Manipulating metal–sulfur interactions for achieving high-performance S cathodes for room temperature Li/Na–sulfur batteries

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Abstract
Rechargeable lithium/sodium–sulfur batteries working at room temperature (RT-Li/S, RT-Na/S) appear to be a promising energy storage system in terms of high theoretical energy density, low cost, and abundant resources in nature. They are, thus, considered as highly attractive candidates for future application in energy storage devices. Nevertheless, the solubility of sulfur species, sluggish kinetics of lithium/sodium sulfide compounds, and high reactivity of metallic anodes render these cells unstable. As a consequence, metal–sulfur batteries present low reversible capacity and quick capacity loss, which hinder their practical application. Investigations to address these issues regarding S cathodes are critical to the increase of their performance and our fundamental understanding of RT-Li/S and RT-Na/S battery systems. Metal–sulfur interactions, recently, have attracted considerable attention, and there have been new insights on pathways to high-performance RT-Li/Na sulfur batteries, due to the following factors: (1) deliberate construction of metal–sulfur interactions can enable a leap in capacity; (2) metal–sulfur interactions can confine S species, as well as sodium sulfide compounds, to stop shuttle effects; (3) traces of metal species can help to encapsulate a high loading mass of sulfur with high-cost efficiency; and (4) metal components make electrodes more conductive. In this review, we highlight the latest progress in sulfide immobilization via constructing metal bonding between various metals and S cathodes. Also, we summarize the storage mechanisms of Li/Na as well as the metal–sulfur interaction mechanisms. Furthermore, the current challenges and future remedies in terms of intact confinement and optimization of the electrochemical performance of RT-Li/Na sulfur systems are discussed in this review.
1 | INTRODUCTION

Sulfur, as a potential alternative material for conventional cathodes, has been intensively pursued for use in rechargeable systems over a long time because of its high theoretical capacity (1672 mAh g⁻¹), environmental friendliness, and cost-effectiveness.¹⁻⁶ Thus, metal–sulfur batteries have received increasing attention. Rechargeable lithium/sodium–sulfur cells, working at room temperature (RT-Li/S and RT-Na/S cells), as the most important parts of metal batteries, have been extensively investigated and partially commercialized. The advantages of advanced RT-Li/Na sulfur systems offer the high theoretical energy density of Li/Na metals, their similar chemical properties, the nature abundance of elements, capability for low-temperature operation, and environmental friendliness.⁷⁻¹⁷ These advantages of RT-Li/S and RT-Na/S cells make them two of the most promising candidates for energy storage devices in the near future.¹⁸⁻²⁰ Challenges, however, still remained for achieving efficient conversion reactions of sulfur species, which limit the application of RT-Li/Na sulfur batteries with regard to low reversible capacity as well as rapid capacity decay.²¹,²² The insulating nature and poor reactivity of sulfur-related compounds, however, result in poor sulfur utilization and low accessible capacity.²³⁻²⁸ In addition, the dissolution of lithium polysulfides (LiPSs)/sodium polysulfides (NaPSs) is the origin of the “shuttle effect,” where the dissolved LiPSs/NaPSs migrate between the anode and cathode in the organic electrolyte during cycling.²⁹ As a result, the shuttle effect causes loss of active substances, severe capacity decay, and poor stability.³⁰⁻³⁴ Considerable efforts have, therefore, been made to solve these problems to improve the performance of Li/Na sulfur batteries through the confinement of S or constructing chemical bonding between S and its host materials. Effective hosts include carbonaceous materials (graphene, carbon nanotubes, carbon fibers), metal oxides, and metal sulfides, as well as metals.³⁵⁻⁴⁵ Recently, there is a new synthesis and morphology control method that can help the future synthesis of sulfur hosts.⁴⁶ Although the utilization of various hosts can enhance the performance of Li/Na sulfur batteries, a giant leap is a prerequisite for meeting the practical targets for practical applications. In recent years, metals have been introduced into a sulfur host to further optimize the performance of Li/S as well as Na/S systems. For example, metal materials, such as single-atom metals and metal clusters, are arousing extensive interest, owing to their maximum atom utilization and high utilization rate of active materials.⁴⁷⁻⁵⁶ Arava’s group first reported that Pt dispersed on graphene can work as an electrocatalyst and accelerate the kinetics of the transformation of polysulfides in Li/S batteries.¹ The specific capacity of the synthesized graphene-supported Pt nanoparticles (Pt/graphene) was increased by 40% compared with the original graphene, and the stability reached more than 100 cycles under 0.2 C, while the Coulombic efficiency was 99.3%. Tang et al.⁵⁷ also applied a nickel fiber/sulfur composite as a cathode in Li/S systems. With 3 wt% addition of nickel, the initial discharge capacity of the constructed system reached 805 mAh g⁻¹. Furthermore, Yao et al.⁵⁸ enhanced the performance of the Li/S system via the fabrication of metal/graphene composites. Chen’s group also recently successfully synthetized a single atom-based material, nickel (Ni) single atoms loaded on nitrogen-doped graphene (Ni@NG), which exhibited a remarkable rate performance as well as a stable cycling life with ultralow capacity attenuation of 0.06% in each cycle.⁵⁹ Similar concepts have also been proven for the RT-Na/S system. For instance, Zhang et al.⁶⁰ designed a novel S host via using metal (M) nanoclusters (Fe, Ni, Cu) supported on hollow carbon nanospheres (S@M-HC) for achieving excellent performance in RT-Na/S cells. It has been demonstrated that S@M-HC can enhance sulfur reactivity by forming M–S bonds. Moreover, it can electrocatalytically reduce polysulfides to Na₂S, thus efficiently inhibiting the dissolution of NaPSs and alleviating the shuttle effect. In addition, they also prepared a novel sulfur matrix with atomic cobalt anchored on the pores of hollow carbon nanospheres (S@Co₃-HC).⁶¹ The initial reversible capacity of the designed S@Co₃-HC was 1081 mAh g⁻¹, and the sulfur utilization rate was 64.7%. It is worth noting that the battery maintained a superior reversible capacity of 508 mAh g⁻¹ after 600 cycles at a current density of 100 mA g⁻¹.

