang, Baohua Li*, Xiulei Ji*, **Guoxiu Wang*** “Rechargeable anion-shuttle batteries for low-cost energy storage”, **Chem**, 2021, 7, 1993-2021. IF: 22.804. DOI: 10.1016/j.chempr.2021.02.004.

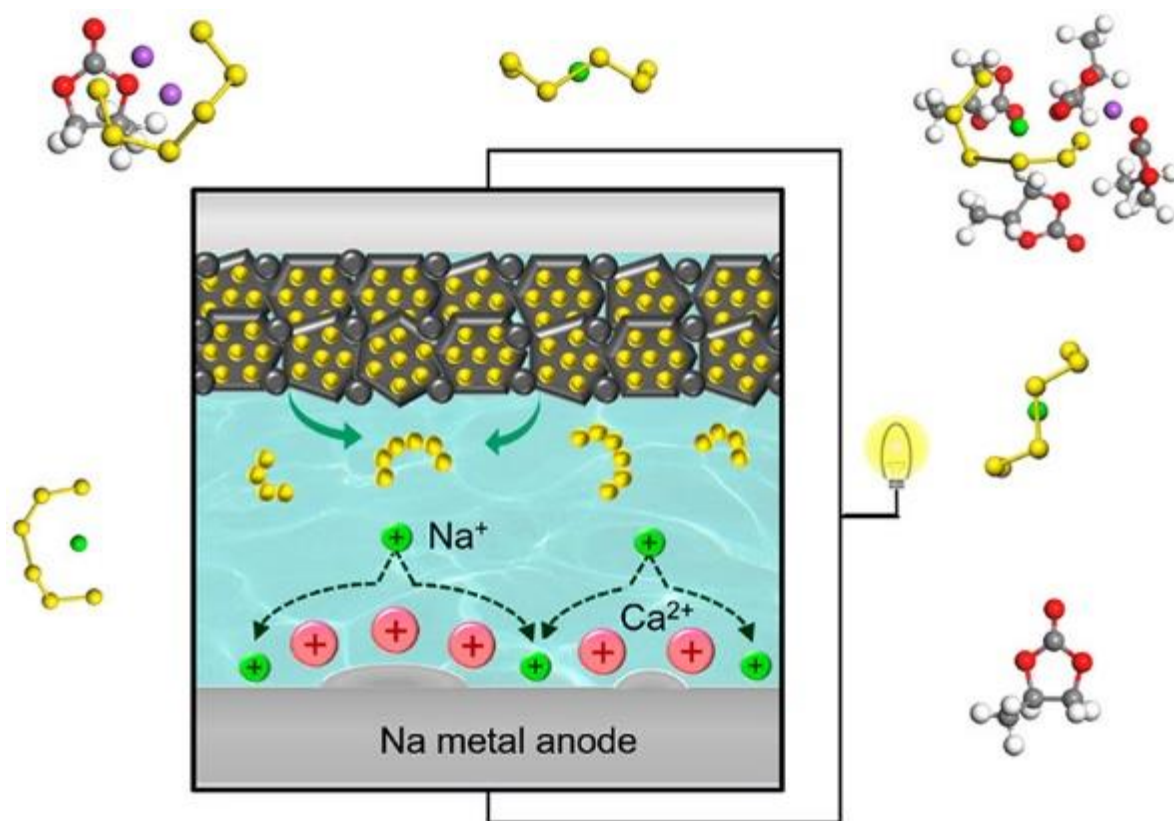
Abstract: As promising alternatives to lithium-ion batteries, rechargeable anion-shuttle batteries (ASBs) with anions as charge carriers stand out because of their low cost, long cyclic lifetime, and/or high energy density. In this review, we provide for the first time, comprehensive insights into the anion shuttling mechanisms of ASBs, including anion-based rocking-chair batteries (ARBs), dual-ion batteries (DIBs), including insertion-type, conversion-type, and conversion-insertion-type, and reverse dual-ion batteries (RDIBs). Thereafter, we review the latest progresses and challenges regarding electrode materials and electrolytes for ASBs. In addition, we summarize the existing dilemmas of ASBs and outline the perspective of ASB technology for future grid storage.



<https://www.sciencedirect.com/science/article/abs/pii/S2451929421000541>

15. Dong Zhou, Xiao Tang, Xiuyun Zhang, Fan Zhang, Junru Wu, Feiyu Kang, Baohua Li*, and **Guoxiu Wang***, “Multi-ion Strategy toward Highly Durable Calcium/Sodium–Sulfur Hybrid Battery”, **Nano Letters**, 2021, 21, 3548–3556. IF=11.189. DOI: 10.1021/acs.nanolett.1c00448.

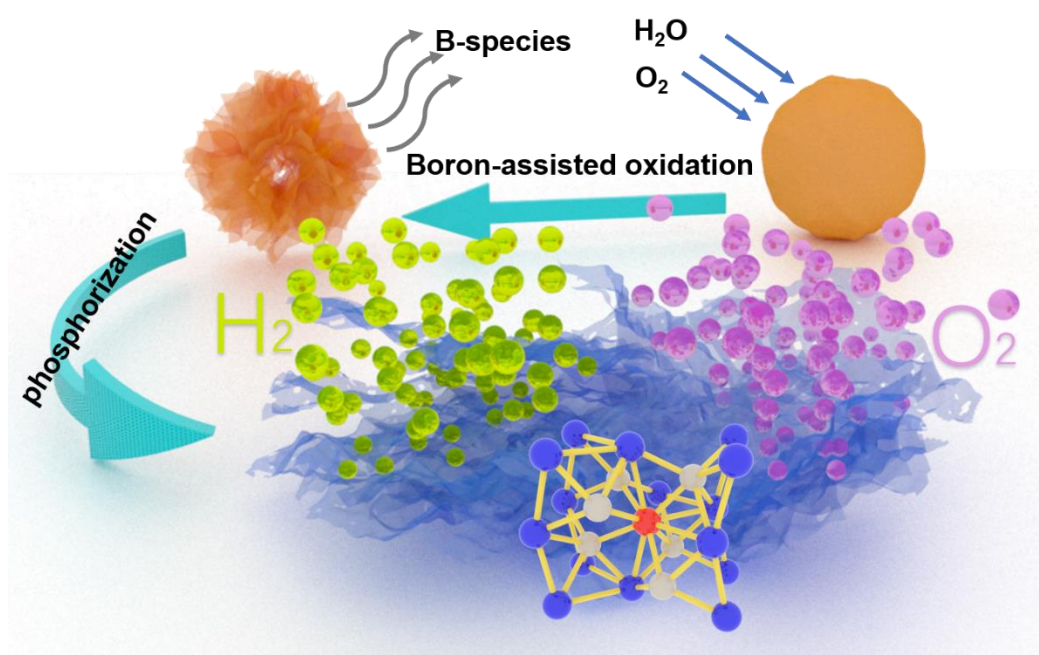
Abstract: Non lithium (Li) metal–sulfur batteries are a viable technology for large-scale energy storage due to their relative high energy densities and low cost. However, their practical application is still hindered by the insufficient reversibility and/or limited cycling stability. Herein, we report a high-performance calcium/sodium–sulfur (Ca/Na–S) hybrid battery enabled by a multi-ion chemistry. The introduction of Na ions in the electrolyte greatly boosts the conversion of Ca polysulfides, which has been verified by theoretical calculation and experimental investigation. Meanwhile, the presence of Ca ions constructs a protective electrostatic shield around the initial protrusions on the Na metal anode without prereduction, thus efficiently suppressing the Na dendrite growth. The as-developed Ca/Na–S cell exhibited a high reversible capacity of 947 mAh g⁻¹ at 0.1 C with long cycle life, clearly demonstrating the feasibility of this multi-ion strategy for developing low-cost non-Li metal–sulfur batteries.



