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Review

# High-entropy liquid electrolytes in rechargeable batteries: Merits and challenges

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ABSTRACT

The growing severity of environmental challenges has accelerated advancements in renewable energy technologies, highlighting the critical need for efficient energy storage solutions. Rechargeable batteries, as primary short-term energy storage devices, have seen significant progress. Among emerging optimization strategies, high-entropy electrolytes have garnered attention for their superior ionic conductivity and ability to broaden batteries' operational temperature ranges. Rooted in the thermodynamic concept of entropy, high-entropy materials, originally exemplified by high-entropy alloys, have demonstrated enhanced structural stability and advanced electrochemical performance through the synergistic integration of multiple components. High-entropy liquid electrolytes, both aqueous and non-aqueous, offer unique opportunities for entropy manipulation due to their inherently disordered structures. However, their complex compositions present challenges, as minor changes in formulation can lead to significant performance variations. This review introduces the fundamentals of entropy tuning, surveys recent advances in high-entropy liquid electrolytes, and analyzes the interplay between entropy and electrochemical behavior. Finally, it discusses design strategies and future perspectives for the practical implementation of high-entropy liquid electrolytes in next-generation energy storage systems.

## 1. Introduction

Increasingly severe environmental issues are motivating the vigorous advancements of renewable energy techniques. Meanwhile, the inherent intermittency of renewable sources places more demands on energy storage devices. Thus, rechargeable batteries, as the primary short-term energy storage agents, have also witnessed flourishing developments in recent decades. Many researchers have devoted much effort to advanced materials development, configuration upgrades, and management systems tuning for batteries to improve their overall performance. Among these strategies, the evolution of materials is the most rapid, and electrolytes, which interact with both electrodes, have been optimized continuously. Advanced electrolyte matrices with effective additives,[1,2] refined salts/solvents ratios,[3] or fine-tuned (quasi)-solid hosts[4,5] have emerged. Within them, high-entropy electrolytes are becoming a promising newcomer for their superior ionic conductivity and efficient extension of the batteries' working-temperature scenario.

Entropy ($S$) was first formalized by Rudolf Clausius in the 1850s to describe the randomness of the system.[6] As the core concept of the second law of thermodynamics, entropy, combined with enthalpy ($H$), can be used to describe the thermodynamic properties of an electrolyte system.[7,8] As described in the Gibbs free energy calculation ($G = H - TS$), the increase in entropy could significantly decrease the overall Gibbs free energy.[9] Thus, entropy tuning has been comprehensively employed for materials research, and the most successful representative is the high-entropy alloy.[10,11] By incorporating multiple metals, the entropy of the system can be elevated to a level sufficient against the enthalpy for forming undesired metallic compounds, and the lattice can be finely tuned.[12,13] As a result, the alloy can provide favorable structural stability and rich electroactive sites.[14–16] Meanwhile, the synergistic effect

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among diverse elements integrates all of their electrochemical advantages. Inspired by high-entropy alloys, high-entropy sulfides,[17] nitrides,[18] and oxides[19–21] were subsequently proposed, and the employment of high-entropy materials in electrochemical storage devices started with the high-entropy oxides. Berardan et al. first proposed the $(MgCoNiCuZn)_{1-x-y}Ga_yA_xO$ (where A = Li, Na, or K) high-entropy oxide as the solid-state electrolyte for Li-ion batteries.[10] Then, Sarkar et al. reported a $(Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})O$ anode in 2018,[22] and Wang et al. reported a $Li_x(Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})OF_x$ cathode in 2019,[23] for advanced Li-ion batteries.

Given the highly disordered structure of electrolytes, especially liquid ones, the manipulation of entropy can be more convenient. Meanwhile, challenges remain under the complicated composition of high-entropy electrolytes. Besides the view from entropy, the single element within the matrix can exert influence independently, and its intercorrelated functions are highly interactive and overlapped. Thus, even a minimal change of electrolyte composition or the stoichiometric ratio will result in significant discrepancies. As the development of high-entropy electrolytes is just at the beginning stage, the underlying mechanism between the entropy and electrochemical behavior needs further explanation. Herein, starting from the introduction of fundamentals of entropy tuning, a comprehensive review of recent developments of high-entropy liquid electrolytes, including both aqueous and non-aqueous electrolytes, will be carried out. The role of entropy in electrolytes and design strategies of high-entropy liquid electrolytes will be analyzed. Then, a perspective regarding the future practical implementation and development routines of high-entropy liquid electrolytes will be provided.

## 2. Mechanism and advantages of entropy tuning

As an abstract description, the entropy can be quantified through diverse routines, and these quantifications are closely related to the physicochemical properties of electrolytes. These indices do not exist independently. Instead, the increase of the overall entropy causes the parallel tendency for all these different descriptions of entropy, causing corresponding variations in electrolyte characteristics. Major entropy concepts related to liquid electrolytes mainly include the systematic entropy change ($\Delta S$), which influences the Gibbs free energy change ($\Delta G$), the configuration entropy ($S_{config}$), the excess entropy ($S_{ex}$), the migration entropy ($S_m$), and the tetrahedral entropy ($Sq_{tet}$).

