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MOF-Architected Transition Metal Chalcogenides as electrodes for Next-Generation Alkali-Ion Batteries

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Abstract- : Metal-organic framework (MOF)-architected d-block metal chalcogenides have attracted significant attention in recent times as assuring electrode materials for advanced alkali-ion batteries on account of their porous nature, high surface area, and enhanced electrochemical reactivity. Progress achieved in the fabrication of MOF-architected sulfides, selenides, tellurides, and hybridized compounds has demonstrated remarkable electrochemical performance in lithium-ion (LIBs), sodium-ion (SIBs), and potassium-ion batteries (KIBs). The unique structures architected from MOFs lead to enhanced ion transport kinetics and cyclability. This review highlights recent developments in MOF-architected TMCs as potent electrode materials for alkali metal-ion secondary batteries.

Keywords:- Metal–organic frameworks (MOFs), Transition metal chalcogenides, Alkali-metal ion batteries, Electrode materials, Energy storage

1. INTRODUCTION

Amidst a limitless surge in the global energy demand, as a consequence of technological and economic advances, the lashing geopolitical tension in the Middle East, engaging vital energy anchors like Iran, Israel, and the United States, has once again cast the spark on the intense delicacy of the global fossil fuel economy. War-driven energy crises, including uncontrolled hikes in crude oil prices, global power-deficiency, and energy volatility, have transformed the sense of global energy security from a sustained environmental goal to an instant nationwide security urgency. To attain self-reliance in the energy economy, an immediate transformation in the direction of consistent green energy storage is the need of the hour.

Alkaline ion batteries, among other electrochemical energy storage systems, have been widely studied for their top-notch energy density, extended cyclic performance, and ecofriendly nature.[1] Lithium-ion batteries (LIBs) represent the most common battery types commercially used at present; however, scarcity in lithium availability and increasing prices have led to the development of alternative battery types like Na-ion batteries (SIBs) along with K-ion batteries (KIBs).[2,3]

Metal-organic frameworks (MOFs) stand as a genre of absorptive crystal-like materials constituted of agglomerated metal ions linked to organic ligands and are promising starting materials for electrochemistry due to their customizable porosity and morphology.[4] Chemical or thermal processing of MOFs results in carbonized nanoparticles with well-dispersed active sites.[5] Consequently, MOF-based materials have demonstrated remarkable performance in the application of electrodes in alkali-ion batteries

Transition metal chalcogenides (TMCs), consisting of sulfides, selenides, and tellurides, have attracted a great deal of interest compared to other MOF-based materials because of their outstanding electrochemical kinetics, abundant redox activities, and high theoretical capacities.[6,7] Moreover, the incorporation of carbon architectures, heterostructures, and multi-metallic architectures can greatly enhance conductivity while minimizing structural instability during the cycling process.[8]

This review article provides a detailed insight into the current developments of MOF-based transition metal chalcogenides (TMCs) and their composites for application as electrode material in metal cation (Li^+, Na^+, K^+) secondary batteries.

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II. MOF-ARCHITECTED TRANSITION METAL CHALCOGENIDES FOR NEXT-GEN LI-ION BATTERIES

Li-ion batteries stated as LIBs find application in consumer electronics and automotive industry because of their excellent properties including high energy density, longer cycling life, and superior electrochemical behavior.[1] The rise in demand for better performing devices has resulted in the discovery of new electrode materials, which not only enhance their energy storage capacity but also improve the cycling stability. MOF-based transition metal chalcogenide (TMC) materials have been introduced as alternative candidates owing to their porous nature, larger electrochemically active surface area, and enhanced conductivity.[5] Electrochemical performances of MOF-based TMC electrodes for LIBs are summarized in Table .

Table 1. Electrochemical performance of MOF-architected transition metal chalcogenides and its composites as lithium-ion battery electrode materials.