<https://pubs.acs.org/doi/10.1021/acs.nanolett.1c00448>

16. Yufei Zhao, Jinqiang Zhang*, Yuhao Xie, Bing Sun, Junjie Jiang, Wen-Jie Jiang, Shibo Xi, Hui Ying Yang, Kang Yan, Shijian Wang, Xin Guo, Peng Li, Zhaojun Han, Xunyu Lu, Hao Liu*, and **Guoxiu Wang***, “Constructing Atomic Heterometallic Sites in Ultrathin Nickel-Incorporated Cobalt Phosphide Nanosheets via a Boron-Assisted Strategy for Highly Efficient Water Splitting”, **Nano Letters**, 2021, 21, 823-832. IF= 11.189. DOI: 10.1021/acs.nanolett.0c04569.

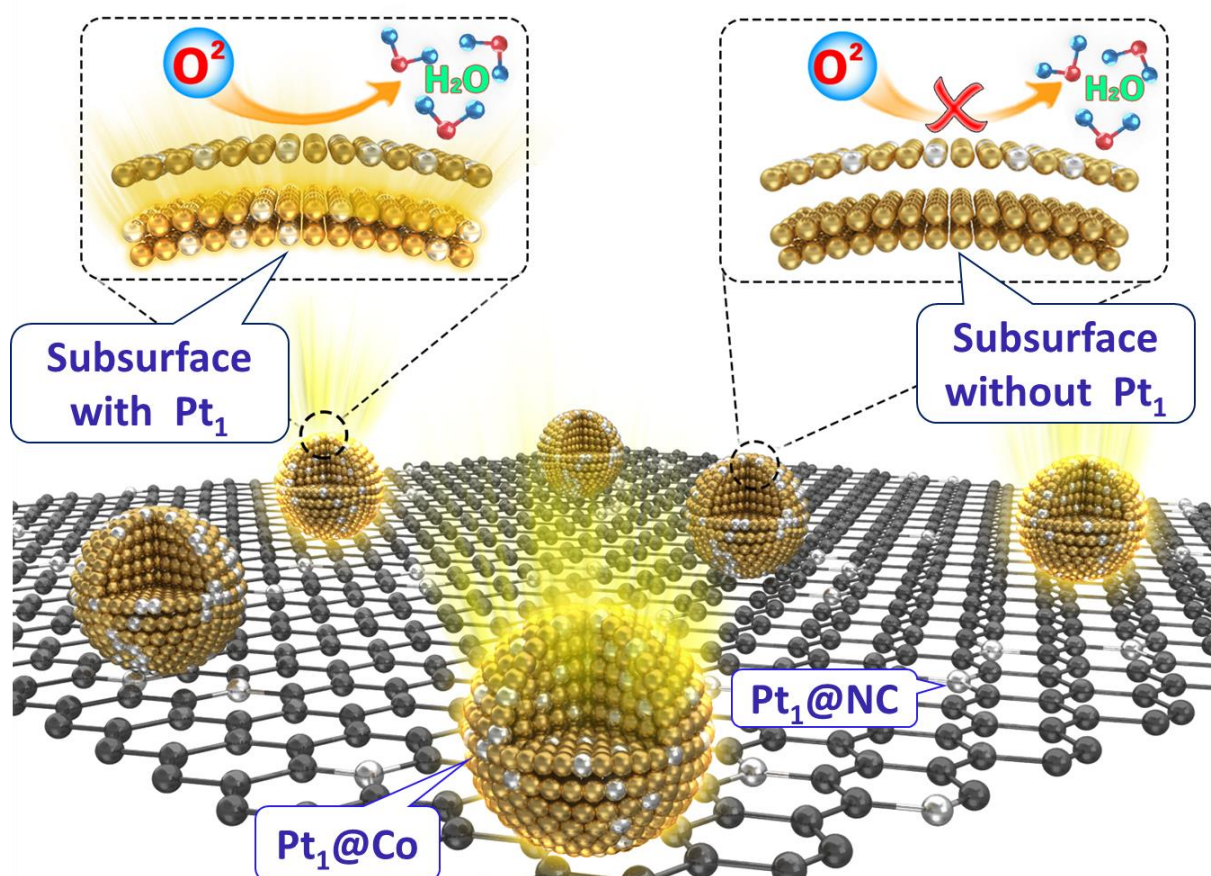
Abstract: The identification of active sites for highly efficient catalysts at the atomic scale for water splitting is still a great challenge. Herein, we fabricate ultrathin nickel-incorporated cobalt phosphide porous nanosheets (Ni-CoP) featuring an atomic heterometallic site ($\text{NiCo}_{16-x}\text{P}_6$) *via* a boron-assisted method. The presence of boron induces a release-and-oxidation mechanism, resulting in the gradual exfoliation of hydroxide nanosheets. After a subsequent phosphorization process, the resultant Ni-CoP nanosheets are implanted with unsaturated atomic heterometallic $\text{NiCo}_{16-x}\text{P}_6$ sites (with Co vacancies) for alkaline hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). The optimized Ni-CoP exhibits a low overpotential of 88 mV and 290 mV at 10 mA cm^{-2} for alkaline HER and OER, respectively. This can be attributed to reduced free energy barriers, owing to the direct influence of centre Ni atoms to the adjacent Co/P atoms in $\text{NiCo}_{16-x}\text{P}_6$ sites. These provide fundamental insights on the correlation between atomic structures and catalytic activity.



<https://pubs.acs.org/doi/10.1021/acs.nanolett.0c04569>

17. Wei-Hong Lai, Lifu Zhang, Zichao Yan, Weibo Hua, Sylvio Indris, Yaojie Lei, Hanwen Liu, Yun-Xiao Wang, Zhenpeng Hu, Hua Kun Liu, Shulei Chou, Guoxiu Wang, Shi Xue Dou. "Activating inert surface Pt single atoms via sub-surface doping for oxygen reduction reaction", *Nano Letters*, 2021. IF= 11.189. DOI: 10.1021/acs.nanolett.1c02013.

