Molybdenum-based materials for sodium-ion batteries

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Abstract
Sodium-ion batteries are considered one of the most promising candidates for affordable and scalable energy storage as required in smart grid and renewable energy. One of the principal challenges sodium-ion batteries being faced is to search suitable anode materials that can accommodate and store large amounts of Na+ ions reversibly and sustainably at reasonable galvanostatic rates. Molybdenum-based materials such as oxides and sulfides might meet this challenge as they afford a capacity much greater than those of the carbonaceous materials and exhibit rich Na-reaction chemistry. However, these materials are facing several technical issues, such as multiple-phase transformation, particle pulverization as the result of volume swelling, and low surface activity during sodiation/desodiation. To tackle these issues, materials design and engineering are of indispensability. In this brief review, we present a state-of-the-art overview of the research progress of molybdenum-based materials for sodium storage, and highlight materials engineering strategies that are capable of addressing the mentioned challenges. We also offer valuable insights into their further development direction and discuss their potentiality in practical batteries.

KEYWORDS
materials engineering, molybdenum-based materials, sodium-ion batteries

1 | INTRODUCTION

The excessive consumption of fossil fuels and the consequent environmental pollution have been driving us to consider utilizing renewable energy.1-3 However, many of the renewable energies such as solar and wind energies are intermittent, so that the integration of energy storage into these renewable systems is indispensable.4,5 Among various energy storage technologies, the currently available, electrochemical rechargeable batteries and sodium-ion batteries (SIBs) in particular are a key solution of choice due to the virtually unlimited sodium resource and the similarity in electrochemical behavior to that of lithium.6-11

Currently, one of the key challenges the application of SIBs being facing is lack of satisfactory anode materials. The larger radius of Na+ ion (0.102 nm) compared to that of Li+ ion (0.076 nm) presents a core challenge to the host structure. As a result, the prevailing graphite anode for rechargeable lithium-ion batteries only exhibits a negligible capacity for Na+ ion. Meanwhile, the standard electrode potential of Na+/Na is higher than that of Li+/Li, which reduces the operating voltage and the energy of full cells.12-14 To achieve a performance...
comparable to lithium technology, it is imperative to explore Na anode materials with large capacity, rapid reaction kinetics, and long cycle life.\textsuperscript{15-18}

Anode materials based on transition metal elements have drawn substantial attention in recent years. Particularly, materials based on molybdenum (Mo) have been regarded as promising electrode materials due to their unique structural and performance advantages.\textsuperscript{19} Mo is inexpensive and abundant in the earth’s crust, and most Mo materials have a high theoretical capacity as a result of intercalation and/or conversion reactions, as summarized in Figure 1. For example, the theoretical capacity of MoO\textsubscript{3} is 1117 mAh g\textsuperscript{−1},\textsuperscript{20} which is more than 3 times of TiO\textsubscript{2} (335 mAh g\textsuperscript{−1}).\textsuperscript{21,22} Moreover, owing to multiple valence states with varied coordination numbers, Mo-based compounds show rich structural chemistry with a variety of stoichiometries such as oxides (MoO\textsubscript{3}, MoO\textsubscript{2}), dichalcogenides (MoS\textsubscript{2}, MoSe\textsubscript{2}), and carbides (Mo\textsubscript{2}C, MoC). Their representative crystal structures are illustrated in Figure 2.

Similar to other transition metal compounds,\textsuperscript{23-26} Mo-based electrode materials suffer from many practical challenges, such as multiple-phase transformation, serious electrode pulverization, slow electron/ion transfer, and low surface reactivity.\textsuperscript{27} Therefore, to fully utilize the advantageous of Mo-based electrode materials for SIBs, the Mo-based electrode materials must be tailored and engineered to mitigate these challenges (Figure 3). Over the past years, researches in this area are so rigorous and significant progress has been achieved. Therefore, it is desirable to summarize the state-of-the-art of the advancement of the Mo-based compounds in the sodium storage application. Although several excellent reviews regarding Mo-based electrode materials have been published recently,\textsuperscript{19,28} they are primarily focused on the application in lithium storage. In this review, however, we will solely deal with their application in sodium storage. We hope this work will sever as a guidance for the further advancing of Mo materials or other transition metal compounds.