Material	**MOF**	**Gravimetric Capacity (mAh/g)**	**Current Density (A/g)**	**Cycle count**	**SoH Retention (%)**	**Ref.**
NiS Nanoplates	Ni-MOF	213	10	100	-	[9]
$CoTe_2$@N-C	ZIF-67	157.2	0.1	100	90.95 %	[10]
Co-Zn-S@N-S-C-CNT	Co-Zn-ZIF-67	941	0.1	250	-	[11]
FeS_2@S-C NR-NS	MIL-88-Fe	1336.5	0.1	200	108.6 %	[12]
Ni_3Se_4/$NiSe_2$@C	Ni-MOF	362.3	4	2000	-	[13]
Fe_7S_8@C-S	MIL-88-Fe	1503	100	0.05	95%	[14]
CoSe@PCP	ZIF-67	675/708.2	0.2/1	100/500	-	[15]
α-MnSe/CNF	Mn-BTC	845.54/544.6	0.1/1	100/500	-	[16]
$CoNiSe_2$/C	Co Ni-MOF	549.9	1	1000	-	[17]
$AlCuSe_2$@C	Al-Cu MOF	351	4	2000	71%	[18]
$Cu_{1.8}Se$@C	Cu-BTC	320.3	1	1000	-	[19]
Co-Zn-Se@C	Co/Zn-MOF-74	949	1	500	-	[20]
ZnCoS@Co_9S_8/NC	Co Zn-ZIF	1813/1095	0.5/2	500/400	98%	[21]
Co_3S_4@C @MoS_2	ZIF-67	672.6	0.2	200	89.8 %	[22]
Ni-Co-Se@C	Ni-Co-BTC	2061	0.5	300	325%	[23]
$Co_{1-x}S$/C	Co-BTC	791	0.2	100	88%	[24]
ZCS@NC/CNTs	ZIF-8/67	873/768	0.5/1	500/1000	-	[25]
$MoSe_2$@NPC	ZIF-8	527	1	500	82%	[26]
$Co_{1-x}S$/NCS	Co-MOF	796.3	0.2	100	-	[27]

The $CoNiSe_2$/C nanododecahedrons were obtained from Han et al.[17] through the selenization of Co-Ni-MOF within an Ar atmosphere. The optimal $CoNiSe_2$/C-700 electrode exhibited a preliminary discharge capacity of 2125.5 mAh/g under a specific current of 0.1 A/g, beside an additional capacity of

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549.9 mAh/g, achieved past 1000 cycle count of testing under 1.0 A/g current density. The enhanced electrochemical characteristics were due to the synergistic effect of the porous nanododecahedral structure, carbonaceous matrix, and bimetallic selenide structure. Figure 1 demonstrates the fabrication process as well as the electrolytic performance of the $CoNiSe_2$/C compound.

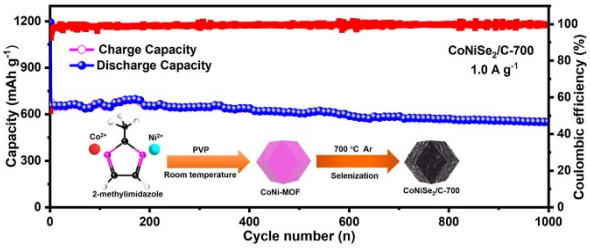

Figure 1. Schematic demonstration of the fabrication with electrochemical behaviour of MOF-architected $CoNiSe_2$/C nanododecahedral electrodes for lithium-ion batteries. Reproduced from Ref. 17; Copyright 2024, American Chemical Society.

Aslam et al.[21] employed a bimetallic metal-organic framework precursor for the fabrication of mono-porous void backing fundamental-shell ZnCoS@Co_9S_8/NC through sulfurization by kinetics control. These structures enhanced the performance of the material due to their yolk shell structure and nitrogen-nobbled carbon matrix leading high specific capacities of 2141 mAh/g at 0.1 A/g along with 1095 mAh/g following 400 cycle count at 2 A/g. According to Li et al.[14], the sulfurized sulfur-activated carbon bound Fe_7S_8@C-S nanorods were prepared from MIL-88 by using in-situ sulfurization with thiourea. The core-shell structure improved Li ion diffusion kinetics and limited volume change while cycling. The Fe_7S_8@C-S electrode possessed a capacity of 1000 mAh/g after 100 cycles at 0.1 A/g and 95% capacity back-up past 1000 cycles at 2 A/g