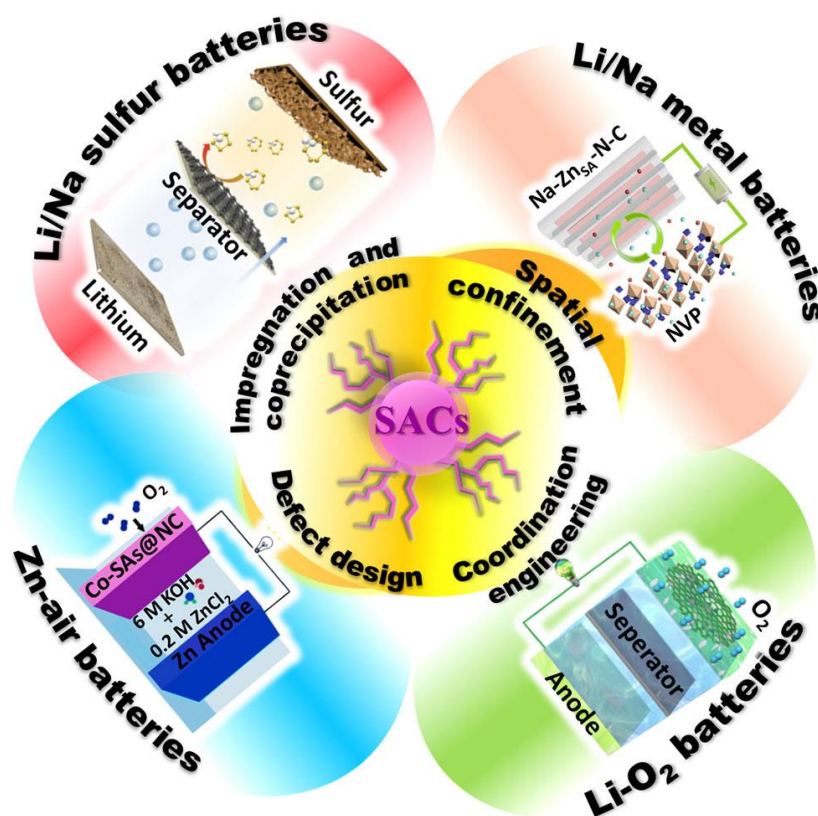
Abstract: The performance of single-atom catalysts strongly depends on their particular coordination environments in the near-surface region. Herein, we discover that engineering extra Pt single atoms in the subsurface ($\text{Pt}_{\text{subsurf}}$) can significantly enhance the catalytic efficiency of surface Pt single atoms toward the oxygen reduction reaction (ORR). We experimentally and theoretically investigated the effects of the $\text{Pt}_{\text{subsurf}}$ single atoms implanted in different positions of the subsurface of Co particles. The local environments and catalytic properties of surface Pt_1 are highly tunable via $\text{Pt}_{\text{subsurf}}$ doping. Specifically, the obtained $\text{Pt}_1@Co/NC$ catalyst displays a remarkable performance for ORR, achieving mass activity of $4.2 \text{ mA } \mu\text{gPt}^{-1}$ (28 times higher than that of commercial Pt/C) at 0.9 V versus reversible hydrogen electrode (RHE) in 0.1 M HClO_4 solution with high stability over 30000 cycles.



<https://pubs.acs.org/doi/10.1021/acs.nanolett.1c02013>

- 18.Hao Tian, Ailing Song, Huajun Tian, Jian Liu, Guangjie Shao*, Hao Liu* and **Guoxiu Wang***, “Single-atom catalysts for high energy rechargeable batteries”, **Chemical Science**, 2021, 12, 7656-7676. IF=9.35. DOI: 10.1039/D1SC00716E.

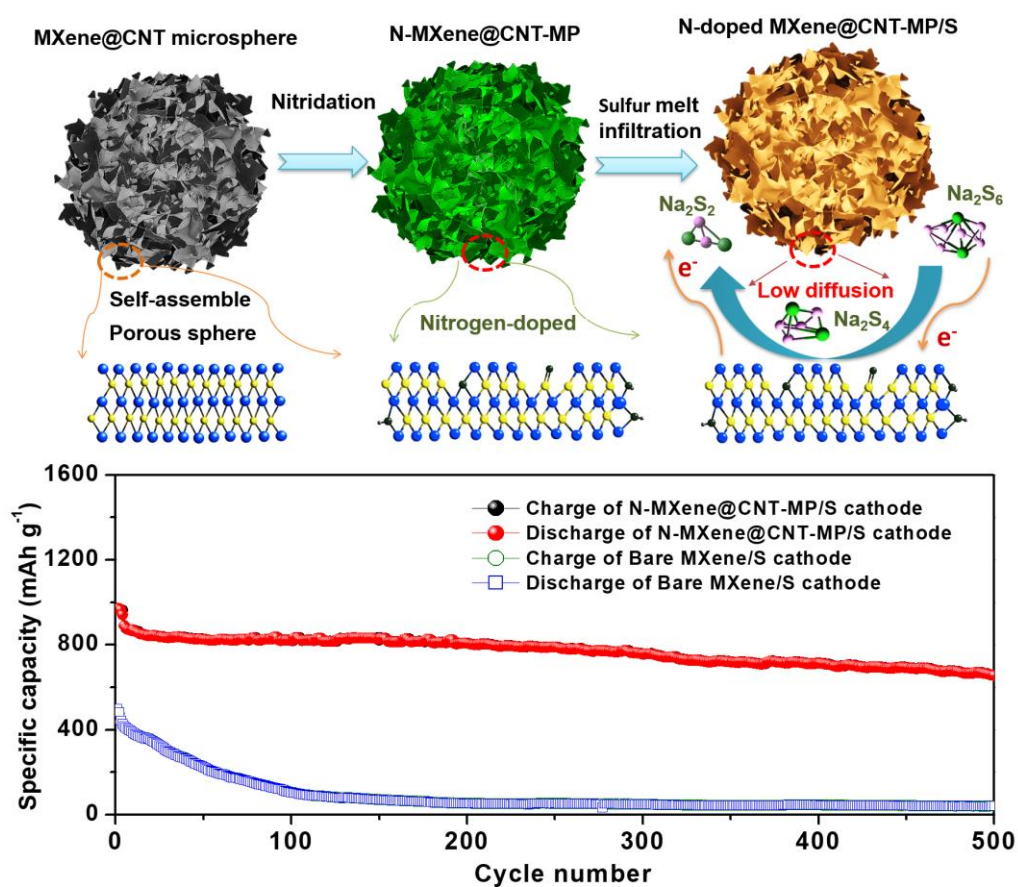
Abstract: Clean and sustainable electrochemical energy storage has attracted extensive attention. It remains great challenges to next-generation rechargeable battery systems with high energy density, good rate capability, excellent cycling stability, efficient active material utilization, and high Coulombic efficiency. Many catalysts have been explored to promote electrochemical reactions during the charge and discharge process. Among reported catalysts, single-atom catalysts (SACs) have attracted extensive attention due to their maximum atom utilization efficiency, homogenous active centres, and unique reaction mechanisms. In this perspective, we summarize the recent advances of the synthesis methods for SACs and highlight the recent progress of SACs for new generation rechargeable batteries including lithium/sodium metal batteries, lithium/sodium-sulfur batteries, lithium-oxygen batteries, and zinc-air batteries. The challenges and perspectives for the future development of SACs are discussed to shed light on future research of SACs for boosting performances of rechargeable batteries.



<https://doi.org/10.1039/D1SC00716E>

19. Weizhai Bao, Ronghao Wang, Chengfei Qian, Zherui Zhang, Ruijun Wu, Yuhao Zhang, Fangyang Liu*, Jingfa Li*, and **Guoxiu Wang***, “Porous Heteroatom-Doped $\text{Ti}_3\text{C}_2\text{T}_x$ MXene Microspheres Enable Strong Adsorption of Sodium Polysulfides for Long-Life Room-Temperature Sodium–Sulfur Batteries”, *ACS Nano*, 2021. IF=15.88. DOI: 10.1021/acsnano.1c05193.