The common formation process of a liquid electrolyte includes two ordered procedures: the dissociation of the lattice existing in the original solutes, and the recombination of solutes with solvent molecules, which is also called the solvation process.[24] This process is governed by the second law of thermodynamics and can be evaluated through the Gibbs energy change, calculated with Equation (1).

$$\Delta G = \Delta H - T\Delta S \tag{1}$$

In this equation, $\Delta G$ is the Gibbs free energy change; $\Delta H$ is the enthalpy change; $T$ is the temperature of the system, and $\Delta S$ is the change of entropy. The enthalpy can be calculated through the summation of the internal energy of system ($U$) and the product of the pressure ($p$) and volume ($V$) of the system. Meanwhile, the entropy of this process can be simplified as the combination of the entropy for dissociation and solvation. Thus, the dominant equation can be rewritten into Equation (2).

$$\Delta G = U + pV - T(\Delta S_{dis} + \Delta S_{sol}) \tag{2}$$

The Gibbs free energy gap is typically used to describe the possibility of the spontaneous initiation, and a process with a negative Gibbs free energy gap can be expected to happen without the requirement of any external inputs to overcome the energy barrier. Thus, the $\Delta G$ of the solution fabrication should be less than 0.[25] As a negatively contributing factor, the elevation of entropy can significantly compensate for the potential increase of the systematic enthalpy.[26] Thus, the entropy tuning makes the Gibbs free energy change more negative, making the electrolyte more stable and facilitating the overall electrochemical reaction rate.

The degree of chaos can be described by the randomness of the distribution of different compositions. The concept regarding the position of constituent particles can be described with the $S_{config}$, which can be calculated with Equation (3).[27]

$$S_{config} = -k_B \sum_{n=1}^{W} P_n \ln P_n \tag{3}$$

In this equation, $k_B$ is the Boltzmann constant, and $P_n$ represents the possibility for the system to be in the state $n$. The summation is carried out with a total of $W$ possible configurations. Thus, with a more complicated matrix, by introducing diverse solutes and solvents, the number of states can be significantly increased to elevate the configurational entropy. From the physicochemical view, the higher $S_{config}$ makes the degree of freedom of ions more advanced, reducing their inclination to crystallization.[27] Consequently, the solubility of all these salts increases, and the more complicated solute-solvent interactions can be established.[28,29]

The ionic conductivity, as the core criterion for evaluating an electrolyte, is also related to entropy through the calculation of the ion diffusion coefficient. Equation (4) describes the logarithmic relationship between the excess entropy ($S_{ex}$) and ion diffusion coefficient ($D$).[30]

$$D = ae^{\frac{bS_{ex}}{k_B}} \tag{4}$$

The excess entropy represents the difference between the actual entropy of the system and the entropy of the ideal mixture at the same temperature and density. Furthermore, as a and b are all empirical fitting constants, the ion diffusion coefficient will be directly improved with the growth of the excess entropy. By promoting ion movement in the electrolyte and at the electrolyte/electrode interface, the energy barrier for the electrochemical reactions can be reduced.[31,32]

Another entropy concept describing ionic conductivity is the migration entropy ($S_m$). The $S_m$ arises from the vibration generated during the migration of the charge carriers, which differ in different batteries. The connection between $S_m$ and ionic conductivity can be established with the bridge of Arrhenius pre-factor, and the whole calculation includes two equations shown below.[33]

$$\sigma T = \sigma_0 e^{-\frac{E_a}{k_B T}} \tag{5}$$

$$\sigma_0 = \frac{znq^2}{k_B} e^{\frac{\Delta S_m}{k_B}} \alpha_0^2 \omega_0 \tag{6}$$

In Equation (5), $\sigma$ is the ionic conductivity; $\sigma_0$ is the Arrhenius pre-factor, and $E_a$ is the activation energy. Therefore, the ionic conductivity is proportionally related to $\sigma_0$, and $\sigma_0$ can be obtained through Equation (6), with $z$ as the geometrical factor, $n$ as the concentration of the charge carrier, $q$ as the charge, $\alpha_0$ as the migration distance, and $\omega_0$ as the attempt frequency. These factors are commonly settled once the electrochemical system is fabricated. Therefore, once $\Delta S_m$ is elevated, $\sigma_0$ will become higher, leading to an enhanced ionic conductivity of the electrolyte.[12]

The last one is the tetrahedral entropy ($Sq_{tet}$), representing solvent molecules' local tetrahedral orderliness.[34] It can be described by Equation (7). In this equation, $q_{tet}$ is the tetrahedral parameter, and $S_0$ can be calculated through Equation (8).

$$S_{q_{tet}} = S_0 + \frac{3}{2} k_B \int_{q_{tet,min}}^{q_{tet,max}} \ln(1 - q_{tet}) P(q_{tet}) dq_{tet} \tag{7}$$

$$S_0 = k_B \left( \frac{\ln \Omega_0 + \frac{3}{2}\ln 8}{3} \right) \tag{8}$$

The influence of $Sq_{tet}$ on the properties of electrolytes cannot be identified directly from the calculation. Instead, the description of the physical property of $Sq_{tet}$ implies that the increase in the value contributes to reducing the freezing point and improving dynamics. The tetrahedral configuration is closely related to the H-bond network, so the higher $Sq_{tet}$ breaks the ordered network, effectively lowering the freezing point.[35] Meanwhile, the loose tetrahedral configuration means the enlargement of both the translational and rotational entropy of solvent molecules, indicating facilitated kinetics.

In summary, entropy is an objective physical property related to the randomness of the system. For the electrolyte, the superposition of diverse components can significantly improve the systematic entropy. By separating different concepts from the overall entropy, and establishing their connections to the physicochemical properties of the electrolyte, it can be concluded that high-entropy liquid electrolytes typically obtain a higher ionic conductivity and a broader range of working temperature. Meanwhile, considering the synergetic effects of these functional constituent parts, high-entropy liquid electrolytes are extremely attractive for rechargeable batteries (Fig. 1). However, the complexity not only confers advantages but also casts a shadow over the underlying mechanism. Thus, a comprehensive review of recent progress of high-entropy electrolytes consisting of both aqueous and non-aqueous ones becomes necessary to bridge the gap between those traditional entropy theories and the practical projection of these merits into electrolyte design.