In this review, we analyze the effects of metals loaded on carbonaceous host materials as efficient sulfur cathode electrocatalysts on the performance of RT-Li/S and RT-Na/S cells. We aim to summarize the Li/Na storage mechanisms and challenges, and metal–sulfur interaction mechanisms, as well as improvements in electrochemical performances (Figure 1). Then, the remaining challenges and perspectives on developing RT-Li/Na sulfur batteries are discussed.
LI/NA STORAGE MECHANISM AND CHALLENGES

As is well known, the RT-Na/S and RT-Li/S batteries share similar mechanisms. The working mechanism of RT-Na/S cells during the discharging process is shown as follows:

Anode: \[ \text{Na} \leftrightarrow \text{Na}^+ + e^- \]  

Cathode: \[ n\text{S} + 2\text{Na}^+ + 2e^- \rightarrow \text{Na}_2\text{S}_n(4 \leq n \leq 8) \]  

Overall: \[ \text{S}_8 + 2\text{Na}^+ + 2e^- \leftrightarrow \text{Na}_2\text{S}_8 \]  

\[ \text{Na}_2\text{S}_8 + 2\text{Na}^+ + 2e^- \leftrightarrow 2\text{Na}_2\text{S}_4 \]  

\[ \text{Na}_2\text{S}_4 + 2/3\text{Na}^+ + 2/3e^- \leftrightarrow 4/3\text{Na}_2\text{S}_3 \]  

\[ \text{Na}_2\text{S}_4 + 2\text{Na}^+ + 2e^- \leftrightarrow 2\text{Na}_2\text{S}_2 \]  

\[ \text{Na}_2\text{S}_2 + 2\text{Na}^+ + 2e^- \leftrightarrow 2\text{Na}_2\text{S} \]  

To sum up, the RT-Na/S undergoes a solid (S8)–liquid (Na2S8, Na2S6, Na2S4)–solid (Na2S2, Na2S) transition.62 The RT-Li/S and RT-Na/S systems still face huge challenges, however.29,63 First, on the sulfur cathode, the poor conductivity of sulfur as well as its final discharge product, Li2S/Na2S, usually leads to a low utilization rate of the active material, which results in unsatisfactory discharge capacity of the batteries. Second, the long-chain LiPSs/NaPSs (Li2Sx/Na2Sx, 4 \leq x \leq 8) can be dissolved in the organic electrolyte, leading to loss of active materials, and inevitably capacity decay. In particular, the higher solubility of NaPSs in the electrolyte than that of the LiPSs in RT-Li/S cells causes a more serious shuttle effect in RT-Na/S cells. In addition, the conversion from sulfur to the final products Li2S/Na2S causes large volume changes with a volume expansion of 260% in RT-Na/S batteries and 80% in RT-Li/S cells. The large volume expansion will cause rapid capacity decay and serious safety issues. Furthermore, on the anode side, the growth of lithium/sodium dendrites, also poses challenges for developing both Li/S cells and Na/S cells. In addition, the unstable solid electrolyte interphase, which arises from the side reaction between lithium and electrolyte, always leads to poor Coulombic efficiency and rapid capacity loss. The introduction of metals on the sulfur host has recently attracted increasing attention owing to the excellent catalytic activity of metals. The atomically dispersed metal atoms are reported to form polar interactions with sulfur and LiPSs/NaPSs, which are expected to suppress the shuttle effect so as to enhance the performance of RT-Li/Na sulfur batteries. Furthermore, the catalytic activity of atomic metals can reduce the decomposition energy barrier of LiPSs/NaPSs, thus ensuring fast conversion kinetics between long-chain polysulfides and short-chain sulfides. Generally speaking, various metals have different catalytic effects. Therefore, it is highly significant to introduce new kinds of atomic-scale metals into the polarized sulfur supports, thus efficiently enhancing the electrochemical performance of RT-Li/Na sulfur cells. Moreover, the doping of different metals can play a synergistic effect, and the exploration of the doping ratio is also of great importance. For instance, Chen’s group reported the catalytic performance of nickel single atoms supported on nitrogen-doped graphene (Ni@NG) for the conversion of LiPS, based on the strong chemical affinity between Ni@NG and LiPSs.59 As depicted in Figure 2A, \( \Delta G_x \) \((x=1–5)\) represents different Gibbs free energy values of adjacent LiPSs. In general, the smaller the value of \( \Delta G_x \), the faster the reaction kinetics. Impressively, compared with NG, the lower \( \Delta G_x \) of LiPSs on Ni@NG indicates that the conversion reaction of LiPSs on Ni@NG is faster during cycling. Furthermore, the binding energy also plays a crucial role in immobilizing the LiPSs during the cycling process. Generally speaking, the greater binding energy value between LiPSs and the prepared Ni@NG indicates the efficient confinement of LiPSs and, therefore, confirms the inhibition of the shuttle effect (Figure 2B). To sum up, the uniformly dispersed Ni single atoms speed up the conversion between LiPSs and Li2S due to their outstanding catalytic activity and
simultaneously avoid the shuttle effect due to the formation of strong \( S_x^2-\text{Ni}-\text{N} \) chemical bonds (Figure 2C). A similar mechanism has been found in RT-Na/S systems. According to the ab initio molecular dynamics results, Fe nanoclusters show the biggest dissociative adsorption energy of \(-10.24\ eV\), which indicates that they promote the best reactive activation between Na\(_2\)S\(_4\) and Fe clusters, and this is consistent with the density functional theory (DFT) result (Figure 2D). The theoretical calculations indicate that the introduced electrocatalyst S@Fe-HC can significantly reduce the energy barrier and facilitate the breaking of sodium sulfides from the long-chain polysulfides to form short-chain polysulfides, thus inhibiting the shuttle effect. Also, the S@Fe-HC was suggested to play a crucial role in enhancing the electrical conductivity as well as the reactivity of sulfur through the formation of M-S chemical bonds. In addition, Zhang et al.\(^{61}\) also demonstrated the catalytic and immobilization effects of cobalt single atoms on NaPSs via in situ characterizations. Generally, we use in situ Raman to obtain the phase transition of sulfur and infer the role of metal catalysts during the phase transition. As depicted in Figure 2E,F, the first discharge mechanism of S@Co-HC can be expressed as follows:

\[
S \rightarrow \text{Na}_2\text{S}_x \rightarrow \text{Na}_2\text{S}_4 \rightarrow \text{Na}_2\text{S}_2 \rightarrow \text{Na}_2\text{S}.
\]