III. MOF-ARCHITECTED TRANSITION METAL CHALCOGENIDES FOR ADVANCED SODIUM-ION BATTERIES

Sodium-ion secondary batteries stated as SIBs have become a practical alternative to Li-ion batteries owing to the high availability, lower cost, and widespread geographical sources of sodium.[2] Moreover, SIBs offer great promise for energy storage applications on a commercial scale. Nonetheless, the bigger ionic radius of Na^+ results in volumetric changes and slow kinetics in the system. To address these limitations, transition metal chalcogenides (TMCs) architected from MOFs have drawn much attention on account of their porosity, controllable morphologies, and enhanced conductivity.[5] Table 2 provides an analysis of the electrochemical properties of TMCs used for SIBs

Table 2. Electrochemical behaviour of MOF-architected transition metal chalcogenides as sodium-ion battery electrodes.

Material	MOF	Gravimetric capacity (mAh/g)	Current Density (A/g)	Cycle count	Retention (%)	Ref.
$Cu_{2-x}Se$@C	Cu-BTC	216	2	1800	97.3 %	[28]
Fe_3Se_4/NC	MIL-88-A	290.8	8	1000	94.9 %	[29]
$CoTe_2$@CNTs	ZIF-67	112.7	2	500	-	[30]
N-ZnSe@rGO	ZIF-8	285	0.3	500	-	[31]
Fe_2CoSe_4@NC	FeCo-MOF	352	1	2100	-	[32]
$CoNiSe_2$/C	Co-Ni-BTC	421.1	2	2000	-	[33]
Fe_7S_8@C	MIL-88-Fe	341.3	1.25	150	-	[34]
$Cu_4Mo_6Se_8$/C	Cu-Mo BMOF	474	2	2400	107.7 %	[35]
CuS@N-C	Cu-MOF	300.2	5	1200	95.8 %	[36]
NSGA/$CoTe_2$@NPC	Co-MOF	518.2/445.6	1/2	1000/2000	>99%	[37]
(FeCoNiMnCu)Se@C	(FeCoNiMnCu)-MOF	455	1	2300	-	[38]
$NiSe_x$@C/CNT	Ni-BTC	387.1	0.1	2000	-	[39]
$CoSe_2$/ZnSe@NC	ZIF-67/ZIF-8	264.8	5	800	91.7 %	[40]
Co/(NiCo)Se_2	ZIF-67	497	0.2	80	92%	[41]

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$CoSe_2$/ZnSe @C	CoZn-BIM	376	2	2500	92%	[42]
$CoSe_2$@NC	ZIF-67	404	0.2	120	80%	[43]
CoNiZnS/C	Co-Ni-Zn MOF	410.7	2	960	-	[44]
Ni_3S_2@C	Ni-MOF	268.2 /140. 6	0.1/ 1	100/10 00	-	[45]

Wang et al.[35] manufactured MOF-architected $Cu_4Mo_6Se_8$ nanosheets with a carbon conductive framework for efficient sodium storage applications. The nanosheet arrangement increased the rate of Na ion diffusion, thereby improving electrochemical kinetics. CMSe/C exhibited an impressive adjustable capacity of 411 mAh/g at 0.1 A/g and could even maintain a capacity of 365 mAh/g when subjected to a high current of 5 A/g. Furthermore, the battery was capable of delivering 474 mAh/g past 2400 cycle count at a rate of 2 A g^{-1}, demonstrating exceptional solidity.

In-situ synthesis of nitrogen-doped carbon-coated CuS nanoparticles (CuS@N-C) was reported by Liu et al.[36], through carbonization followed by sulfidation of a Cu-MOF precursor. Porous octahedral architecture along with a conducting carbon matrix with nitrogen doping facilitated better charge transfer without causing any volume alteration throughout cycling. The CuS@N-C material exhibited a discharge/charge capacity of 584.2/479.0 mAh/g at 0.1 A/g and withstood a capacity of 300.2 mAh/g past 1200 cycle count at 5 A/g, resulting in 95.8% of capacity retention. Moreover, when cycled at an impressive rate of 10 A/g for 2670 cycles, the capacity even stood at 161.8 mAh g^{-1}.