## 3. Aqueous high-entropy liquid electrolytes

Aqueous electrolytes employ water as the basic solvent, so the inherent safety can be ensured. Meanwhile, the higher degree of freedom of molecules' movement in aqueous agents and an abundant reservoir of water endow them with additional advantages of the favorable ionic conductivity and economic effectiveness.[36] However, compared to organic solvents, water maintains a high freezing point ($T_f = 0$ °C), so the ions' movements are significantly restricted when the temperature drops.[37] As a result, the low-temperature application of aqueous batteries becomes challenging. Meanwhile, due to the water electrolysis, the electrochemical stability window (ESW, 1.23 V) is narrow for the aqueous electrolyte.[38] Once the voltage exceeds this range, the hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) will happen. Taking the aqueous zinc metal battery as an example, for the HER, which is the major competing reaction to the $Zn^{2+}$ reduction process, will plunder electrons to cause damage to the overall capacity. Furthermore, the electrolysis of the water will continuously consume electrolytes. Noticeably, these parasitic reactions are interactive, and they will exacerbate the dendrite formation. Specifically, the HER will decrease the content of protons, which results in the elevation of local $OH^-$ concentration. This alkaline micro-environment favors the reaction with zinc to form the insulating products like $Zn(OH)_2$, ZnO, $Zn_4(OH)_6SO_4{\cdot}xH_2O$, etc. These side-products will passivate the zinc anode surface to cause a more non-homogeneous local electric field, resulting in the promotion of dendrite growth.[39] Fortunately, because these two drawbacks are both derived from the water activities, the H-bond network trimming brought by the entropy tuning mentioned in Section 2 can effectively solve these issues.

Once aqueous batteries are mentioned, electrodes employing transition metals like Zn, Mg or Al with acceptable inertness to water are commonly discussed. Different from Li, Na, and K metals, which are extremely active in water, these metals are relatively stable, and only moderate water-induced side reactions will happen. Thus, unlike Li-ion batteries, these batteries usually utilize metal directly as the anode material, so many Zn, Mg, and Al-ion batteries can also be classified as metal batteries.[40] On the contrary, once aqueous electrolytes are utilized in batteries with alkali metal (Li, Na, K) ions as charge carriers, only ion-containing compounds can be used to avoid the explosion. At the same time, the excessive redox potential for these types of ions will be wasted because of the narrow ESW of the aqueous electrolyte. Thus, research regarding aqueous high-entropy electrolytes is still focusing on the rechargeable aqueous zinc-ion batteries (AZIBs).

Besides the merits associated with aqueous electrolytes, zinc anodes brought additional advantages, including the favorable theoretical capacity (820 mAh $g^{-1}$, or 5855 mAh $cm^{-3}$), acceptable redox potential (−0.763 V versus standard hydrogen electrode (SHE)), and abundant storage amounts.[41,42] To the best of our knowledge, the first attempt to design high-entropy electrolytes in AZIBs was reported by Yang et al.[43] They added 2 M LiCl salts into the 2 M $ZnCl_2$ to form the high-entropy