2 | MOLYBDENUM OXIDES

Molybdenum oxides exhibit a wealth of structural diversity and unique electrochemical properties with a large range of applications. Molybdenum trioxide (MoO\textsubscript{3}) and dioxide (MoO\textsubscript{2}) are two typical compounds that have captured tremendous attentions in the battery applications.

2.1 | Molybdenum trioxide

MoO\textsubscript{3} contains two well-known phases based on the MoO\textsubscript{6} octahedron building block, that is, thermodynamically stable orthorhombic α-phase (α-MoO\textsubscript{3}) and metastable monoclinic β-phase (β-MoO\textsubscript{3}). α-MoO\textsubscript{3} has a layered structure that is made of atomically thin sheets along [010] direction with a wide bandgap energy of about 3 eV. The thickness of the sheet is about 0.7 nm, the distance between the two layers is 1.4 nm, and the van der Waals interaction would be the major binding force between the two layers.\textsuperscript{27}

The electrochemical storage of sodium ions in MoO\textsubscript{3} has been extensively investigated since 1995. In the voltage range of 1.0–4.0 V, insertion reaction is the main mechanism for Na\textsuperscript{+} ion storage in MoO\textsubscript{3}. Specifically, a reduction potential of about 2.4 V versus Na/Na\textsuperscript{+} indicates the formation of a sodium bronze (Na\textsubscript{x}MoO\textsubscript{3}), which will further reduce to Na\textsubscript{x}MoO\textsubscript{3} at a lower potential.\textsuperscript{29} In the potential window between 0.01 and 3 V, MoO\textsubscript{3} serves as an anode and its sodiation process involves both insertion and conversion reactions. The overall reaction is MoO\textsubscript{3} + 6Na\textsuperscript{+} + 6e\textsuperscript{−} ↔ Mo + 3Na\textsubscript{2}O with a theoretical capacity of 1117 mAh g\textsuperscript{−1}. Due to significant volume swelling upon the insertion of Na\textsuperscript{+} ions, the reversibility of this reaction is poor.\textsuperscript{30} Recently, by in situ transmission electron microscopy (TEM), Xia et al unveiled that Mo\textsuperscript{6+} can be fully reduced to Mo upon sodiation, but only re-oxidized to Mo\textsuperscript{4+} upon subsequent desodiation (Mo + 3Na\textsubscript{2}O → Na\textsubscript{2}MoO\textsubscript{3} + 4Na\textsuperscript{+} + 4e\textsuperscript{−}).\textsuperscript{31} This clearly explains the limited reversibility of MoO\textsubscript{3}.

In an early work, Hariharan et al. demonstrated the feasibility of reversible sodium storage in α-MoO\textsubscript{3} anode material.\textsuperscript{20} They reported a specific capacity of 771 mAh g\textsuperscript{−1} in the first discharge and 410 mAh g\textsuperscript{−1} in the following charge. A full cell with the α-MoO\textsubscript{3} anode and Na\textsubscript{3}V\textsubscript{2}(PO\textsubscript{4})\textsubscript{3} cathode was capable of affording an average voltage of 1.4 V and a discharge capacity of 164 mAh g\textsuperscript{−1}. However, bulk MoO\textsubscript{3} frequently suffers from sluggish Na-reaction kinetics due to low electronic

![Figure 1](image-url) Comparison between gravimetric and volumetric capacities of major molybdenum-based materials

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conductivity ($10^{-5}$ S cm$^{-1}$),$^{32}$ large ion transport distance, and poor electrochemical activity. Also, bulk MoO$_3$ shows poor cycling stability due to large volume expansion during sodiation. To solve these problems, two general materials strategies have been proposed, one of which is to design and produce nanomaterials or nanostructures of MoO$_3$, which exhibits a larger surface area and shorter ion transport path versus bulk phase. The other one is to tailor the intrinsic property of MoO$_3$ by materials engineering.