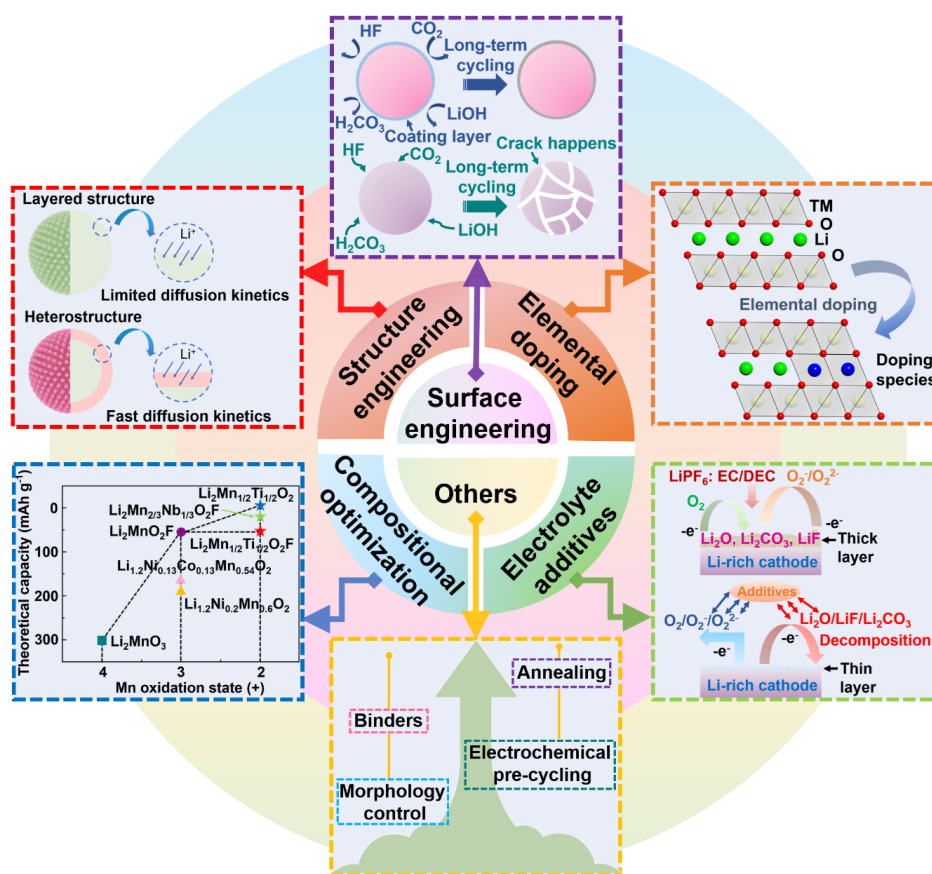
Abstract: The practical application of Na–S batteries is largely hindered by their low mass loading, inferior rate capability, and poor cycling performance. Herein, we report a design strategy for encapsulation of sodium polysulfides using $\text{Ti}_3\text{C}_2\text{T}_x$ MXene. Porous nitrogen-doped $\text{Ti}_3\text{C}_2\text{T}_x$ MXene microspheres have been synthesized by a facile synthesis method. Porous nitrogen-doped $\text{Ti}_3\text{C}_2\text{T}_x$ MXene microspheres contain abundant pore structures and heteroatom functional groups for structural and chemical synergistic encapsulation of sodium polysulfides. Sodium–sulfur batteries, based on the as-proposed cathode, demonstrated outstanding electrochemical performances, including a high reversible capacity (980 mAh g^{-1} at 0.5 C rate) and extended cycling stability (450.1 mAh g^{-1} at 2 C after 1000 cycles at a high areal sulfur loading of 5.5 mg cm^{-2}). This MXene-based hybrid material is a promising cathode host material for polysulfide-retention, enabling high-performance Na–S batteries.



<https://pubs.acs.org/doi/10.1021/acsnano.1c05193>

20. Shuoqing Zhao, Ziqi Guo, Kang Yan, Shuwei Wan, Fengrong He, Bing Sun* and **Guoxiu Wang***, “Towards High-energy-density Lithium-ion Batteries: Strategies for Developing High-capacity Lithium-rich Cathode Materials”, **Energy Storage Materials**, 2021, 34, 716-734. IF=16.28. DOI: 10.1016/j.ensm.2020.11.008.

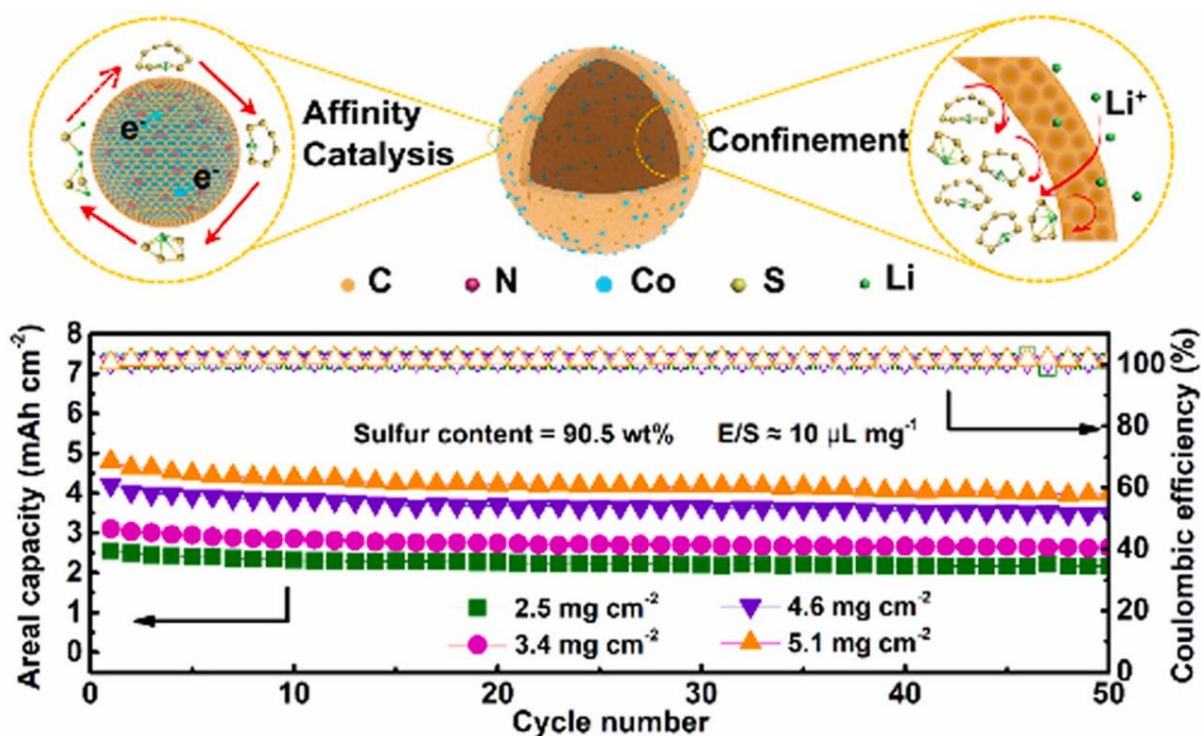
Abstract: With the growing demand for high-energy-density lithium-ion batteries, layered lithium-rich cathode materials with high specific capacity and low cost have been widely regarded as one of the most attractive candidates for next-generation lithium-ion batteries. However, issues such as voltage decay, capacity loss and sluggish reaction kinetics have hindered their further commercialization for decades. Intensive investigations have been devoted to developing high-performance lithium-rich cathode materials, highlighting the importance of improvement strategies as a potential approach. Herein, we summarize various strategies for improving performances of layered lithium-rich cathode materials for next-generation high-energy-density lithium-ion batteries. These include surface engineering, elemental doping, composition optimization, structure engineering and electrolyte additives, with emphasis on the effect and functional mechanism of corresponding techniques. In the subsequent section, we illustrate opportunities and challenges for designing high-performance lithium-rich cathode materials and bridging the gap between the laboratory and practical applications.