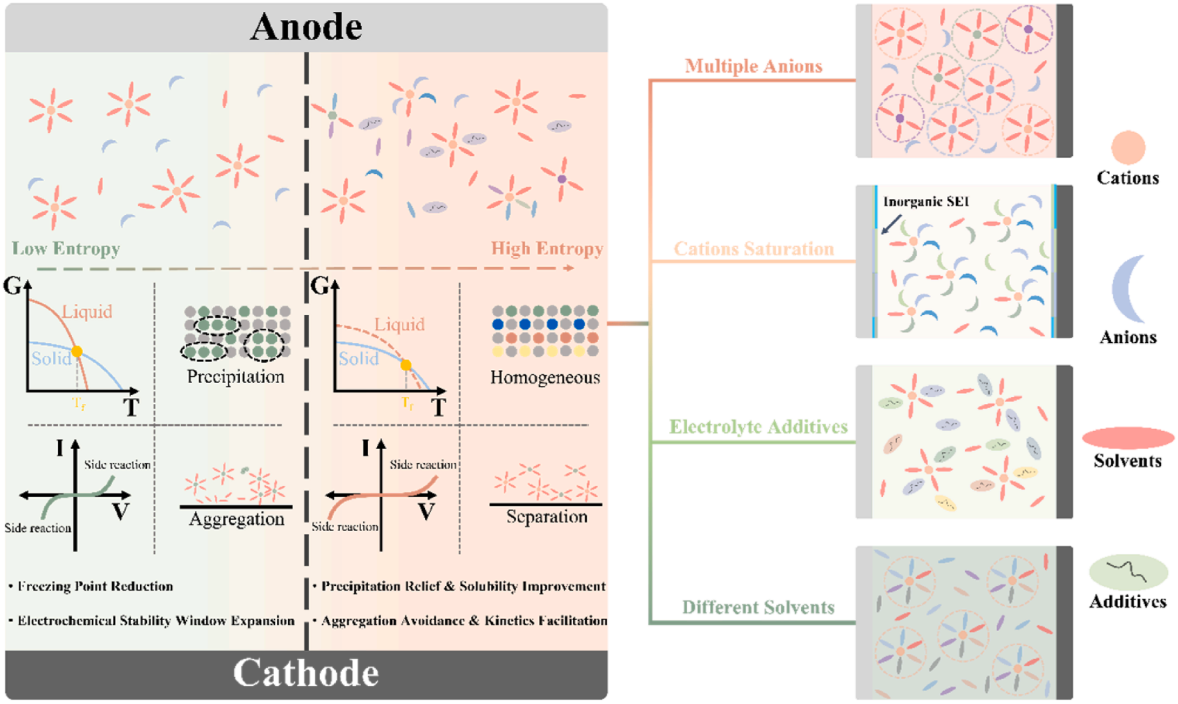


**Fig. 1.** Scheme of the strategies, the underlying mechanism, and the influence of entropy tuning.

electrolytes. With the duplication of the constituent elements, the systematic entropy was significantly increased. At low concentration of LiCl, water remained as free solvent. As more LiCl was added, water molecules increasingly coordinated with $Li^+$. Meanwhile, the $Cl^-$ anions preferentially replace water molecules from the solvation sheath of $Zn^{2+}$, and then help form $ZnCl_4^{2-}$ anions or $[ZnCl_{4-m}^{2-m}]_n$ anion clusters as shown in Fig. 2a. As a result, the content of free water was reduced to avoid water-induced parasitic reactions (Fig. 2b). At the same time, the increasing concentration of Li salts resulted in less chemically bound water in the $Li_2ZnCl_4{\cdot}xH_2O$ complex, so the length of anion clusters was restricted, resulting in less movement resistance and facilitated transportation kinetics. The strong adsorption of water molecules by $Li^+$ can cause continuous stretching and breaking of H-bonds, avoiding the crystallization of water even at low temperatures, which is represented by a decreased $T_f$. Combining all these merits, as indicated in Fig. 2c, this pioneering high-entropy aqueous electrolyte contributed to a stable operation of the zinc-air batteries for more than 800 h within a broad range of temperatures from −60 °C to 80 °C.

The entropy can be further increased through integrating more cationic constituents. For example, Wang et al. proposed a ternary electrolyte containing $Zn(ClO_4)_2$, $LiClO_4$, and $NaClO_4$ to take advantage of their different solvation structures.[44] With varying solvation sheaths and distinctive binding strengths with molecules, ion clusters can be formed to keep ions at a favorable distance (Fig. 2d). Thus, the aggregation of ions and solvents can be avoided to facilitate both the ion transportation and desolvation kinetics. Meanwhile, an optimized H-bond environment also reduces the water activity to suppress parasitic reactions and ensure the stable operation at low temperatures (Fig. 2e). Notably, most of the cathodes adopted in AZIBs, like vanadium-based oxides and manganese-based oxides, also suffer from water-induced active materials loss because of their non-negligible dissolution in water. Therefore, the restricted water network in high-entropy electrolytes also contributes to the cathode stability. Thus, as shown in Fig. 2f, this ternary electrolyte presented no capacity decay in both Zn||NaVO and Zn||PANi full batteries for over 30000 and 20000 cycles, respectively, at −20 °C.

Another design strategy besides employing multiple cations to disrupt the H-bond network is to introduce diverse anions. The anions possess an intrinsic attraction to cations, so the universal existence of anions will make them highly likely to substitute water molecules in the solvation sheath of $Zn^{2+}$. As a result, the reconstruction of the water environment and the trimming of interfacial water contents can be achieved (Fig. 2g). For instance, Wang et al. fabricated the aqueous electrolytes containing five kinds of Zn salts, including $ZnSO_4$, $Zn(Ac)_2$, $Zn(OTf)_2$, $Zn(ClO_4)_2$, and $Zn(BF_4)_2$.[45] As depicted in Fig. 2h, their calculations based on the density functional theory and molecular dynamics simulation confirmed that only 50 mM of these salts can exclude water from the solvation sheath of most $Zn^{2+}$, and all anion-substituted solvation structures presented a higher binding energy compared to the original $Zn[H_2O]_6^{2+}$ complex. Attributed to the highly disordered solvation structure, both the anode and cathode stabilities were highly improved. Consequently, an exceptionally long operation of symmetric cells for over 2000 h and a high capacity retention of 70.4% after 10000 cycles in Zn||PANi full cells were exhibited. These anions are catchers for $Zn^{2+}$ to interrupt the pristine water-dominated electrolyte environment. Therefore, the position of catchers can also be occupied by polar organic contents with a stronger binding force compared to water. Thus, Zheng's group prepared a series of electrolytes containing salts, water, and 'tentacles' like Ethylene glycol, propylene glycol, glycerol, etc.[46] These additives presented a stronger tendency to substitute water in the solvation sheath compared to anions, and their inherent low freezing temperature further expanded the range of working temperature,