Nevertheless, when the battery was recharged to 2.8 V, the peaks of Na\(_2\)S\(_2\) and S disappeared, indicating that only the conversions from Na\(_2\)S to Na\(_2\)S\(_4\) and to Na\(_2\)S\(_x\) were reversible. Also, the sluggish reactivity of Na\(_2\)S, as shown in the results of Raman spectra and XRD patterns, could cause the accumulation of Na\(_2\)S over the long discharge/charge process. Unlike pure carbon support, Na\(_2\)S\(_4\) can spontaneously decompose on Co\(_6\) clusters. When adsorbed on Co\(_6\), the adsorption energy of Na\(_2\)S\(_4\), Na\(_2\)S\(_3\), Na\(_2\)S\(_2\), and Na\(_2\)S were \(-4.33\), \(-4.85\), \(-7.85\), and \(-10.67\ eV\), respectively (Figure 2F). The strong adsorption of Na\(_2\)S demonstrated that the electrocatalytic decomposition of Na\(_2\)S\(_4\) to Na\(_2\)S was kinetically fast in the presence of Co\(_6\) clusters. As a result, Co\(_6\) clusters efficiently alleviated the dissolution of Na\(_2\)S\(_4\) in
electrochemical processes, so as to achieve an excellent electrochemical performance. The DFT results also confirmed the decomposition of $\text{Na}_2\text{S}_4$ through the electrocatalytic capability of Co$_{10}$, which is in accordance with the results of Raman spectroscopy and XRD characterization. The dissolution of polysulfides can be effectively alleviated by surface vulcanization of atomic Co. Therefore, the chemical interaction formed between the metal and sulfur can significantly improve the cycling stability and reversible capacity of Na/S cells, due to the efficient catalytic conversion capability from $\text{Na}_2\text{S}_4$ to $\text{Na}_2\text{S}$. The rapid phase-transfer can inhibit the dissolution of polysulfides, thus impeding the shuttle effect.

3 | METAL–SULFUR INTERACTION MECHANISMS

As discussed above, the introduced metal species on sulfur hosts can effectively work toward the improvement of the performance of RT-Na/S and RT-Li/S from many aspects. Pioneering research on the mechanisms of metal–sulfur interaction has been conducted and reached conclusions on the following features: (1) sulfur immobilization, (2) catalytic capability of metal species, (3) inhibition of the solubility of sulfur/sulfide-related materials, and (4) improved conductivity of sulfur/sulfide-related compounds.

3.1 | Sulfur immobilization

A porous structure with a large specific surface area can store more sulfur. Sulfur/sulfide-related species, however, still show a gradual loss during cycling because of the weak interaction between physical adsorption and the porous structure. Therefore, measures to further immobilize sulfur, are urgently needed. It has been confirmed that achieving the chemical interaction of sulfur species and specific hosts is an efficient way to trap and immobilize sulfur species. In brief, it has been demonstrated that some metals used as catalysts supported on sulfur hosts can ensure superior performance of Li/S or Na/S cells. The transition metal nanoparticles are uniformly dispersed in the porous carbon to absorb S and polysulfides. The strong interaction between the highly conductive transition metal and S can fix and load more S in the porous carbon. For instance, there are strong chemical interactions between transition metals (Ni, Cu, Co, etc.) and sulfur. Zheng et al.\textsuperscript{64} used microporous carbon doped with Cu nanoparticles to capture S/polysulfides through chemical interaction in Li/S batteries. Compared with the physical confinement within pores, the chemical interactions of metal atoms and sulfur species displayed stronger and more efficient adsorption of S/polysulfides.

Li et al.\textsuperscript{65} reported metal–organic framework (MOF)-derived porous carbon polyhedra with Co doping, which were coated with reduced graphene oxide (RGO) for sulfur immobilizing cathodes for Li/S batteries. Here, the Co-doped porous carbon derived from MOFs could not only confine sulfur by physical adsorption on porous carbon, but also further fix sulfur via chemical interaction of Co and sulfur.

3.2 | Metal catalysis

Various metals, which are often used to improve electrochemical reactivity, have high electrocatalytic activity. The metals can accelerate the redox reactions of long-chain polysulfides during the charge/discharge process. For instance, Salem et al.\textsuperscript{1} reported the electrocatalytic effect of the metal catalyst Pt on the redox reactions of LiPSs. It was shown that Pt can promote reversible transformation between $\text{Li}_2\text{S}_2$/$\text{Li}_2\text{S}$ and LiPSs. The Pt nanoparticles can not only adsorb soluble polysulfides during the discharge process, but also convert them to long-chain LiPSs and elemental sulfur during the charge process, avoiding their excessive deposition on the electrode. And the presence of Pt electrocatalyst can enhance the reaction kinetics at high current densities. Furthermore, Chen’s group prepared nitrogen-doped graphene with uniformly embedded single Ni atoms ($\text{Ni}@(\text{NG})$ for the Li/S system.\textsuperscript{59} The distinctive Ni–N$_4$ structure can cause reversible catalysis of the transformation of LiPSs via the formation of $\text{S}_x^{2−}$–Ni–N bonds. Also, the formed $\text{S}_x^{2−}$–Ni–N bonds enable the bonding between Li$^+$ and negative N atoms by electrostatic adsorption, thus trapping LiPSs. In addition, the single Ni atoms on nitrogen-doped graphene can effectively promote the catalysis of breaking Li–S bond, thereby achieving the rapid delithiation reaction kinetics. Due to their excellent conductivity, the single Ni atoms on Ni@NG can speed up electron transfer during the charge/discharge process, leading to a highly improved electrochemical performance. More recently, Lai et al.\textsuperscript{60} reported a general method for synthesizing various single-atom metals ($\text{V}$, $\text{Mn}$, $\text{Fe}$, $\text{Co}$, $\text{Ni}$, $\text{Cu}$, $\text{Ge}$, $\text{Mo}$, $\text{Ru}$, $\text{Rh}$, $\text{Pd}$, $\text{Ag}$, $\text{In}$, $\text{Sn}$, $\text{W}$, $\text{Ir}$, $\text{Pt}$, $\text{Pb}$, $\text{Bi}$) on N-doped carbon matrix (M$_1$@NC@S), and the synthesized single atoms showed excellent and adjustable catalytic performance in RT-Na/S batteries. The constructed systems composed of single-atom metal and NC exhibit different catalytic capability to S. For example, compared with the noble metal single-atom catalyst Pt$_1$@NC@S whose discharge products are mainly long-chain NaPSs, the Fe$_1$@NC@S can promote the conversion of long-chain NaPSs to short-chain Na$_2$S effectively, thus leading to a stronger adsorption ability on short-chain NaPSs. Moreover,
Fe$_1$@NC@S presents much small polarization even at high current rates, indicating that Fe$_1$ single atom can be efficient in immobilizing and converting NaPSs.