Tailoring the intrinsic property can be realized by the introduction of defects or exotic elements. Oxygen vacancy is a typical defect capable of tuning the electric properties of oxides. Figure 4A,B shows oxygen-deficient MoO$_3$ (MoO$_3$$_{-x}$) nanosheet electrodes for sodium storage.$^{35}$ Oxygen vacancy brought the color of MoO$_3$ nanosheets from white to blue, and significantly boosted the electronic conductivity and Na$^+$ diffusivity.$^{37}$ However, the high conductivity of MoO$_3$$_{-x}$ may not be well preserved upon Na cycling, as the desodiated phase cannot recover to MoO$_3$$_{-x}$.
Modulation of MoO₃ can also be done by additional phases such as nanocarbons, which are highly conductive, flexible, and resilient.³⁸,³⁹ Previously, we have constructed MoO₃/CNT hybrid via a simple hydrothermal reaction and achieved a high Li-storage activity.³⁷ By analogy, Figure 4C,D shows a composite of MoO₃ and reduced graphite oxide (rGO) showing a reversible capacity of 665 mAh g⁻¹ over 100 cycles.³⁶ This capacity is the highest value reported for cycled MoO₃ to date. It was proposed that the Mo–O–C bonds might contribute to such impressive cycling stability. Alternatively, Xu et al. utilized an ultrathin (~0.8 nm) Al₂O₃ layer by atomic layer deposition to improve the Na cycling of MoO₃ nanosheets.³⁵ The amorphous Al₂O₃ layer might act as a nanoglue to avoid the possible pulverization of active materials. Regarding coating, conductive polymers might be a promising agent, as their large elasticity and resilience offer a perfect protection layer to bind active particles, and thus maximize the structural integrity and stability.⁴⁰

2.2 | Molybdenum dioxide

MoO₂ is an unusual transition metal oxide owing to the semi-metallic nature with low electrical resistivity (8.8 × 10⁻⁵ Ω cm at 300 K)⁴¹ and high thermodynamic stability. Monoclinic MoO₂ crystallizes in a rutile structure, in which the chains of MoO₆ octahedra share opposite edges along the crystallographic c axis.⁴² Each Mo atom is surrounded by six O atoms, while each O is coordinated by three Mo atoms at the corners of an equilateral triangle. The characteristic pairs of Mo centers form distorted octahedron, within it the Mo–Mo distance is 0.251 nm, while the distance between the pairs is 0.311 nm along the rutile c axis.

Similar to MoO₃, sodium storage in MoO₂ involves stepwise insertion and conversion reactions.⁴³ Firstly, Na⁺ ions insert into MoO₂ at a potential about 0.79 V forming NaₓMoO₂ with the formation of solid–electrolyte interphase. Upon further sodiation, NaₓMoO₂ converts into metallic Mo and Na₂O at the potential of 0.33 V. Despite high conductivity and theoretical specific capacity (838 mAh g⁻¹), MoO₂ has been plagued by large volume expansion of about 325%, resulting in breaking of conductive paths upon Na cycling. To address these problems, nanotechnology has found its critical role in the design and engineering of active MoO₂ materials.⁴⁴,⁴⁵

Construction of sheet structure has demonstrated to be effective as the two-dimensional configuration greatly reduces electron and ion transport length.⁴⁶ Jiang et al. synthesized MoO₂/C nanosheets, in which crystallized
MoO₂ nanoparticles at an average particle size of ~10 nm were embedded in the amorphous carbon matrix. The MoO₂/C nanosheets exhibited a discharge capacity of 307 mAh g⁻¹ at 83.8 mA g⁻¹ after 50 cycles. In both cases, the amorphous carbon matrix could prevent the aggregation of MoO₂ nanosheets and maintain the mechanical integrity of the MoO₂ electrode during sodiation/desodiation. Figure 5 shows another case as an example regarding a hierarchical architecture in which TiO₂ nanotubes are coated with ultrafine MoO₂ nanoparticles. The tubular architecture not only relieves the stress associated with the large volume change but also reduces the Na⁺ ion transport path and accelerates the electron transfer. As a result, this hierarchical electrode delivered excellent stability up to 10 000 cycles even at a high rate of 10 A g⁻¹.