<https://www.sciencedirect.com/science/article/abs/pii/S2405829720304207>

21. Li Su, Jinqiang Zhang*, Yi Chen, Wu Yang, Jing Wang, Zhipeng Ma, Guangjie Shao*, **Guoxiu Wang***, “Cobalt-Embedded Hierarchically-Porous Hollow Carbon Microspheres as Multifunctional Confined Reactors for High-Loading Li-S Batteries”, *Nano Energy*, 2021, 85, 105981. IF=16.88. DOI: 10.1016/j.nanoen.2021.105981.

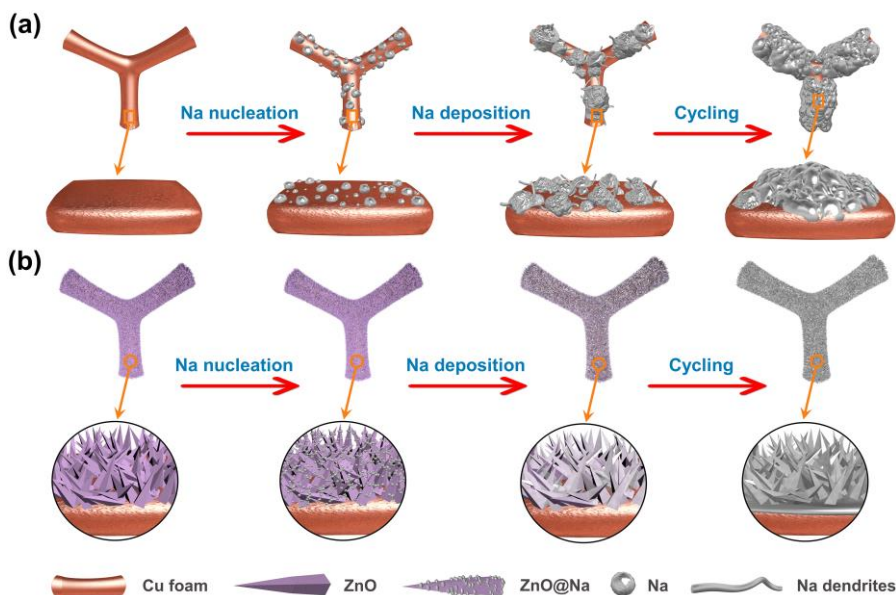
Abstract: The shuttle effect of dissolved polysulfides migrating to and depositing on anodes often leads to low round-trip efficiency and short cycle life for lithium-sulfur (Li-S) batteries. Herein, we report the rational design of cobalt-embedded nitrogen-doped hollow carbon microspheres (Co@N-HCMSs) as a multifunctional sulfur host for Li-S batteries. The hollow carbon microspheres exhibit large central cavities wrapped by a hierarchically porous shell, showing a large surface area of $1954 \text{ m}^2 \text{ g}^{-1}$. Furthermore, the carbon shells display a unique porous architecture, in which small pores are scattered on the outside and large pores are inside, thereby functioning as a selection barrier to confine polysulfides and diffuse Li^+ simultaneously. Moreover, the homogeneously distributed cobalt nanoparticles in the porous shell activate the surrounding N-doped carbon layer, which not only promote chemical adsorption of polysulfides but also catalyze polysulfide conversion. This facilitation effect has been confirmed by Bader charge and density function theory (DFT) calculations. When applied in Li-S batteries, the sulfur-impregnated Co@N-HCMSs cathode material exhibits excellent electrochemical performances, especially with a high sulfur content of 90.5 wt% and a high areal sulfur loading of 5.1 mg cm^{-2} .



<https://www.sciencedirect.com/science/article/abs/pii/S2211285521002391>

22. Wu Yang, Wang Yang, Liubing Dong, Guangjie Shao*, **Guoxiu Wang*** and Xinwen Peng*, “Hierarchical ZnO nanorod arrays grown on copper foam as an advanced three-dimensional skeleton for dendrite-free sodium metal anodes”, **Nano Energy**, 2021, 80, 105563. IF=16.88. DOI: 10.1016/j.nanoen.2020.105563.

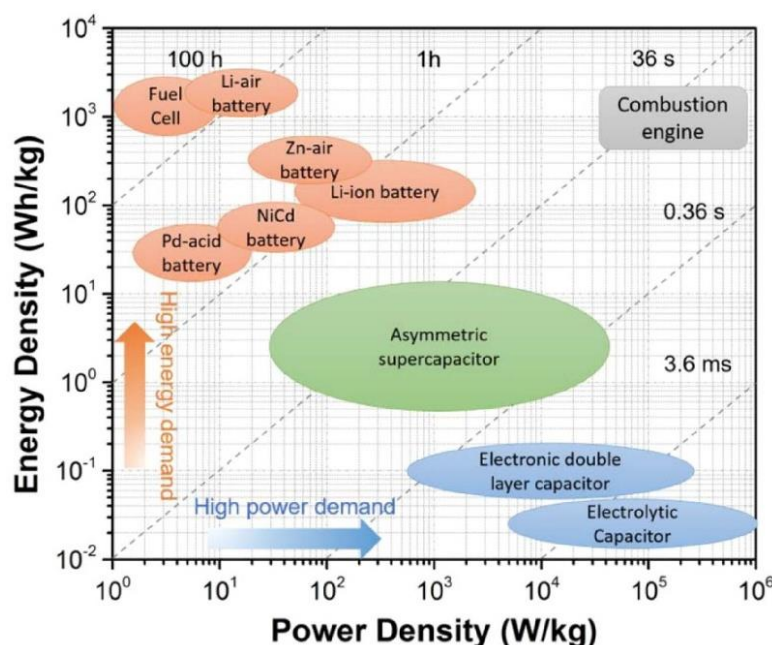
Abstract: Na metal is regarded as a potential anode material for next-generation Na metal batteries owing to its high theoretical capacity and low cost. However, the severe dendrite growth and infinitely volume changes of Na limit its practical applications as an anode material. In this study, a three-dimensional (3D) Cu foam skeleton with hierarchical ZnO nanorod arrays (CF@ZnO) was prepared as a stable host for dendrite-free Na metal anodes. Commercially available Cu foam was treated via a simple chemical precipitation method to grow hierarchical ZnO nanorod arrays, exhibiting a 3D porous structure with a cylindrical core-shell skeleton. The highly “sodiophilic” ZnO nanorod arrays provided abundant Na nucleation sites and exhibited low nucleation overpotential, which facilitated the homogeneous nucleation and uniform growth of Na on the electrode. Moreover, the 3D porous core-shell cylindrical structure of the nanorod arrays efficiently reduced the local effective current density, thus suppressing the growth of Na dendrites, as indicated by COMSOL Multiphysics simulations. As a result, the CF@ZnO electrode exhibited dendrite-free morphology during repeated Na plating/stripping and excellent cycling stability with very small voltage hysteresis even at high current densities. When paired with a $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ cathode, the CF@ZnO/Na electrode showed significant potential for application in full cells. This study provides a facile approach to design 3D sodiophilic hosts for high-energy-density Na metal batteries.