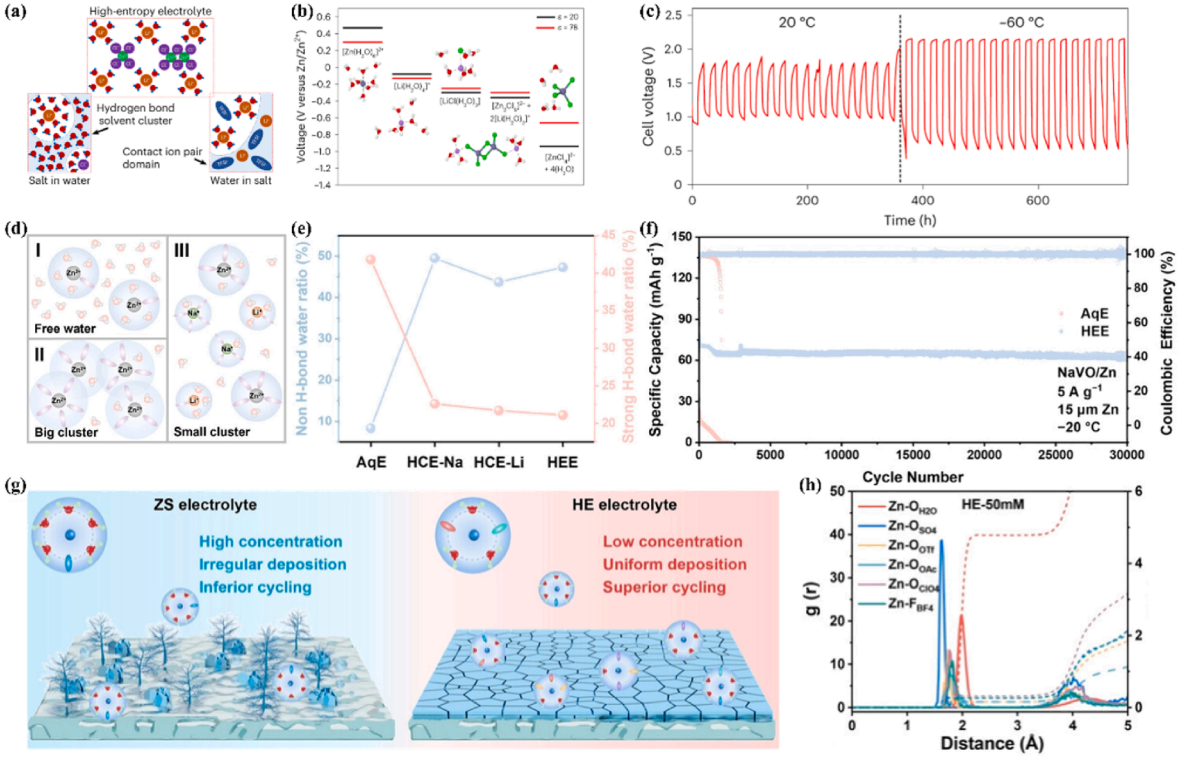


**Fig. 2.** Entropy tuning strategies in aqueous zinc ion batteries. (a) Schematic demonstration of solvation structure variation in different electrolytes. (b) Reduction potentials for hydrogen evolution reactions with different coordination structures. (c) Cycling performance for Zn-air batteries. (a–c) Reproduced with permission from Ref. 43. (d) Illustration of the electrolyte environment in different electrolytes. (e) Distribution of non-H-bond water and strong H-bond water. (f) Cycling performance of Zn||NaVO full batteries at 5 A $g^{-1}$. (d–f) Reproduced with permission from Ref. 44. (g) Schematic demonstration of the zinc anode interfacial environment in $ZnSO_4$ and high-entropy electrolytes. (h) $Zn^{2+}$ coordination environment in 50 mM high-entropy electrolytes. (g, h) Reproduced with permission from Ref. 45.

conjugated to the entropy tuning.

These design strategies can also be applied to alkali metal (Li, Na, K) ion batteries. Even though a zinc metal battery can be described as a kind of zinc ion battery, these alkali-metal ion batteries can only use ion-containing materials instead of pure metal as electrodes due to their active performance in water. In 2022, Xu et al. developed a ternary eutectic aqueous electrolyte containing LiTFSI, KOH, and $CO(NH_2)_2$, suitable for $LiMn_2O_4||Li_4Ti_5O_{12}$ Li-ion battery.[47] The firm H-bond between $CO(NH_2)_2$ and $H_2O$ stabilized the free water and reduced the interfacial water contents; as a result, an astonishing ESW of over 3.3 V was achieved in this aqueous electrolyte (Fig. 3a). This wide ESW makes the formation of a solid electrolyte interphase (SEI) more feasible in the aqueous electrolyte. The SEI is a thin film that is conductive for ions and insulating for electrons. This robust layer can effectively protect the electrode from the corrosion of the electrolyte and make the distribution of charge carriers more uniform. This layer can be formed at the beginning when the electrode is in contact with the electrolyte or during the charging/discharging process. In the latter case, the major components of the SEI are generated through the decomposition of the electrolyte. Therefore, in aqueous electrolytes, if the decomposition voltage exceeds the basic ESW of 1.23 V, water consumption will happen, and corresponding parasitic reactions will be severe. Thus, an extended ESW of aqueous electrolyte is favorable for the SEI establishment. Just as demonstrated in Fig. 3b and c, this wide ESW of 3.3 V ensured a two-step SEI formation. The addition of KOH created an alkaline environment that promoted the precipitation of $F^-$ to form the inner LiF SEI layer. Meanwhile, $CO(NH_2)_2$ can be polymerized into polyurea at low voltages to form a double SEI structure. This reconstructed H-bond configuration and more stable electrode/electrolyte interfaces contributed to an outstanding capacity retention of 92% for over 470 cycles in a $LiMn_2O_4||Li_4Ti_5O_{12}$ pouch cell (Fig. 3d and e). This deep eutectic electrolyte engineering was also applied to aqueous Na- and K-ion batteries by Zheng et al. They chose $TFSI^-$, $OTf^-$, and $PTFSI^-$ anions to fabricate $Na(PTFSI)_{0.65}(TFSI)_{0.14}(OTf)_{0.21}\cdot 3H_2O$, and $K(PTFSI)_{0.12}(TFSI)_{0.08}(OTf)_{0.8}\cdot 2H_2O$ ternary electrolytes (Fig. 3f).[48] As depicted in Fig. 3g and h, the introduction of asymmetric PTFSI-salts with the favorable solubility in water significantly increased the cations/$H_2O$ ratio and strengthened the fragile S-F bond. As a result, the ESW was significantly expanded.