For better comparison, we have enclosed some typical electrodes of molybdenum oxide and their sodium storage performance in Table 1. It is worth noting that the accessible capacity for MoO₂ is only limited to ~300 mAh g⁻¹ and would be much less if the voltage window is narrowed to a practical level. Hence, further improvement in the sodium storage activity will be of a high priority.

3 MOLYBDENUM CHALCOGENIDES

Molybdenum chalcogenides show a typical layered structure with enough space for transport and accommodation of guest ions, rendering them an optional choice for battery application. Among these chalcogenides, molybdenum disulfide (MoS₂) and diselenide (MoSe₂) have captured most interest due to their large interlayer space and high electrochemical activity. Table 2 tabulates the electrochemical performances of some molybdenum chalcogenides reported recently.

3.1 Molybdenum disulfide

MoS₂ is stacked by covalently bonded 2D sandwiched S–Mo–S layers with a weak van der Waals force, structurally similar to that of graphite. There are several polymorphs of MoS₂ according to the positions of Mo and S atoms and stacking ways of adjacent layers: the two-layer-stacked hexagonal polymorph 2H-MoS₂, the three-layer-stacked rhombohedral 3R-MoS₂, and the
one-layer-stacked trigonal 1T-MoS2, 2H-MoS2 and 3R-MoS2 are stable, while 1T-MoS2 is unable in air. Therefore, 1T-MoS2 is generally ruled out for electrode application despite its metallic conductivity. The 2H-MoS2 is the most extensively explored material for energy storage but suffers from several crucial challenges upon sodiation such as low conductivity, large volume swelling, and dissolution loss of sulfur species in organic electrolytes.  

Storage of sodium in 2H-MoS2 is also a two-step reaction defined by the terminal voltage as shown in Figure 6. Generally, Na⁺ intercalation occurs at the voltage above 0.4 V, while conversion occurs at lower voltage. The details can be described by equations as follows:

\[ \text{MoS}_2 + x\text{Na}^+ + xe^- \leftrightarrow \text{Na}_x\text{MoS}_2 \] (Equation 1, \( \geq 0.4V \))

\[ \text{Na}_x\text{MoS}_2 + (4-x)\text{Na}^+ + (4-x)e^- \rightarrow \text{Mo} + 2\text{Na}_2\text{S} \] (Equation 2, < 0.4V)

Park et al. explored the structure change of MoS2 upon Na uptake and concluded a Na-concentration-dependent evolution. When \( \sim 1.1 \) Na⁺ ions insert into 1 MoS2 formula (equivalent to discharge potential of 0.4 V), the interlayer along with the c-axis of MoS2 expands with an evident structural change. By in situ technique, Wang et al. could explore the structural evolution of MoS2 upon sodium intercalation in detail. With the insertion of Na⁺ ion, the evolution of 2H-MoS2 is 2H-MoS2 → 2H-Na0.5MoS2 → 1T-Na0.5MoS2 → 1T-NaMoS2 → Mo + NaS. It was shown that \( x = 1.5 \) in NaₓMoS₂ is a critical point, where microstrains cause the distortion of MoS₂ and loss of structural reversibility. Compared to bulk MoS2 with low capacity, the nanostructured MoS2 usually exhibits a higher activity toward sodium storage. Nonetheless, the MoS2 nanostructures still experience a significant capacity decay, due most possibly to the dissolution of polysulfide species upon Na cycling. To mitigate this issue, several strategies such as strong coupling and surface coating have been proposed. Nano-carbon coupling not only offers a conductive percolation but also strongly holds polysulfide species by chemical bonds. Wang et al. demonstrated vertically aligned MoS2 nanosheets grown onto graphene nanosheets as efficient Na anode. In this hybrid, the intimate binding between MoS2 and graphene avoided structural deterioration of MoS2 and minimized the dissolution of polysulfides. As such, the coupled hybrid exhibited high utilization of active materials and stable cycling, affording a capacity of 509 mAh g⁻¹ at 1 A g⁻¹. Another case reported a MoS2/Ni3S2@MoS2 coupled heterostructure with enhanced sodium storage. Surface coating is also effective to hold the structural integrity and to maximize the cycling stability of electrodes. Ryu et al. coated MoS2 nanofiber with a TiO2 nanolayer using atomic layer deposition and achieved drastic improvement in the cycling stability cycle. Similarly, Ahmed et al. deposited an ultrathin (~1 nm) amorphous HfO2 layer onto MoS2 nanosheets. The coated MoS2 remained intact crystallinity upon Na cycling, retaining 91% of its initial capacity in contrast to 62% of the bare MoS2.