### 3.3 Inhibition of polysulfide dissolution

The shuttle effect mainly originates from the dissolution of LiPSs into electrolyte as well as the slow transformation of LiPSs. In general, contact between the electrolyte and the sulfur is vital for sulfur to be highly reactive in this reaction, which, in turn, leads to the dissolution of LiPSs. Many efforts have been made to avoid contamination of the metal anodes induced by the shuttle effect. For instance, nonpolar materials, such as mesoporous carbon or graphene, can physically trap polysulfides due to their porous structure or interlayer space. During long-term cycling, however, the adsorption capability of porosity to trap polarized polysulfides becomes weaker, resulting in the loss of polysulfides. Thus, it has been developed that stronger interactions with sulfur species can alleviate the shuttling of polysulfides into the electrolyte during cycling. Zhang et al.\textsuperscript{61} successfully prepared a high-efficiency sulfur carrier for RT-Na/S batteries via using atomic Co as an electrocatalyst loaded on hollow carbon nanospheres. The atomic Co species with their surfaces vulcanized can effectively prevent the intensive shuttling of NaPSs. Notably, the sulfur diffusion can induce the migration of atomic Co to the carbon shell to form S@Co$_n$-HC. In addition, the polysulfides that are confined in the carbon shell can be completely reduced to Na$_2$S due to the catalysis by atomic Co, thus improving the utilization rate of S and the cycling performance of batteries.

Du’s group synthesized Co–N/G (0.29 at%) by embedding monatomic Co catalyst into a nitrogen-doped graphene framework for a high-efficiency cathode for Li/S batteries.\textsuperscript{67} The sulfur loading of the S@Co–N/G-based cathode was as high as 90%. The stable Co–N–C center accelerated the redox reaction of polysulfides and efficiently captured polysulfides within the cathode, which greatly enhanced the performance.

### 3.4 Enhanced conductivity of sulfur

One of the main causes for low utilization of sulfur and poor long-term cycling in Li/S or Na/S batteries is the poor conductivity of sulfur as well as its discharge products. The low conductivity of sulfur contributes to the low utilization rate of active materials as well as poor electrochemical activity. Therefore, the cathode matrix must be conductive to make up for the nonconductivity of sulfur and its corresponding discharge products. Metals with high electrical conductivity can significantly improve the utilization rate of S. Lim et al.\textsuperscript{68} used Fe–N–C catalyst as the sulfur host for the cathode of Li/S batteries. There are strong chemical interactions between the uniformly distributed Fe–N–C sites and the soluble LiPSs, which could improve the reaction kinetics of elemental sulfur and increase the sulfur loading on matrix to 80%, as well as enhance the conductivity of sulfur.

Furthermore, Du et al.\textsuperscript{69} prepared a free-standing cathode with Co nanoparticles decorated on porous carbon nanofiber doped with nitrogen (Co@NPCNFs/S) in RT-Na/S batteries. The introduction of homogeneously distributed Co nanoparticles provided abundant active sites for sulfur, leading to improved sulfur utilization and sulfur electroactivity. In addition to the lower charge-transfer resistance, Co@NPCNFs/S showed fast electronic and ionic conductivity.

In general, the sodium storage performance of RT-Li/S and RT-Na/S batteries would be enhanced comprehensively by virtue of these four kinds of metal–sulfur interaction mechanism, which are shown in Figure 3.

### 4 Improvement of electrochemical performance

#### 4.1 Metal–sulfur interactions in RT-Li/S batteries

The intact physical confinement via using a graphene carbon matrix has been confirmed to increase the reversible capacity and improve cycling performance through the prevention of the shuttle effect. The inhibition of the shuttle effect is the key to addressing the issues of instability and fast capacity decay in Li/S batteries. The nonporous carbon matrix, however, cannot chemically and effectively interact with polar LiPSs and totally address the issues related to capacity attenuation. Although it was proved that porous carbon materials can endow a carbon matrix with multifaceted structural advantages in transferring electrons/ions, accommodating sulfur, and immobilizing polysulfides, porosity engineering is not effective for maintaining capacity and improving rate performance. As electrocatalysts, the additional introduction of metals on the carbon matrix is necessary and significant progresses have been made recently, thus attracting increasing attention from researchers. The uniformly dispersed metals are reported to form polar interactions with sulfur and LiPSs, which are expected to greatly improve RT-Li/S cells in terms of electrochemical performance. Also, the metals as sulfur/
sulfur polysulfide catalysts can rapidly catalyze S cathode into short-chain sulfides, so that the reversibility and activity of RT-Li/S batteries can be significantly improved.

For example, Cheng’s group\textsuperscript{70} studied the effects of a new cathode material based on three-dimensional (3D) graphene combined with graded porous N-doped carbon (3DG@NPC) on the electrochemical performance of Li/S systems. As depicted in Figure 4A, the sulfur grains were well embedded in 3DG@NPC/S, and no significant accumulation of sulfur was observed throughout the porous composite. Although the monoclinic phase of sulfur can enhance the rate capability of Li/S batteries, its structural instability also results in unstable cycling performance of battery systems at room temperature. 3DG@NPC/S presented a high initial capacity of 1280 mAh g\(^{-1}\), but only retained a lower reversible capacity of 667 mAh g\(^{-1}\) at 0.2 C over 500 cycles (Figure 4B). The reduction reactions from elemental S to long-chain LiPSs (Li\(_2\)S\(_n\), 4 \(\leq n \leq 8\)), as well as further to Li\(_2\)S\(_2\)/Li\(_2\)S, were not fully carried out, which is in accordance with the low reversible capacity. These results indicated that the irreversible deposition and accumulation of Li\(_2\)S\(_2\)/Li\(_2\)S on 3DG@NPC/S electrode would lead to a rapid decay in capacity for Li/S batteries. Therefore, it is necessary to introduce an electrocatalyst to promote the conversion of S to Li\(_2\)S, and effectively inhibit the serious capacity attenuation of the RT-Li/S battery. For that reason, Yao et al.\textsuperscript{58} proposed using a metal embedded in graphene to form metal (Cu, Ni, Sn)/graphene (G) composites as a high-efficiency sulfur carrier for RT-Li/S batteries. With or without the presence of graphene substrates, bond interactions between the three different metals and sulfur clusters were compared. The metal nanoparticles in the synthesized metal/graphene composites were evenly dispersed and tightly attached to the surface of graphene, with particle sizes of 30, 50, and 10 nm, respectively, which meant that all three types of metals were well embedded in the graphene materials (Figure 4C–E). The strong metal and sulfur chemical bonding could effectively immobilize sulfur species and suppress the formation of soluble polysulfide intermediates. Cu, Sn, and Ni in metal/G composites all have a good adsorption effect on sulfur clusters. Compared with the bare G-S electrode, all types of metal/G–S cathodes presented impressive capacity retention at 0.2 C, which was caused by the pivotal role of enhanced sulfur immobilization (Figure 4F). Also, the rate capacities of...
metal/graphene composites with different metals (Ni, Sn, and Cu) all exhibited excellent performance from 0.2 to 5 C (Figure 4G). The Ni/G–S cathode, in particular, presented the best electrochemical performance, with an excellent initial capacity (1092 mAh g\(^{-1}\)) and an outstanding reversible capacity of 832 mAh g\(^{-1}\), with only a 0.05% capacity decay rate over 500 cycles (Figure 4H). Most notably, the average Coulombic efficiency was also high (98.5%) over the whole electrochemical process for the Ni/G–S cathode.