In summary, the entropy tuning in aqueous electrolytes can be described as regulating H-bonds by disrupting the original water mapping. Effective strategies can be divided into two clusters: the employment of water-catchers (other cations) to form diverse solvation sheaths to avoid the aggregation of ions and solvents, and the introduction of cation-catchers (anions, polar organic molecules, etc.) to exclude water from the solvation sheath for reducing interfacial water contents. Both routines can reduce free water to improve the stability of both electrodes, and once the temperature decreases, a free-water-lean environment can retard the crystallization of water to lower the freezing point. The decreased freezing point means an improved ionic conductivity at low temperatures. As a result, not only the expansion of working scenarios is achieved, but also the performance of batteries is improved at feasible but not favorable conditions, like temperature higher than the freezing point but much lower than the ambient temperature.

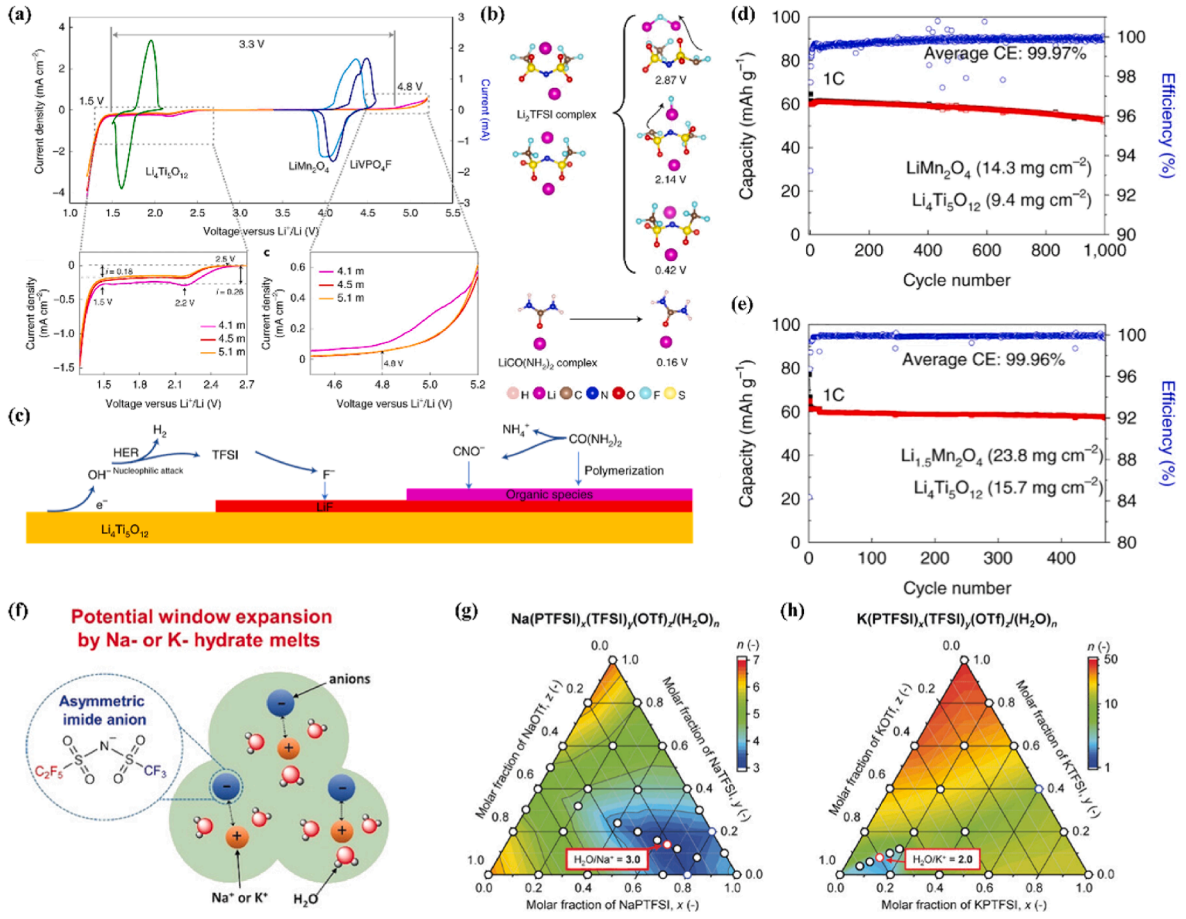


**Fig. 3.** Entropy tuning strategies in aqueous non-zinc batteries. (a) Electrochemical stability windows of different electrolytes. (b) Reduction potentials for different components. (c) Schematic demonstration of the double-layer SEI formation. (d, e) Cycling performance of $LiMn_2O_4||Li_4Ti_5O_{12}$ pouch cell with different amounts of active materials. (a–e) Reproduced with permission from Ref. 47. (f) Illustration of changes in bonds with different additives. Ternary liquidus phase diagrams of (g) aqueous high-entropy Na-ion electrolyte, and (h) aqueous high-entropy K-ion electrolytes. (f–h) Reproduced with permission from Ref. 48.

## 4. Non-aqueous high-entropy liquid electrolytes

Most commercially successful rechargeable batteries, like Li-ion and Na-ion batteries, still employ organic electrolytes to take full advantage of their high voltage output. Meanwhile, because Li, Na and K are stable in organic solvents, it is becoming a trend to use pure Li, Na, and K metals as electrodes to maximize the energy density. These burgeoning batteries are called Li-, Na-, and K-metal batteries.