To mitigate kinetic issues, on the one hand, designing ultrathin or even single-layered MoS2 nanosheets has

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**TABLE 1**  
Electrochemical performances of Mo-based oxides

| Sample      | Capacity/rate (mAh g⁻¹/A g⁻¹) | Retention/retention number | Ref. |
|-------------|-------------------------------|---------------------------|------|
| a-MoO₃      | 100/1.117                     | 10.5%/500                 | 20   |
| MoO₃₋ₓ      | 180/1                         | 25.4%/100                 | 37   |
| P-MoO₃₋ₓ    | 147/5                         | 22.9%/1500                | 31   |
| MoO₂/rGO    | 330/1.117                     | 59.5%/100                 | 40   |
| MoO₂        | 80/10                         | 21.0%/50                  | 44   |
| MoO₂/C      | 52.5/4.19                     | 36.6%/50                  | 48   |
| TiO₂@MoO₂/C | 76/20                         | 25.1%/500                 | 49   |

*Based on the theoretical capacity.

**TABLE 2**  
Electrochemical performances of Mo-based dichalcogenides

| Sample       | Capacity/rate (mAh g⁻¹/A g⁻¹) | Retention/retention number | Ref. |
|--------------|-------------------------------|---------------------------|------|
| MoS₂         | 175/10                        | 58.2%/1500                | 52   |
| MoS₂/rGO     | 352/0.64                      | 37.9%/300                 | 53   |
| MoS₂/C       | 390/1.34                      | 59.7%/300                 | 54   |
| MoS₂/rGO     | 234/10                        | 48.1%/600                 | 55   |
| MoS₂/carbon  | 187/20                        | 70.9%/200                 | 56   |
| MoS₂/carbon  | 75/50                         | 72.2%/100                 | 57   |
| MoS₂/HFOr   | 347/1                         | 95.1%/50                  | 58   |
| MoS₂/G      | 420/2                         | 76.0%/250                 | 59   |
| MoS₂/NiS₂@MoS₂ | 283/5                 | 72.1%/100                | 60   |
| MoSe₂       | 345/1.5                       | 102.6%/50                 | 61   |
| MoSe₂/C     | 410/0.5                       | 91.0%/2000                | 62   |
| MoSe₂/N-C   | 339/5                         | 114.0%/120                | 63   |
| C-MoSe₂/rGO | 228/4                         | 100.7%/350                | 64   |
| MoSe₂/CNT   | 255/5                         | 70.1%/250                 | 65   |
| MoSe₂/carbon| 216/1.5                       | 89.6%/1000                | 66   |
| VG/MoSe₂/N-C| 113/10                        | 89.8%/500                 | 67   |