Li et al. [28] used selenization treatment on the Cu-BTC-based precursors to synthesize MOF-derived $Cu_{2-x}Se$@C composites that contained $Cu_{2-x}Se$ nanoparticles homogeneously dispersed in carbon matrix. When cycling at 0.2 A/g, the resulting $Cu_{2-x}Se$@C electrode delivered initial specific discharge/charge capacities of 473/319 mAh/g and retained an impressive specific capacity of 310 mAh/g with 97.3% capacity retention after 70 cycles. Furthermore, the electrode achieved reversible capacities of 317 and 257 mAh/g at current densities of 0.1 A/g and 10 A/g, respectively. Notably, after 1200 cycles at 2 A/g, the electrode had 88.1% capacity remaining. The long-term sodium-ion storage performance of $Cu_{2-x}Se$@C electrode is shown in Figure 2.

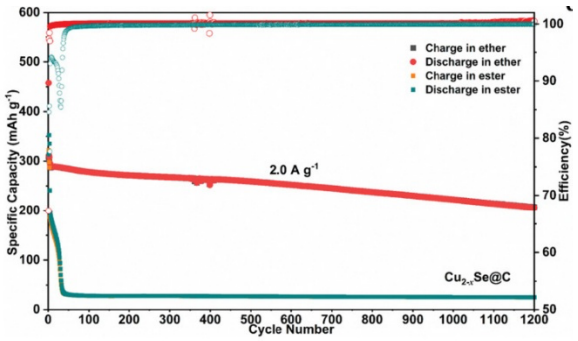

Figure 2. Cycling performance of MOF-derived $Cu_{2-x}Se$@C electrode tested at 2 A/g current density for sodium-ion storage. Reproduced from Ref. 28; under creative common licence.

Xiao et al.[42] successfully prepared ultra-nano nitrogen-nobbled carbon-layered $CoSe_2$/ZnSe nanospheres through a bimetallic MOF precursor via a selenization process. The utilization of bimetallic selenides along with conductive carbon wrapping significantly enhanced the storage capability of Na^+ ions. These fabricated electrodes demonstrated excellent steadiness of 346 mAh/g after 2500 cycle count under 2 A/g.

IV. MOF-ARCHITECTED TRANSITION METAL CHALCOGENIDES FOR POTASSIUM-ION BATTERIES

KIBs can be considered one of the feasible alternatives for energy system subject to the abundance of potassium ions along with the low standard reduction potential (K/K^+), similar to that of lithium.[46] In addition, the relatively lower Lewis acidity of K^+ results in a faster ion movement in electrolytes. In contrast, the large ionic size of potassium ions gives escalation to substantial structural strain and sluggish kinetic behavior during cycling. In order to overcome these challenges, TMCs based on MOFs with porous nanostructures and conductive carbon-based architectures have been studied in detail as advanced anode materials.[5] Table 3 illustrates the electrochemical behavior of certain MOF-based TMCs used for KIBs

Table 3. Electrochemical behaviour of MOF-architected transition metal chalcogenides as potassium-ion battery electrode materials.

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Material	MOF	Gravimetric Capacity (mAh/g)	Current Density (A/g)	Cycle count	Retention	Ref.
Co_9S_8@NPC	ZIF-67	315	0.1	200	67.5 %	47
NSGA/$CoTe_2$@NPC	Co-MOF	410/301.1	0.1/2	200/1500	>90 %	37
CoSe2@NC/HMCS	ZIF-67	442	0.1	120	-	48
$MoSe_2$-$CoSe_2$@NC	ZIF-67	327	0.5	150	-	49
CoS_2/NC@VS_4	ZIF-67	291.54	1	430	-	50
ZnSe@PCNF	ZIF-8	270	0.5	1000	-	51
CoPSe/NC	Dia-Co(Melm)$_2$	317/203	0.1/5	100/2000	89.2 %	52
NPCP@$MoSe_2$	ZIF-8	325	0.1	80	82%	53
ZnTe@C/$Ti_3C_2T_x$	ZIF-8	408	0.1	100	82%	54
ZnSe/$CoSe_2$@PDA	ZIF-8@ZIF-67	182	1	250	-	55
$CoSe_2$@NC/MX	ZIF-67	358	0.5	100	-	56
CoSSe@C/G	ZIF-67	208.1	0.2	100	-	57
$CoTe_2$/ZnTe	ZIF-67/ZIF-8	175.3	3	4000	89.4 %	58
CoS_2	ZIF-67	537.3	0.1	50	-	59