Instead of water, solvents such as ethylene carbonate (EC), diethyl carbonate (DEC) and other linear carbonates, including dimethyl carbonate (DMC) and ethyl methyl carbonate (EMC), are typically used.[49–51] Even though these organic solvents typically obtain a much lower $T_f$ compared to water, the precipitation of salts generally happens once an extremely low working temperature is required.[52] Furthermore, the higher ionic conductivity should be consistently pursued, especially at low temperatures. Similar to aqueous high-entropy electrolytes, entropy tuning for non-aqueous electrolytes is summarized as two clusters: introduction of multiple salts or solvents. Because organic additives are heterogeneous to aqueous base, but are homogeneous to organic solutions, the strategy to add organic catchers mentioned in Section 3 can be classified as a multiple-solvent strategy here. As the most successful representative, the Li-ion battery will be mainly discussed, and the mapping from the Li-ion battery to other organic batteries will be followed.

Similar to aqueous electrolytes, the utilization of multiple salts can bring diverse solvation structures to the system to avoid the aggregation of salts and solvents, ensuring the boosting of the transportation kinetics of charge carriers. These merits contribute to a suitable ionic conductivity under low temperatures, and the corresponding elevation of solubilities relieves the salting-out issue. Furthermore, the entropy tuning strategy makes the adjustment of the SEI formation possible. Firstly, the entropy tuning can increase the solubility of the necessary sources of SEI formation. Secondly, variation and expansion of constituents can change interfacial reactants. Thirdly, the expansion of ESW also makes the decomposition of potential SEI contributors possible. For example, Wang et al. designed a high-entropy organic electrolyte by dissolving LiFSI, LiDFOB, LiTFSI, and $LiNO_3$ with 0.1 M of each in a commercial 1.0 M $LiPF_6$ EC/DMC solution to form a 1.4 M electrolyte.[53] As depicted in Fig. 4a and b, the diverse components improve the homogeneous distribution of different constituent ions. The most noteworthy advancement here is the increased solubility of $LiNO_3$. $LiNO_3$ is now one of the most prevailing additives because it is the main contributor to the multiple stable SEI compositions, like $Li_3N$ or $LiNO_2$. Therefore, the elevation of entropy improves the content of these sacrificial additives, leading to the formation of a more extensive and homogeneously distributed SEI. As a result, both anode and cathode interphase stabilities were improved, and a capacity retention was kept at 82% after 400 cycles at 0.5 C (Fig. 4c). More encouragingly, the fast charge/discharge at 9 C in the Li||NCM811 battery (Fig. 4d), and a favorable discharge capacity at an extremely low temperature of −50 °C (Fig. 4e) were realized. Different from aqueous electrolytes, for which SEI is hard to establish due to the narrow ESW of water, the SEI formation in organic batteries is always a key topic. Thus, in the later work, the SEI formation with the same constituent salts was investigated in more detail. Wang et al. used the five kinds of salts with different anions, but they kept the concentration of all these salts at 0.2 M instead of making the $LiPF_6$ excessive.[54] They proposed that the diversity of salts stimulated the participation of anions in the solvation sheath. Because the organic solvent was replaced with anions, as illustrated in Fig. 4f, more salt content instead of solvents would arrive at the electrode/electrolyte interface. Consequently, SEI would be mainly derived from the decomposition of salts instead of solvents. For traditional SEI established from