*Based on the theoretical capacity.
been proven to be effective. MoS$_2$ single layer can be fabricated via either top-down approach, such as exfoliation,\textsuperscript{58} or bottom-upon method, such as chemical vapor deposition.\textsuperscript{79} As single-layered MoS$_2$ nanosheets approached atomic thickness (~0.4 nm), transport would no longer be a problem, provided that the electronic percolation network is well established.\textsuperscript{80} As a result, the single-layered MoS$_2$ retained a capacity of 484 mAh g$^{-1}$ over 100 cycles at a rate of 1 A g$^{-1}$. To further boost the insertion kinetics, on the other hand, it is feasible to expand the interlayer spacing of MoS$_2$ where the movement of Na$^+$ ions could be facilitated.\textsuperscript{57} Density functional theory simulations showed that the interlayer expansion could reduce the ion intercalation energy and the ion diffusion energy barrier.\textsuperscript{81} To this end, Shi et al. engineered hierarchical nanotubes consisting of carbon-sandwiched monolayers of MoS$_2$, in which the (002) interlayer spacing of MoS$_2$ increase to 0.98 nm (0.615 nm for bulk MoS$_2$).\textsuperscript{82} Benefiting from larger spacing, the nanotube electrode afforded a capacity of 620 mAh g$^{-1}$ with an initial Coulombic efficiency of 84% at 200 mA g$^{-1}$. In addition, it sustained 200 cycles without evident capacity decay, demonstrating its great potentiality in sodium storage (Figure 7). The interlayer expansion strategy may pave a way for exploring high-performance electrode materials for hosting Na$^+$ ions and other large ions.\textsuperscript{56}

**Figure 6**  (A) The high-angle-annular dark-field (HAADF) image reveals the transition of MoS$_2$ phases, (B,C) the annular bright-field (ABF) image of MoS$_2$ with a cutoff of (B) 0.8 V and (C) 0.2 V, and (D) Schematic of the sodium storage mechanism of 2H-MoS$_2$. Reproduced with permission.\textsuperscript{72} Copyright 2014, American Chemistry Society
If the lower cut-off voltage is set at ~0.4 V, the reaction would only involve the intercalation/deintercalation of Na\textsuperscript{+} ions. This is of considerable appeal, as intercalation reaction usually features high reversibility and sustainability. Compared to conversion reaction, neither structural disruption nor the dissolution of polysulfides occurs, and therefore, the structural integrity and cycling performance would be much improved.\textsuperscript{73} Hu et al. have demonstrated that graphene-like Mo\textsubscript{S}_\textsubscript{2} nanoflowers with expanded interlayers of 0.67 nm could achieve a reasonable capacity of 350 mAh g\textsuperscript{−1} with considerable stability over 200 cycles in the range of 0.4–3 V.\textsuperscript{83} This higher capacity might be attributed to the increased interlayer distance. Alternatively, Li et al. further increased the

**FIGURE 7** (A) Schematic illustration of synthesis of Mo\textsubscript{S}_\textsubscript{2}:C superstructure nanosheets, (B) HRTEM images of the as-prepared Mo\textsubscript{S}_\textsubscript{2}:C nanotubes. Inset of (B): profile plot of the calibration for measuring the spacings, (C) HRTEM images of the Mo\textsubscript{S}_\textsubscript{2}:C superstructure nanotubes after post thermal treatment, (D) Profile plot of the calibration for measuring the spacings in panel (C), (E) CV curves of the annealed Mo\textsubscript{S}_\textsubscript{2}:C nanotube electrode at a scan rate of 0.1 mV s\textsuperscript{−1} over the voltage range of 0.0–3.0 V, (F) cycling performance of the annealed Mo\textsubscript{S}_\textsubscript{2}:C nanotube electrode. Reproduced with permission.\textsuperscript{82} Copyright 2016, Elsevier
interlayer distance of MoS$_2$ to 1.45 nm by insertion of poly(ethylene oxide), but only achieved a moderate capacity of 210 mAh g$^{-1}$.52

3.2 | Molybdenum diselenide

Sharing a similar structure to MoS$_2$, layered MoSe$_2$ is composed of individual layers of MoSe$_2$ held together by weak van der Waals force.61,84 MoSe$_2$ is interesting for sodium storage due to its large interlayer spacing (~0.65 nm), small bandgap (~1.1 eV), and acceptable capacity (~422 mAh g$^{-1}$).51 MoSe$_2$ also exhibits a typical stepwise process as MoS$_2$ does. However, the exact mechanism and the reversibility of Se are still under debate.