<https://www.sciencedirect.com/science/article/pii/S221128552031137X>

23. Jianmin Ma, Yutao Li, Nicholas S Grundish, John B Goodenough, Yuhui Chen, Limin Guo, Zhangquan Peng, Xiaoqun Qi, Fengyi Yang, Long Qie, Chang-An Wang, Bing Huang, Zeya Huang, Linhui Chen, Dawei Su, **Guoxiu Wang**, Xinwen Peng, Zehong Chen, Junliang Yang, Shiman He, Xu Zhang, Haijun Yu, Chaopeng Fu, Min Jiang, Wenzhuo Deng, Chuan-Fu Sun, Qingguang Pan, Yongbing Tang, Xianfeng Li, Xiulei Ji, Fang Wan, Zhiqiang Niu, Fang Lian, Caiyun Wang, Gordon G Wallace, Min Fan, Qinghai Meng, Sen Xin, Yu-Guo Guo and Li-Jun Wan, “The 2021 battery technology roadmap”, **Journal of Physics D: Applied Physics**, 2021, 54 ,183001. IF=3.041. DOI: 10.1088/1361-6463/abd353

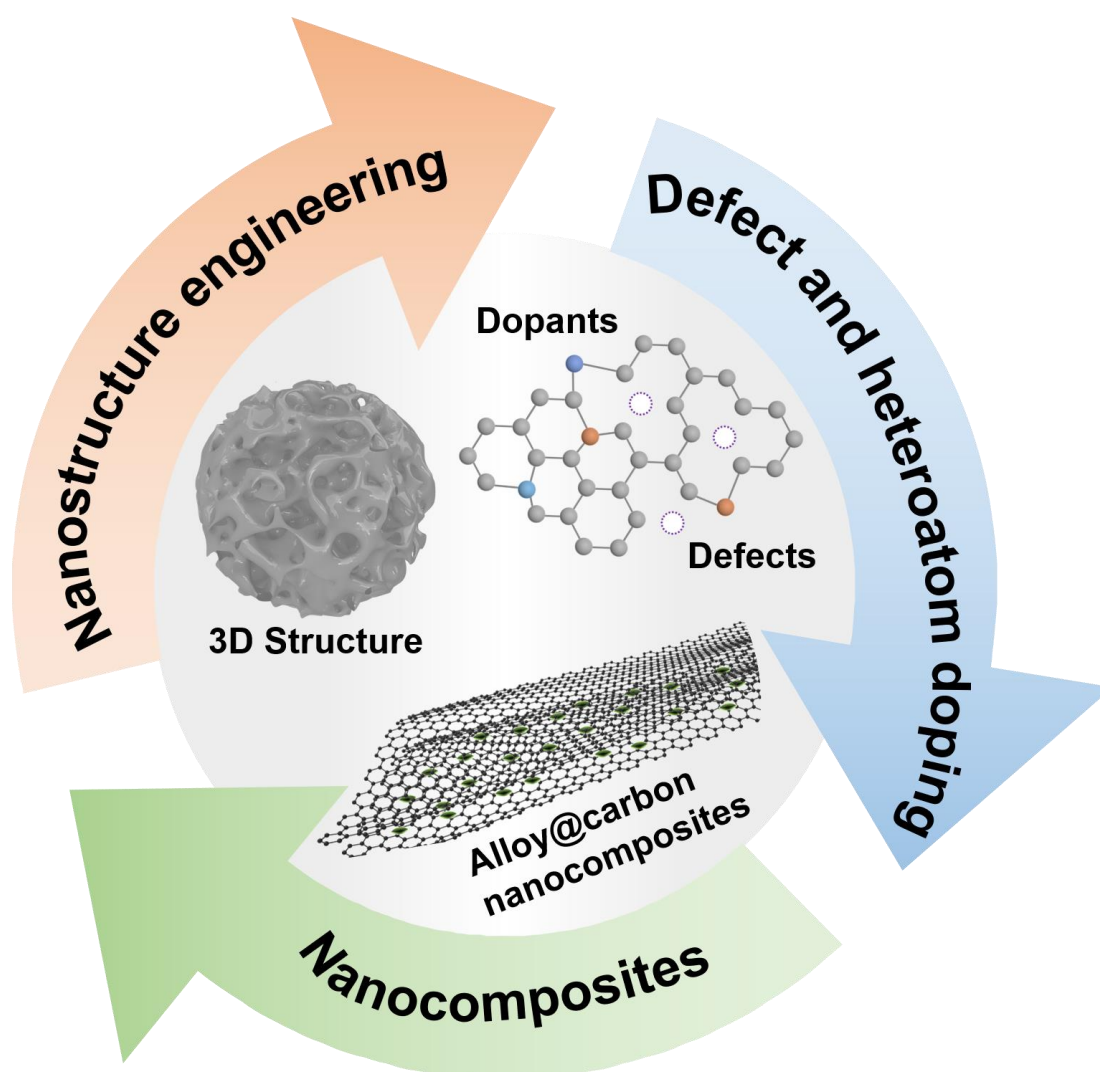
Abstract: Sun, wind and tides have huge potential in providing us electricity in an environmental-friendly way. However, its intermittency and non-dispatchability are major reasons preventing full-scale adoption of renewable energy generation. Energy storage will enable this adoption by enabling a constant and high-quality electricity supply from these systems. But which storage technology should be considered is one of important issues. Nowadays, great effort has been focused on various kinds of batteries to store energy, lithium-related batteries, sodium-related batteries, zinc-related batteries, aluminum-related batteries and so on. Some cathodes can be used for these batteries, such as sulfur, oxygen, layered compounds. In addition, the construction of these batteries can be changed into flexible, flow or solid-state types. There are many challenges in electrode materials, electrolytes and construction of these batteries and research related to the battery systems for energy storage is extremely active. With the myriad of technologies and their associated technological challenges, we were motivated to assemble this 2020 battery technology roadmap.



<https://iopscience.iop.org/article/10.1088/1361-6463/abd353>

24. Shuoqing Zhao, Ziqi Guo, Jian Yang, Chengyin Wang, Bing Sun* and Guoxiu Wang*, “Nanoengineering of Advanced Carbon Materials for Sodium-ion Batteries”, *Small*, 2021, 2007431. IF=12.02. DOI: 10.1002/sml.202007431.

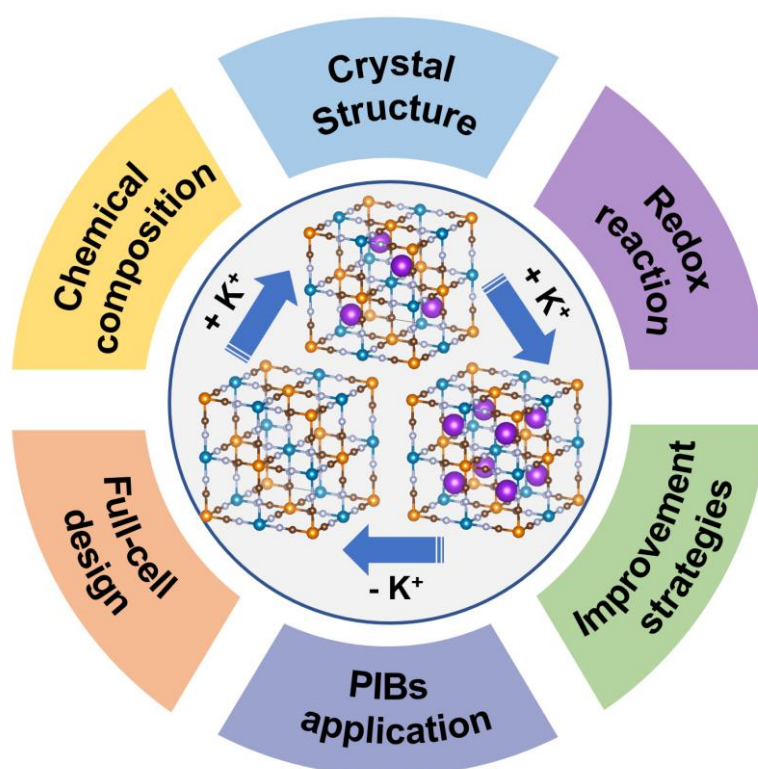
Abstract: Recent research shows that the continuing importance of carbon anode materials plays an important role in the development of sodium-ion batteries. Nevertheless, the practical deployment of sodium-ion batteries still faces many challenges such as mediocre sodium storage capability and short cycle life. Therefore, it is imperative to explore improvement methods to boost their competitiveness. Herein, various nanoengineering strategies, including nanostructure design, defect and heteroatom doping, and nanocomposite optimization, are proposed as reliable and effective approaches to improve electrochemical performances and structural stability of carbon-based anode materials for SIBs. We highlight the feasibility of nanoengineering as a promising approach to develop next-generation carbon materials for sodium-ion batteries.