Most recently, Chen’s group synthesized Ni@NG with excellent electrochemical performance for Li/S systems.\(^{59}\) As shown in Figure 5A, the elements in the layered structure of Ni@NG were uniformly distributed. The single Ni atoms were well embedded in the substrate and there were no apparent Ni nanoparticles on the Ni@NG surface (Figure 5B). Ni@NG can speed up the kinetic transformation of LiPSs during cycling via its superior catalytic activity. Moreover, the distinct discharge plateaus, even at high rates, indicated that Ni@NG can immobilize LiPSs (Figure 5C). The designed Ni@NG separator also ensured high-rate performance of the battery, as shown in Figure 5D. The battery delivered a higher capacity of 1598 mAh g\(^{-1}\) at 0.1 C than that of the battery based on Ni particles/NG (1306 mAh g\(^{-1}\)). Furthermore, with the current rate as high as 10 C, the capacity could still remain at 612 mAh g\(^{-1}\) because of the fast conversion of LiPSs, which is nearly twice that of the Li/S system based on Ni particles/NG. Upon recovering back to 0.1 C, the Ni@NG showed a high capacity, with almost 86.8% of the initial capacity over 35 cycles. Apart from excellent rate performance, the cycling performance of Li/S cells is also of great importance. The Li/S cell using Ni@NG as the separator presented a high initial capacity of 826.2 mAh g\(^{-1}\) at a current rate of 1 C after 500 cycles, and its corresponding capacity retention was 78%. Even at 10 C, the designed Ni@NG could also achieve a capacity retention of 70%
over 500 cycles (Figure 5E). Furthermore, the binding energy is also of great significance for immobilizing the LiPSs during the cycling process. The binding energy between LiPSs and Ni@NG is greater than that between LiPSs and NG, indicating efficient constraint of LiPSs and thus confirming the inhibition of the shuttle effect. The excellent conductivity of NG makes it possible to embed the Ni single atoms and speed up electron transfer. In short, the active Ni sites of the designed Ni@NG can not only fix the LiPSs, but also efficiently catalyze the reversible conversion of LiPSs, thus inhibiting the shuttle effect due to the formation of $S_{2}^{2-}$–Ni–N.

In summary, the cathode matrix typically exhibits excellent rate and cycling performances when it contains metal in Li/S battery systems. This is mainly attributed to the fact that metals can fix polysulfides by forming metal–sulfur bonds, thus efficiently impeding the shuttling of polysulfides. In addition, the introduction of metals can also improve the conductivity of matrices and the utilization rate of sulfur. Finally, metals can serve as catalysts to accelerate the conversion reaction of polysulfides. As shown in Table 1, we further summarize recent research on cathode matrices with/without metal in Li/S battery systems.

### 4.2 Metal–sulfur interactions in RT-Na/S batteries

RT-Na/S batteries, as is well known, share an analogous working mechanism to the RT-Li/S batteries, so they are confronted with crucial problems in relation to rapid capacity decay and low reversible capacity. It has been demonstrated that complete physical confinement in hollow/mesoporous carbon matrices could increase the reversible capacity and cycling stability, although the nonpolar carbon matrix cannot effectively react with polar NaPSs, nor can completely block the capacity decay. Effective material modification is the main method to increase the conductivity and reactivity of sulfur as well as to avoid the shuttling of polysulfides. Similar to RT-Li/S batteries, it is reasonable to introduce new metals into the sulfur host of RT-Na/S batteries, which can not only maximize the effect of polarized sulfur supports, but also achieve remarkable performance of RT-Na/S systems.

For instance, Wang et al. prepared a new type of sulfur host via employing interconnected mesoporous carbon hollow nanospheres (iMCHS) to investigate their electrochemical performance for RT-Na/S batteries. As
Table 1 Comparison of recent research on cathode matrices with/without metal in RT-Li/S batteries