Wang et al. proposed that the discharged product, Na$_3$Se, would react with metallic Mo to generate MoSe$_2$ during the subsequent desodiation process, although this process is poorly reversible.62 In contrast, by ex situ TEM technique, Xie et al. believed that Se was one of the charge products rather than MoSe$_2$.85 Hence, the Na reaction of MoSe$_2$ deserves further efforts and in situ techniques may play a decisive role in clarifying the process.64

Naturally, MoSe$_2$ suffers from poor conductivity and stability associated with large volume swelling and the “shuttle effect” of polyselenides. Again, nanocomposite offers an opportunity to neatly address these issues.63,65,86,87 MoSe$_2$ nanosheet grown on N, P-co-doped rGO (MoSe$_2$/N, P-rGO) is a case in point.51 A large contact between MoSe$_2$ and rGO enhances the charge transportation and prevented the particles from aggregation, benefiting both the rate and cycling capability. Moreover, both N and P atoms at the edge of graphene were capable of tightly binding polyselenide species, thus minimizing the dissolution loss of active Se. Also, a full cell assembled this anode and Na$_3$V$_2$(PO$_4$)$_3$/C cathode afforded specific energy of 113.6 Wh kg$^{-1}$ at 0.1 kW kg$^{-1}$ (calculate by the mass of both electrode materials). Similarly, Xie et al. had sandwiched MoSe$_2$ nanosheets between vertical graphene and N-doped C.88 The resulting array electrode sustained a capacity of 398 mAh g$^{-1}$ at 1 A g$^{-1}$ after 1000 cycles.

Although the batteries based on liquid electrolytes have been extensively examined, solid-state batteries feature higher energy density and safety.67,89 In this regard, Chen et al. designed molybdenum sulfide selenide (MoSSe) nanoribbons for solid-state sodium batteries.90 When tested in a Na$_3$PS$_4$ electrolyte, MoSSe nanoribbons exhibited a high charge capacity of 523.1 mAh g$^{-1}$ and Coulombic efficiency of 73.6% in the first cycle. In comparison to MoS$_2$, MoSSe exhibited enhanced sodium storage kinetics, as a result of lower energy for Na adsorption on MoSSe than on MoS$_2$.

4 | CARBIDES OF MOLYBDENUM

Although molybdenum chalcogenides show a high reactivity toward sodium storage, their poor conductivity presents a formidable challenge. By contrast, the molybdenum carbides having a platinum-like electronic structure feature a metallic conductivity, which is beneficial for high-rate sodium storage.91 Several types of molybdenum carbides are explored, including α-MoC$_1$–$x$, β-MoC$_3$, η-MoC, and γ-MoC. Among them, β-MoC$_3$, η-MoC, and γ-MoC share a similar hexagonal structure with varied stacking sequences,92 while cubic α-MoC$_1$–$x$ has an isomorphic structure with NaCl.93

Mo$_2$C is well known in hydrogen evolution reaction, but its application in sodium storage has yet been fully investigated until recently. First-principles calculations revealed a low energy barrier for sodium storage in monolayer Mo$_2$C and a final chemical stoichiometry of Na$_{10}$Mo$_2$C, indicating an intercalation mechanism.92 The theoretical capacity of monolayer Mo$_2$C is 132 mAh g$^{-1}$, and the average open-circuit voltage is 0.166 eV. These results have then been experimentally confirmed.94,95 Mo$_2$C might not be appealing for high-energy applications owing to its relatively small capacity but can be used for high-power utilization, particularly in a hybrid system. For example, Mo$_2$C/N-C demonstrated a stable capacity of 204 mAh g$^{-1}$ upon 200 cycles at 100 mA g$^{-1}$.95