<https://onlinelibrary.wiley.com/doi/abs/10.1002/sml.202007431>

25. Shuoqing Zhao, Ziqi Guo, Kang Yan, Xin Guo, Shuwei Wan, Fengrong He, Bing Sun* and Guoxiu Wang*, “The Rise of Prussian Blue Analogs: Challenges and Opportunities for High-performance Cathode Materials in Potassium-ion Batteries”, **Small Structures**, 2021, 2, 2000054. IF=11.459. DOI: 10.1002/sstr.202000054.

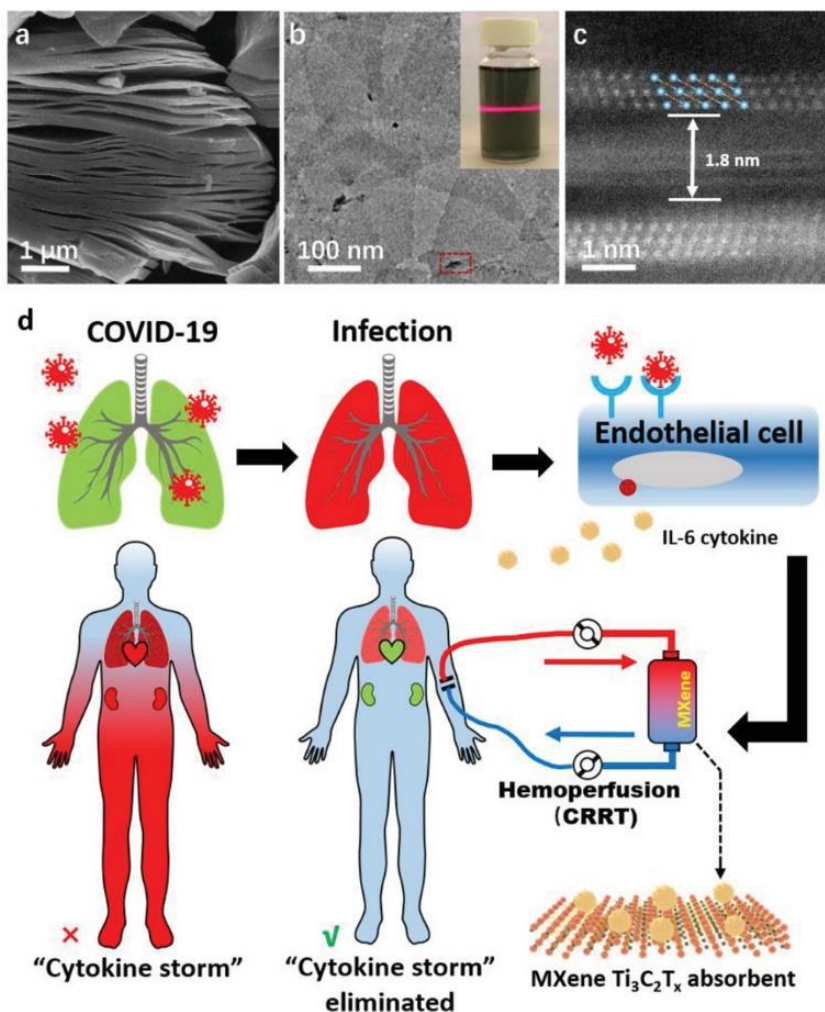
Abstract: Potassium-ion batteries are emerging as one of the potential alternatives to lithium-ion batteries for next-generation rechargeable battery systems. Nevertheless, the lack of suitable cathode materials with high capacity hinders their practical applications. Recently, Prussian blue analogs cathode materials stand out as promising candidates for potassium-ion batteries. Their unique crystal structure with open three-dimensional frameworks and large interstitial voids favors fast K^+ intercalation without causing drastic volume expansion, which is the prerequisite for high-rate and long-term battery operation. Herein, we present a fundamental review on the development and advance of Prussian blue analogs cathode materials for potassium-ion batteries with in-depth elucidation of their crystal structures, chemical compositions and electrochemical performances. Particularly, we highlight the unique and prominent advantages of Prussian blue analogs in both aqueous and non-aqueous potassium ion batteries. In addition, to bridge the current gap from the laboratory to future commercialization, potential improvement strategies are proposed to overcome the present drawbacks. Finally, we provide perspectives and new insights for further exploration and research in Prussian blue analogs for better potassium-ion batteries.



<https://onlinelibrary.wiley.com/doi/10.1002/sstr.202000054>

26. Tianyi Wang, Xiaoyu Sun, Xin Guo, Jinqiang Zhang, Shouxuan Tao, Jun Guan, Lin Zhou, Jie Han, Chengyin Wang, Hang Yao*, **Guoxiu Wang** *, “Ultraefficiently Calming Cytokine Storm using $Ti_3C_2T_x$ MXene”, **Small methods**, 2020, 5, 2001108. IF= 14.188. DOI: 10.1002/smt.202001108.