| Material               | Morphology (metals) | Specific capacity (mAh g\(^{-1}\)) | Rate capability (mAh g\(^{-1}\)) | Cycling performance (mAh g\(^{-1}\)) | References |
|------------------------|---------------------|------------------------------------|----------------------------------|--------------------------------------|------------|
| 3DSCS                  | –                   | 1066/0.12 A g\(^{-1}\)             | 492/2.33 A g\(^{-1}\)            | 946/200th/0.12 A g\(^{-1}\)          | [71]       |
| P@CNC–S               | –                   | 1398/0.167 A g\(^{-1}\)           | 711/16.7 A g\(^{-1}\)           | 927/1000th/1.67 A g\(^{-1}\)        | [72]       |
| SFC/S                 | –                   | 1123/0.335 A g\(^{-1}\)           | 231/3.35 A g\(^{-1}\)           | 404/200th/0.335 A g\(^{-1}\)        | [73]       |
| HPC/SC                | –                   | 875/0.335 A g\(^{-1}\)           | 477/3.35 A g\(^{-1}\)           | 567/200th/1.68 A g\(^{-1}\)        | [74]       |
| GSH@APC               | –                   | 1266/0.168 A g\(^{-1}\)           | 500/3.35 A g\(^{-1}\)           | 465/250th/1.68 A g\(^{-1}\)        | [75]       |
| N,S-codoped graphene  | –                   | 1200/0.335 A g\(^{-1}\)           | 430/3.35 A g\(^{-1}\)           | 550/500th/0.838 A g\(^{-1}\)       | [77]       |
| NS–PCs                | –                   | 1265/0.168 A g\(^{-1}\)           | 942/0.50 A g\(^{-1}\)           | 629/300th/0.50 A g\(^{-1}\)        | [78]       |
| DHCSs                 | –                   | 1020/0.168 A g\(^{-1}\)           | 350/1.68 A g\(^{-1}\)           | 690/100th/0.168 A g\(^{-1}\)       | [79]       |
| N–PC@uCo              | Cluster             | 1370/0.168 A g\(^{-1}\)           | 740/3.35 A g\(^{-1}\)           | 780/500th/1.68 A g\(^{-1}\)        | [80]       |
| Co–N/G                | Single atom         | 1210/0.335 A g\(^{-1}\)           | 790/3.35 A g\(^{-1}\)           | 681/500th/1.68 A g\(^{-1}\)        | [67]       |
| B/2DMOF–Co            | Single atom         | 1138/0.168 A g\(^{-1}\)           | 478/8.38 A g\(^{-1}\)           | 703/200th/0.838 A g\(^{-1}\)       | [81]       |
| Co–N–C                | Single atom         | 1247/0.335 A g\(^{-1}\)           | 1035/3.35 A g\(^{-1}\)          | 850/300th/0.838 A g\(^{-1}\)       | [82]       |
| CoSA–N–C              | Single atom         | 1046/0.335 A g\(^{-1}\)           | 624/8.38 A g\(^{-1}\)           | 675/1000th/1.68 A g\(^{-1}\)       | [83]       |
| Fe/NG                 | Single atom         | 1200/0.335 A g\(^{-1}\)           | 673/8.38 A g\(^{-1}\)           | 892/750th/0.838 A g\(^{-1}\)       | [84]       |
| Fe–N–C/G              | Single atom         | 1142/0.335 A g\(^{-1}\)           | 848/3.35 A g\(^{-1}\)           | 602/500th/0.838 A g\(^{-1}\)       | [85]       |
| SAV@NG                | Single atom         | 1230/0.335 A g\(^{-1}\)           | 645/5.03 A g\(^{-1}\)           | 551/400th/0.838 A g\(^{-1}\)       | [86]       |
| FeVS–CN/S             | Single atom         | 1123/0.335 A g\(^{-1}\)           | 765/3.35 A g\(^{-1}\)           | 796/200th/0.335 A g\(^{-1}\)       | [87]       |
| Fe–PNC                | Single atom         | 1139/0.168 A g\(^{-1}\)           | 595/1.68 A g\(^{-1}\)           | 557/300th/0.838 A g\(^{-1}\)       | [88]       |
| SC–Co                 | Single atom         | 1140/0.167 A g\(^{-1}\)           | 1130/0.836 A g\(^{-1}\)         | 837/300th/0.836 A g\(^{-1}\)       | [89]       |
| CB–S–Au               | Nanoparticle        | 1061/0.168 A g\(^{-1}\)           | 674/0.838 A g\(^{-1}\)          | 641/160th/0.838 A g\(^{-1}\)       | [90]       |
| Co–N–GC               | Nanoparticle        | 1350/0.168 A g\(^{-1}\)           | 685/3.35 A g\(^{-1}\)           | 625/500th/1.68 A g\(^{-1}\)        | [91]       |
| MC–Cu–S               | Nanoparticle        | 1050/0.1 A g\(^{-1}\)             | 360/2 A g\(^{-1}\)              | 630/500th/0.1 A g\(^{-1}\)         | [64]       |
| Co–N–C/tGO            | Nanoparticle        | 1002/0.335 A g\(^{-1}\)           | 625/8.38 A g\(^{-1}\)           | 616/500th/0.838 A g\(^{-1}\)       | [92]       |
| Pt/C                  | Nanoparticle        | 1158/0.838 A g\(^{-1}\)           | 620/3.35 A g\(^{-1}\)           | 575/200th/0.838 A g\(^{-1}\)       | [93]       |
| Co/N–PCNSs            | Nanoparticle        | 1203/0.335 A g\(^{-1}\)           | 683/3.35 A g\(^{-1}\)           | 619/200th/3.35 A g\(^{-1}\)        | [94]       |
| CoFe@NC               | Nanoparticle        | 916/1.68 A g\(^{-1}\)             | 643/3.35 A g\(^{-1}\)           | 447.4/500th/1.68 A g\(^{-1}\)      | [95]       |
| Co–N–C                | Nanoparticle        | 1291/0.335 A g\(^{-1}\)           | 518/3.35 A g\(^{-1}\)           | 796/300th/0.335 A g\(^{-1}\)       | [96]       |
| Co–NPC–MCs            | Nanoparticle        | 1192/0.838 A g\(^{-1}\)           | 823/3.35 A g\(^{-1}\)           | 650/400th/1.675 A g\(^{-1}\)       | [97]       |
| Co@NHCRs              | Nanoparticle        | 971/0.838 A g\(^{-1}\)            | 747/1.68 A g\(^{-1}\)           | 704/100th/0.838 A g\(^{-1}\)       | [98]       |
| PtNi@C                | Nanoparticle        | 1094/0.334 A g\(^{-1}\)           | 546/3.34 A g\(^{-1}\)           | 600/300th/1.67 A g\(^{-1}\)        | [99]       |
| Co,N–G@CNT            | Nanoparticle        | 1398/0.335 A g\(^{-1}\)           | 611/10.1 A g\(^{-1}\)           | 659/1500th/1.68 A g\(^{-1}\)       | [100]      |

Depicted in Figure 6A, RT-Na/S@iMCHS shows an initial discharge capacity of 1215 mAh g\(^{-1}\) as well as a reversible capacity of 410 mAh g\(^{-1}\). The reversible charge capacity, however, is still far below the theoretical capacity of Na\(_2\)S, which means that the conversion of sodium sulfides is not totally reversible. For RT-Na/S@iMCHS, the electrochemical reaction is mainly attributed to the reversible reaction between long-chain Na\(_2\)S\(_x\) (5 ≤ x ≤ 8) and short-chain Na\(_2\)S\(_4\) (theoretical capacity of 418.75 mAh g\(^{-1}\)), which is consistent with the real capacity of 410 mAh g\(^{-1}\) (Figure 6A). These results indicate that the battery has poor reversibility when pure carbon is used.
as the sulfur host. In addition, when the cycling process exceeds 200 cycles, the accumulation of insulating Na₂S causes an increase in the internal resistance of RT-Na/S@iMCHS and the continuous attenuation of capacity (Figure 6B). Thus, the use of an electrocatalyst is vital to decreasing the energy barrier for the conversion from S to Na₂S, facilitating electrochemical processes in charge and discharge, and preventing capacity loss as well as the further increasing in the internal resistance of RT-Na/S batteries.