MoC seems more interesting than Mo$_2$C as it has a higher capacity of 248 mAh g$^{-1}$ upon the formation of NaMoC. Nonetheless, MoC exhibits conduction and electrochemical activity inferior to Mo$_2$C. Introduction of Mo–N interaction could mitigate the aforementioned issues.96 To this aim, Qiu et al. fabricated N-doped MoC hollow particles, which maintained a capacity of 410 mAh g$^{-1}$ after 200 cycles at 0.16 A g$^{-1}$.97 Such a high capacity was due to the N dopant that drastically decreases the binding energy of sodium absorption (2.22 eV for MoC$_x$N$_y$ versus 6.01 eV for MoC). As the attainable capacity was greater than the assumed value of NaN MoC, reactions other than insertion might occur.97 This assumption has recently confirmed by Li et al., who intended that the redox couple at around 1.4/1.6 V would be attributed to the conversion between MoC and Mo.87 Due to a high capacity, the material of MoC and associated mechanism deserve further research efforts to be systematically explored.

5 | OTHER MATERIALS OF MOLYBDENUM

In recent years, molybdenum phosphides and nitrides have also been applied in SIBs. MoP crystallizes in a
hexagonal structure and possesses a high electronic conductivity of ~5000 S cm⁻¹, making it a promising candidate for battery applications.⁹⁸ MoP exhibits a conversion-reaction mechanism for sodium, as confirmed by in situ high energy XRD.⁹⁹ This conversion can be described as \( \text{MoP} + 3\text{Na}^+ + 3\text{e}^- \rightarrow \text{Na}_3\text{P} + \text{Mo} \), leading to a high theoretical capacity of 633 mAh g⁻¹. However, MoP is very sensitive to oxygen and moisture, and therefore surface engineering should be implemented to enhance the stability of phosphides.¹⁰⁰⁻¹⁰² As for nitrides, Mai et al have shown high sodium performance of mesoporous \( \text{Mo}_2\text{N}_2 \) and \( \text{MoN} \) nanowires.¹⁰³ Strangely, their sodium storage is a capacitance-dominated surface process rather than a bulk behavior, which makes their capacity quite limited.

### 6 | SUMMARY AND PERSPECTIVES

In this review, we deal with electrochemical sodium storage in the molybdenum-based materials, focusing on oxides, chalcogenides, carbides, and others. We have discussed both theoretical and experimental discoveries concerning their sodium-storage performance such as electrochemical activity, sustainable rechargeability, and rate capability. Electrodes made of molybdenum-based materials suffer from one or more issues including electrode pulverization, sluggish charge transfers kinetics, and low electrochemical activity upon electrochemical sodiation/desodiation process. To properly mitigate these challenges, rational materials design and engineering have been proven to be essential and profitable.

Owing to their discrepancy in structure and composition, molybdenum-based materials show different sodium storage mechanisms. Molybdenum oxides and chalcogenides usually undergo insertion and conversion reactions, accompanied by multiphase conversion. They usually exhibit poor reversibility as the sodium ions that are inserted cannot be fully removed. As for other molybdenum materials, their (de)sodiation often involves a Na⁺-driven conversion process. Therefore, they show better reversibility upon Na cycling.

Despite ongoing advancement, substantial challenges remain, and significant efforts are demanded. Many molybdenum-based materials store Na⁺ ions through a conversion reaction, which is accompanied by large volume expansion and drastic structure variation. How to mitigate the volume change of electrodes is a tough topic that is acknowledged throughout the world. In this regard, atomic layer deposition and molecular layer deposition offer tremendous opportunities to stabilize the electrodes under harsh conditions.¹⁰⁴⁻¹⁰⁶ Also, a comprehensive understanding of the electronic and structural evolution of molybdenum materials upon sodium storage remains essential. Such an understanding could help to improve the engineering of electrode materials and the design of battery devices.¹⁰⁷,¹⁰⁸ Acquisition of such information strongly depends on the advancement of in situ or operando analytical techniques and theoretical simulation of the reaction process.¹⁰⁹⁻¹¹¹

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### CONFLICT OF INTEREST

The authors declare no conflict of interest.

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