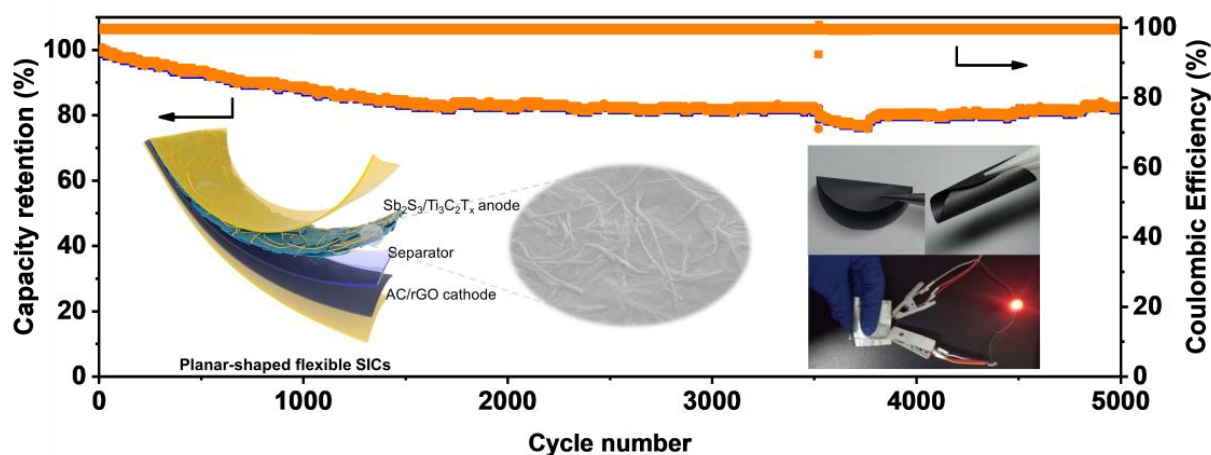
Abstract: During the global outbreak of COVID-19 pandemic, “cytokine storm” conditions are regarded as the fatal step resulting in most mortality. Hemoperfusion is widely used to remove cytokines from the blood of severely ill patients to prevent uncontrolled inflammation induced by a cytokine storm. This article discovers, for the first time, that 2D $Ti_3C_2T_x$ MXene sheet demonstrates an ultrahigh removal capability for typical cytokine interleukin-6. In particular, MXene shows a 13.4 times higher removal efficiency over traditional activated carbon absorbents. Molecular-level investigations reveal that MXene exhibits a strong chemisorption mechanism for immobilizing cytokine interleukin-6 molecules, which is different from activated carbon absorbents. MXene sheet also demonstrates excellent blood compatibility without any deleterious side influence on the composition of human blood. This work can open a new avenue to use MXene sheets as an ultraefficient hemoperfusion absorbent to eliminate the cytokine storm syndrome in treatment of severe COVID-19 patients.



<https://onlinelibrary.wiley.com/doi/10.1002/smt.202001108>

27. Jian Yang, Tianyi Wang, Xin Guo*, Xiaoxue Sheng, Jiabao Li, Chengyin Wang* and **Guoxiu Wang***, “Flexible sodium-ion capacitors boosted by high electrochemically-reactive and structurally-stable Sb_2S_3 nanowire/ $\text{Ti}_3\text{C}_2\text{T}_x$ MXene film anodes”, **Nano Research**, 2021. IF= 8.183. DOI: 10.1007/s12274-021-3933-7.

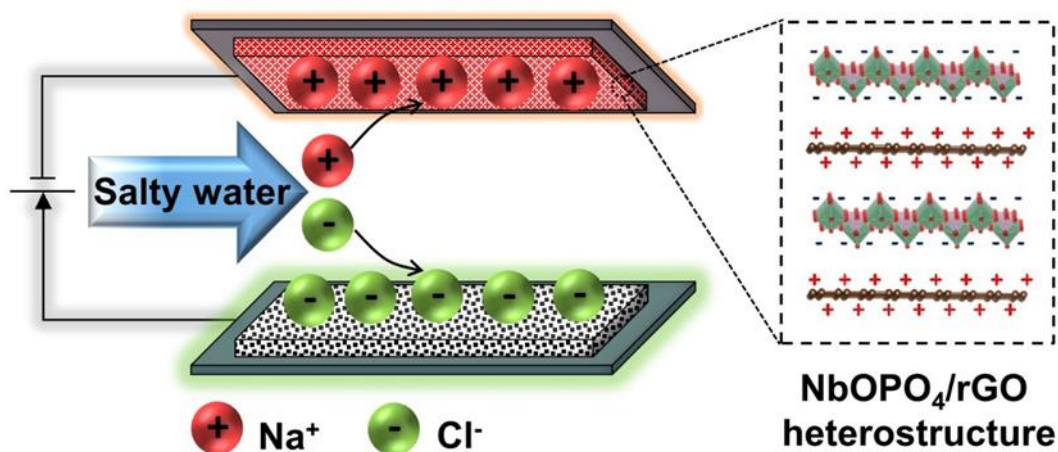
Abstract: The rapid development of portable, foldable, and wearable electronic devices requires flexible energy storage systems. Sodium-ion capacitors (SICs) combining the high energy of batteries and the high power of supercapacitors are promising solutions. However, the lack of flexible and durable electrode materials that allow fast and reversible Na^+ storage hinders the development of flexible SICs. Herein, we report a high-capacity, free-standing and flexible $\text{Sb}_2\text{S}_3/\text{Ti}_3\text{C}_2\text{T}_x$ composite film for fast and stable sodium storage. In this hybrid nano-architecture, the Sb_2S_3 nanowires uniformly anchored between $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets not only act as sodium storage reservoirs but also pillar the 2D $\text{Ti}_3\text{C}_2\text{T}_x$ to form 3D channels benefiting for electrolyte penetration. Meanwhile, the highly conductive $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets provide rapid electron transport pathways, confine the volume expansion of Sb_2S_3 during sodiation, and restrain the dissolution of discharged sodium polysulfides through physical constraint and chemical absorption. Owing to the synergistic effects of the 1D Sb_2S_3 nanowires and 2D MXenes, the resultant composite anodes exhibit outstanding electrochemical performance for sodium storage. Moreover, the flexible SICs using $\text{Sb}_2\text{S}_3/\text{Ti}_3\text{C}_2\text{T}_x$ anodes and active carbon/reduced graphene oxide (AC/rGO) paper cathodes deliver a superior energy and power density in comparison with previously reported devices, as well as an excellent cycling performance with a high capacity retention of 82.78% after 5000 cycles. This work sheds light on the design of next-generation low-cost, flexible and fast-charging energy storage devices.



<https://link.springer.com/article/10.1007/s12274-021-3933-7>

28. Javad Safaei, Seyed Mostafa Hosseinpour Mashkani, Hao Tian, Caichao Ye, Pan Xiong, **Guoxiu Wang***, “Self-Assembled NbOPO₄ Nanosheet/Reduced Graphene Oxide Heterostructure for Capacitive Desalination”, **ACS Applied Nano Materials**, 2021, 4, 12629–12639. IF = 5.097. DOI: 10.1021/acsnm.1c03180.

Abstract: Capacitive deionization (CDI) is a promising and energy-efficient technology for desalination. The development of high-performance capacitive electrodes is essential for enhancing the CDI properties for practical applications. Here, a 2D heterostructure was rationally designed and synthesized by face-to-face restacking of NbOPO₄ nanosheets and reduced graphene oxide (rGO) via an electrostatic self-assembly process. The as-prepared 2D NbOPO₄/rGO heterostructure achieved an excellent ion storage capacity, electronic conductivity, and unimpeded ion kinetics. When applied as electrodes for CDI, the 2D NbOPO₄/rGO heterostructure delivered a high specific capacitance of 258.3 F g⁻¹ and an electrosorption capacity of 73 mg g⁻¹ for NaCl solution of 10 000 mg L⁻¹ at an applied voltage of 1.2 V, which is more than five times larger than that of activated carbon. The heterostructure electrode also showed high desalination stability for up to 50 adsorption/desorption cycles. The high CDI performance is attributed to the strong 2D/2D coupling between NbOPO₄ nanosheets and rGO. The strong 2D/2D coupling reduced the charge transfer resistance, affirmed via the electrochemical impedance spectroscopy technique, attesting to the enhanced charge transportation across the heterointerface. The robust 2D/2D coupling was affirmed via the uniform and identical Raman shifts at various random regions, and larger XPS binding energy shifts for the self-assembled NbOPO₄/rGO heterostructure. This work demonstrated the potential of self-assembled nanoheterostructures for water desalination via capacitive deionization.



<https://pubs.acs.org/doi/abs/10.1021/acsnm.1c03180>