To address these issues, Zhang’s group investigated a new type of sulfur host, transition metal nanoclusters encased in hollow carbon nanospheres (S@M-HC) for RT-Na/S systems. Three different metals were investigated to compare their interactions with S. The metal nanoclusters were well distributed on HC, and their average size was about 1.2 nm (Figure 6C–E). Compared with M-HC matrix, the sizes of metal nanoclusters increased about 0.2–0.3 nm after S loading, which indicated that sulfur was chemically adsorbed on the metal nanoclusters, further leading to the formation of M–S bonds. Although the three cathode materials in the first few cycles presented capacity degradation due to polysulfide dissolution, the later cycles maintained relatively stable cyclability with 1000 cycles, indicating that metal nanoclusters can significantly enhance the stability of RT-Na/S cells (Figure 6F). In particular, the S@Fe-HC
exhibited the best performance, with an initial charge capacity of 1023 mAh g\textsuperscript{−1} as well as an outstanding reversible capacity of 394 mAh g\textsuperscript{−1} after 1000 cycles. Additionally, S@Fe-HC also presented good cycling stability (Figure 6G). In contrast with S@Cu-HC and S@Ni-HC, the Fe–S bonds were the strongest. The three types of cathode materials have shown good adsorption capability toward polysulfides, where the metal nanoclusters interact with Na\textsubscript{2}S\textsubscript{4} and promote its decomposition into Na\textsubscript{2}S. The theoretical calculations indicated that the introduced electrocatalyst S@Fe-HC could greatly reduce the energy barrier and facilitate the breaking up of sodium sulfides from long-chain polysulfides to short-chain polysulfides. Also, these metal nanoclusters were suggested to play a crucial role in enabling the conductivity of the S cathode to be enhanced through the formation of M–S chemical bonds. The strong reactivity of the nanoclusters could greatly increase the adsorption energy of Na\textsubscript{2}S\textsubscript{4}. The results showed that Na\textsubscript{2}S\textsubscript{4} had the fastest degradation rate on Fe nanoclusters, indicating that the Fe nanocluster had the highest reactivity. To further verify the results, DFT calculations were performed. It was shown that S@Fe-HC had the best inhibiting effect on the dissolution of polysulfides and could electrocatalytically achieve the reduction from long-chain polysulfides to short-chain sulfides.

Thus, the chemical coupling between metal nanoclusters and S can improve the performance of Na/S systems with a few advances, including the confinement of S/Na\textsubscript{2}S\textsubscript{4}, improvement of S reactivity, and moderation of the shuttle effect. The size effect of metals, however, is also crucial for the chemical interaction and has not been extensively explored. Next, Zhang et al.\textsuperscript{61} further succeeded in preparing a high-efficiency sulfur carrier via using atomic Co (Co clusters and Co single atoms) as an electrocatalyst loaded on hollow carbon nanospheres (Figure 7). Figure 7A illustrates the synthetic process for S@Co\textsubscript{a}-HC. It was shown that the atomically dispersed Co could further enhance the reactivity of S and form an interaction with it, effectively inhibiting the dissolution of Na\textsubscript{2}S\textsubscript{4} and electrocatalytically reducing Na\textsubscript{2}S\textsubscript{4} to Na\textsubscript{2}S. For instance, S@Co\textsubscript{a}-HC displayed a high initial reversible capacity of 1081 mAh g\textsuperscript{−1} at 100 mA g\textsuperscript{−1} (Figure 7B). In contrast, S@HC presented a lower reversible capacity of 580 mAh g\textsuperscript{−1} (Figure 7C). Hollow carbon without any nanoparticles was evenly dispersed, with both the atomic Co and S well embedded in the carbon shell, which was mainly ascribed to the existence of a strong interaction between Co and S (Figure 7D–F). Also, S@Co\textsubscript{a}-HC showed highly stable cycling performance with a reversible capacity of 508 mAh g\textsuperscript{−1} after 600 cycles (Figure 7G). As shown in Figure 8H, S@Co\textsubscript{a}-HC exhibited high reversible capacity at different current densities. When the rate of discharge/charge was restored to the original 0.1 A g\textsuperscript{−1}, RT-Na/S@Co\textsubscript{a}-HC showed a remarkable reversible capacity of 625 mAh g\textsuperscript{−1} after 100 cycles compared with the 367 mAh g\textsuperscript{−1} of RT-Na/S@HC. Therefore, the chemical bonds between metal and sulfur could efficiently improve the reversible capacity as well as cycling stability of RT-Na/S batteries.

These investigations indicated that sulfur host matrices, which are constructed from functional carbonaceous materials and metal-based electrocatalysts, can provide a promising method for developing high-performance RT-Na/S batteries.

To sum up, cathode matrices with metals introduced can present much better electrochemical performance than those without metals in Na/S batteries. The thus-formed metal–sulfur bonds can immobilize polysulfides and then inhibit the dissolution of polysulfides. Furthermore, the conductivity and sulfur utilization rate will be greatly improved. Also, the polysulfides will be converted quickly under the action of electrocatalysis. As shown in Table 2, we further summarize recent research on cathode matrices with/without metal in Na/S battery systems.

### 5 CONCLUSIONS AND PERSPECTIVES

Rechargeable Li/S and Na/S battery systems represent the most remarkable techniques for future energy storage because of their high energy density, as well as low cost. Unfortunately, tremendous challenges obstruct the development of metal–sulfur batteries due to the following factors: (1) the poor conductivity and severe volume expansion of sulfur; (2) the sluggish reactivity of sulfur and lithium/sodium sulfides; (3) the dissolution of long-chain polysulfides in the organic electrolyte and the formation of low-order polysulfides caused by the diffusion of polysulfides from cathode region to the anode region, which leads to corrosion of the anode and low Coulombic efficiency. Metals have aroused increasing interest for RT-Li/S and RT-Na/S cells because of their strong adsorption effect on sulfur and polysulfides, which can not only enhance the reactivity of S but also impede the shuttle effect. In addition, the introduction of metals can decrease the energy barrier to enhance the reversibility of Li/Na–S batteries. Overall, their electrochemical properties can lead to a significant improvement for RT-Li/Na–S batteries (Figure 8). Unlike the hosts without metal catalysis, as shown in Figure 8, the improved electrochemical performance in both Li/S and Na/S systems that use metal species in sulfur hosts has meant that they make headway to become the prime choice.