Jiang et al.[53] fabricated the NPCP@$MoSe_2$, which is made up of nitrogen-nobbled porous carbon polyhedron alongside $MoSe_2$ nanosheets coating and decorated on ZIF-structured porous carbon frameworks. Upon 80 cycle count at 0.1 A/g, the material exhibited a reversible capacity of 325 mAh g^{-1}, while it maintained a capacity of 128 mAh g^{-1} upon 800 cycles at 0.5 A/g. The enhancement in potassium storage capabilities can be attributed to wider interlayer distances, porous carbon frameworks, and shorter potassium ion diffusion pathways.

Oh et al.[49] fabricated the $MoSe_2$@$CoSe_2$/N-doped carbon nanorods using selenization of MoO_3 nanorods decorated with ZIF-67 coating. At the current density of 0.5 A/g, the electrode delivered a reversible capacity of 327 mAh/g for 150 cycle count and a rate capability of 190 mAh g^{-1} at current density of 2.0 A g^{-1}.

Double-shelled $Co_{0.85}Se_{1-x}S_x$@C/graphene hollow polyhedrons were fabricated by Wang et al.[57] using a cobalt-focused metal organic framework precursor. The electrode demonstrated a capacity of 208.1 mAh/g following 100 cycle count under a current density of 0.2 A g^{-1}, while maintaining a high capacitive retention, indicating enhanced structural stability and reaction kinetics during potassiation/depotassiation processes.

The fabrication of $CoTe_2$/ZnTe heterostructures by tellurization of CoZn-MOF precursors based on $Ti_3C_2T_x$ MXene nanosheets was performed by Pan et al. [58] through 500 cycles under the condition of 0.2 A/g, the porous composite heterostructures yielded high reversible capacities of 339.1 mAh/g; meanwhile, by performing 4000 cycles under the condition of 3 A/g, the material still exhibited a high capacity of 175.3 mAh/g, which represents 89.4% of the initial capacity. Furthermore, the electrode material showed superior structural stability and K^+ diffusion kinetics with capacities of 365.3 and 134.3 mAh/g under the conditions of 0.1 and 10 A/g, respectively. Figure 3 demonstrates the morphology and electrochemical performance of the electrode material of $CoTe_2$/ZnTe.

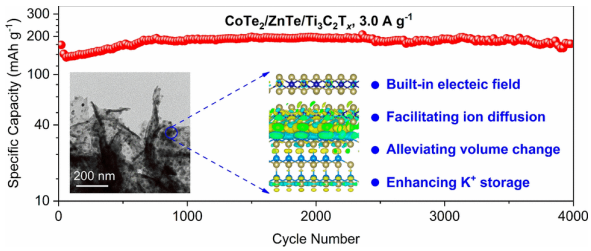

Figure 3. Schematic representation of the $CoTe_2$/ZnTe heterostructure on $Ti_3C_2T_x$ MXene and electrochemical potassium-ion storage performance. Reproduced from Ref. 58; under creative common licence.

V. CONCLUSIONS

In conclusion, MOF-architected transition metal chalcogenides have shown great assurance as anode materials for metal cation like lithium, sodium, and potassium-ion batteries, owing to their

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tunable compositions, hierarchical structures, and excellent electron transfer abilities. The integration of porous structures, carbon-based networks, and multi-metal heterostructures enhances the mobility and stability of the ions during electrochemical reactions. Recent studies have made impressive strides in achieving high specific capacities and long-term cycling stabilities for various alkali-ion-based batteries. Nevertheless, the practical application of such material requires scalable synthetic strategies, initial coulombic efficiencies, and long-term structural stability. Future research can hasten the progress towards the optimization of MOF-architected chalcogenide electrodes

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