In this review, we have summarized the working mechanism of metal catalysts by analyzing the results
FIGURE 7  (A) Schematic illustration of the synthesis of atomic Co-decorated hollow carbon–sulfur host (S@Co₆-HC). Discharge/charge curves of (B) S@Co₆-HC and (C) S@HC at 100 mA g⁻¹. (D) Transmission electron microscopy image (TEM) and (E, F) high-angle annular dark-field scanning TEM (HAADF-STEM) images of S@Co₆-HC. (G) Cycling performances and (H) rate performances of S@Co₆-HC and S@HC. Reproduced with permission: Copyright 2018, Nature.

FIGURE 8  Rate performance of selected cathode materials with/without metals in (A) RT-Li/S and (B) RT-Na/S batteries.
of XRD, Raman spectroscopy, and DFT. It has been demonstrated that metals can be used as electrocatalysts to accelerate the conversion from long-chain polysulfides to short-chain polysulfides, which can alleviate the shuttle effect as well as enhance the electrochemical activity of sulfur. Furthermore, the metal–sulfur interaction mechanism has been comprehensively summarized from four aspects: (1) sulfur immobilization, (2) catalytic capability of metal species, (3) inhibition of the dissolubility of sulfur/sulfide-related materials, and (4) improved conductivity of sulfur/sulfide-related compounds. Moreover, we analyze the influence of introduced metals on the electrochemical performance of Li/Na sulfur batteries.

Nevertheless, the current metal loading mass is relatively low, and the types of metals being explored are relatively limited. Furthermore, our current understanding of the catalytic mechanism of metals is still incomplete. In the future, it is vital to develop novel synthetic methods with easy operation and a large-scale production rate. It is of great significance to develop in situ techniques to deeply understand the catalytic mechanism of metals through integration with theoretical calculations. For example, through in situ Raman spectroscopy and in situ XRD, the sulfur phase transition process during charging and discharging can be obtained, and the role of the metal catalyst in the phase transition processes of sulfur can be inferred by the intensity and position of the peak during charging and discharging. In addition, it is a promising and interesting approach to explore metals on new substrates in detail, including size effects, synergistic effects between metals, and loading ratios. The current status of Li/Na–S batteries falls short of meeting the requirements of practical applications, but in the near future, significant advances in materials, as well as battery designs, are very likely to promote Li/Na–S batteries to suitability for practical applications.

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**CONFLICT OF INTERESTS**

The authors declare that there are no conflict of interests.

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**TABLE 2** Comparison of recent research on cathode materials with/without metal in RT-Na/S batteries

| Material          | Morphology (metals) | Specific capacity (mAh g\(^{-1}\)) | Rate capability (mAh g\(^{-1}\)) | Cycling performance (mAh g\(^{-1}\)) | References |
|-------------------|---------------------|-----------------------------------|----------------------------------|-------------------------------------|------------|
| iMCHS             | –                   | 391/0.1 A g\(^{-1}\)              | 127/5 A g\(^{-1}\)              | 292/200th/0.1 A g\(^{-1}\)         | [101]      |
| CFC               | –                   | 390/1.168 A g\(^{-1}\)            | 48/1.675 A g\(^{-1}\)           | 120/300th/0.168 A g\(^{-1}\)       | [102]      |
| CS90-rGO          | –                   | 650/0.2 A g\(^{-1}\)              | 285/1 A g\(^{-1}\)              | 498/55th/0.2 A g\(^{-1}\)          | [103]      |
| MCS               | –                   | 700/0.168 A g\(^{-1}\)            | 370/1.68 A g\(^{-1}\)           | 306/1500th/1.68 A g\(^{-1}\)       | [104]      |
| MCP               | –                   | 1614/0.168 A g\(^{-1}\)           | 600/0.838 A g\(^{-1}\)           | 800/50th/0.168 A g\(^{-1}\)        | [14]       |
| CNT@MPC           | –                   | 1148/0.168 A g\(^{-1}\)           | 800/3.35 A g\(^{-1}\)           | 1000/20th/0.168 A g\(^{-1}\)       | [15]       |
| CS                | –                   | 1006/0.04 A g\(^{-1}\)            | 730/1.2 A g\(^{-1}\)            | 650/500th/0.4 A g\(^{-1}\)         | [105]      |
| C                 | –                   | 1336/0.268 A g\(^{-1}\)           | 843/0.536 A g\(^{-1}\)           | 811/950th/5.36 A g\(^{-1}\)        | [106]      |
| CM                | –                   | 3962/0.1 A g\(^{-1}\)             | 527/2 A g\(^{-1}\)              | 1250/500th/0.5 A g\(^{-1}\)        | [107]      |
| C\(_{con-NC}\)    | Nanocluster         | 820/0.1 A g\(^{-1}\)              | 220.3/5 A g\(^{-1}\)            | 508/600th/0.1 A g\(^{-1}\)         | [61]       |
| 3D Cu foam        | Foam                | 1403/0.05 A g\(^{-1}\)            | 942/0.05 A g\(^{-1}\)            | 377/5th/0.05 A g\(^{-1}\)          | [108]      |
| Fe@HNC            | Single atom         | 359/0.1 A g\(^{-1}\)              | 56.9/10 A g\(^{-1}\)            | 180/200th/1 A g\(^{-1}\)          | [109]      |
| Fe–N–C/S–MCF      | Single atom         | 1543/0.838 A g\(^{-1}\)           | 798/5.025 A g\(^{-1}\)           | 322/500th/5.025 A g\(^{-1}\)       | [68]       |
| Fe–HC             | Nanoparticle        | 820/0.1 A g\(^{-1}\)              | 220/5 A g\(^{-1}\)              | 394/1000th/0.1 A g\(^{-1}\)        | [60]       |
| Co@NPFCs          | Nanoparticle        | 906/0.168 A g\(^{-1}\)            | 154/5.03 A g\(^{-1}\)           | 411/800th/1.68 A g\(^{-1}\)        | [69]       |
| CN/Au             | Nanoparticle        | 701/0.1 A g\(^{-1}\)              | 181/20 A g\(^{-1}\)             | 369/2000th/10 A g\(^{-1}\)         | [110]      |
| HSMC–Cu           | Nanoparticle        | 710/0.0503 A g\(^{-1}\)           | 350/2.01 A g\(^{-1}\)           | 610/110th/0.0503 A g\(^{-1}\)      | [111]      |
| Co/C/rGO          | Nanoparticle        | 485/0.838 A g\(^{-1}\)            | 291/8.375 A g\(^{-1}\)          | 226/100th/1.838 A g\(^{-1}\)       | [112]      |
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