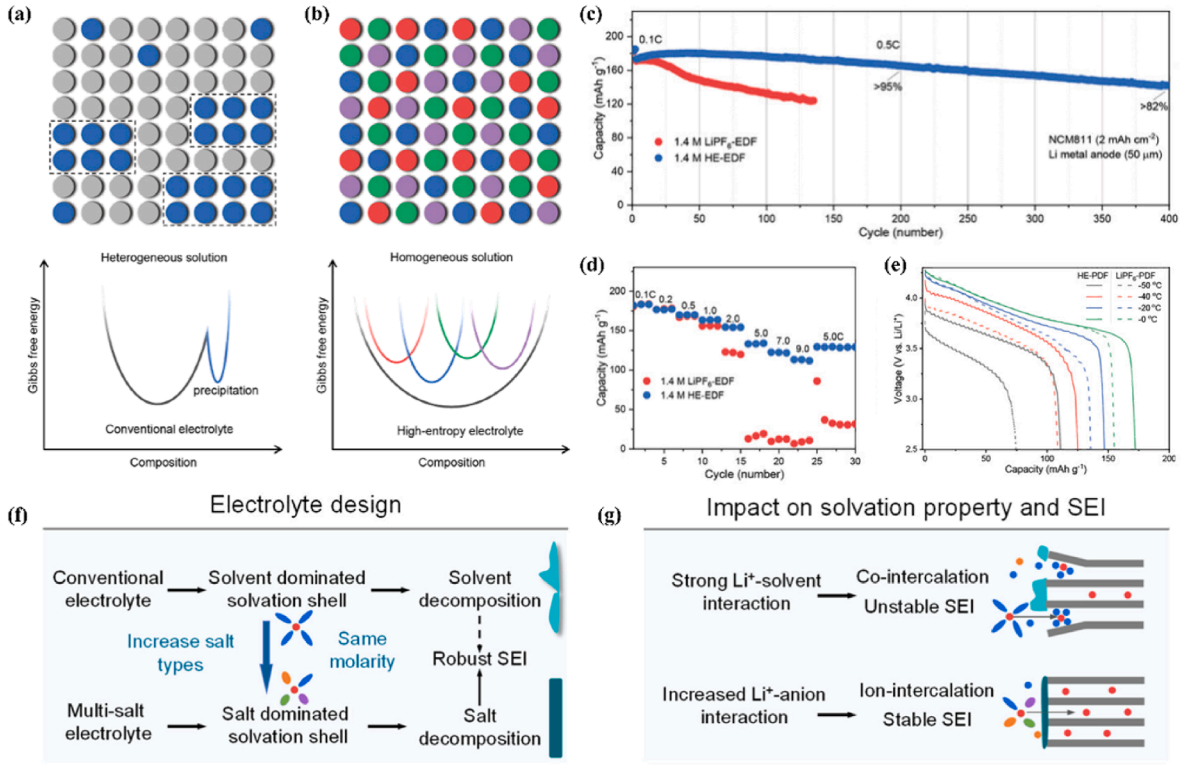


**Fig. 4.** Entropy tuning strategies in Li-ion batteries. (a) Schematic illustration of the structure and energy in the low-entropy electrolyte with indication of precipitation. (b) Schematic illustration of the structure and energy in the high-entropy electrolyte with the homogeneous distribution. (c) Cycling performance of Li||NCM811 batteries with different electrolytes. (d) Cycling performance at different current densities, exhibiting the rate performance. (e) Discharge curves at different temperatures. (a–e) Reproduced with permission from Ref. 53. (f) Schematic demonstration of SEI formation mechanisms with single-salt and multiple-salt electrolytes. (g) Influence of SEI with different electrolytes. (f–g) Reproduced with permission from Ref. 54.

the decomposition of both solvents and salts, the final film will be a mixture of organic and inorganic compositions, with organics dominating. Instead, for this anion-rich interfacial reaction process, an inorganic-dominated SEI can be formed. This type of SEI was commonly believed to be more robust because of its advantageous resistance to dissolution in organic solvents and its higher Young's Modulus. Additionally, ion transportation kinetics were also facilitated in this inorganic SEI (Fig. 4g).[55] As a result, a superior capacity retention of 94% after 600 cycles at 1C was exhibited in the NCM811||graphite battery.

Besides multiple salts, the multiple solvents strategy has also been explored. For traditional electrolytes using only one or two types of solvents, two extreme cases may happen. One is when the solvation strength is very low, the cation-solvent complex will be very large, and the ionic conductivity is reduced. On the contrary, if the solvation strength is high, even though the ions become easier to mobilize, the extremely incompact distribution will result in poor stability.[56] Once one or two solvents and the salt are chosen, the solvation strength is fixed because of the inherent binding energy. Therefore, the balance between stability and ionic conductivity is difficult to establish in the low-entropy electrolyte. To solve this issue, Kim et al. designed high-entropy electrolytes with five solvents.[57] They dissolved 1 M LiTFSI in a complicated solvent matrix consisting of dimethoxyethane (DME), 1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether (TTE), diethoxyethane (DEE), diethylene glycol dimethyl ether (DEGDME), and bis (2,2,2-trifluoroethyl) ether (BTFE). By incorporating solvents with different binding forces for Li ions, the solvation sheath was adjusted to a moderate size, allowing for the formation of small clusters. Thus, the aggregation of ions and solvents was avoided to ensure a high ionic conductivity. Meanwhile, the clusters can improve the stability. They also conducted the component-exclusion study to elucidate the necessity of each solvent. By dividing these five solvents into two clusters, which are solvents and co-solvents, they designed electrolytes with 2, 4, and 5 constituents. Among these, DME, DEGDME, and DEE are solvents because they function to dissociate ions and directly participate in the solvation sheath. On the other hand, TTE and BTFE are defined as the co-solvents, and they function to regulate the viscosity to ensure the fast ion transportation kinetics and provide an anion-rich solvation environment at high concentrations. Therefore, by combining one co-solvent with one or three solvents, they fabricated electrolytes with 2 or 4 components. By adding one additional co-solvent, they settled on the optimal high entropy electrolyte with five constituents. As a result, the ionic conductivity increased from 1.7 mS $cm^{-1}$ to 3.6 mS $cm^{-1}$ as the number of solvents increased from 2 to 5. Consequently, the anode-free Cu||NMC532 pouch cell exhibited a high capacity retention of 70% after 80 cycles at 2C.

These two design routines can also be mapped to other non-aqueous rechargeable batteries. As shown in Fig. 5a, Li et al. prepared a single solvent electrolyte containing $NaPF_6$ and $NaBF_4$ salts.[58] Yang et al. used the single $NaPF_6$ salt in binary solvents with the combination of diethylene glycol dimethyl ether (DIG), tetrahydrofuran (THF), DME, 2-methyltetrahydrofuran (mTHF), and Triglyme.[59] All these electrolytes presented a more robust electrode interface to ensure the stable operation of Na-ion batteries. Meanwhile, these electrolytes enabled operation at progressively lower temperatures (Fig. 5b and c). Even though the high-entropy electrolyte has not attracted much attention in K-ion batteries, pioneering work using multiple solvents for comprehensive K salts has shown efficacy for improving stability, especially for the performance at low temperatures.[61]

The diversification of salts and solvents has been confirmed for their efficacy, so entropy tuning through introducing both multiple salts and solvents seems promising. Wang et al. designed an electrolyte with two different cations, anions and solvents.[60] They dissolved LiOTf and Mg $(TFSI)_2$ into DME and trimethyl phosphate (TMP). As depicted in Fig. 5d, the solvation sheath ($Mg^{2+}$-2DME-$OTf^-$-$Li^+$-DME-TMP) became highly

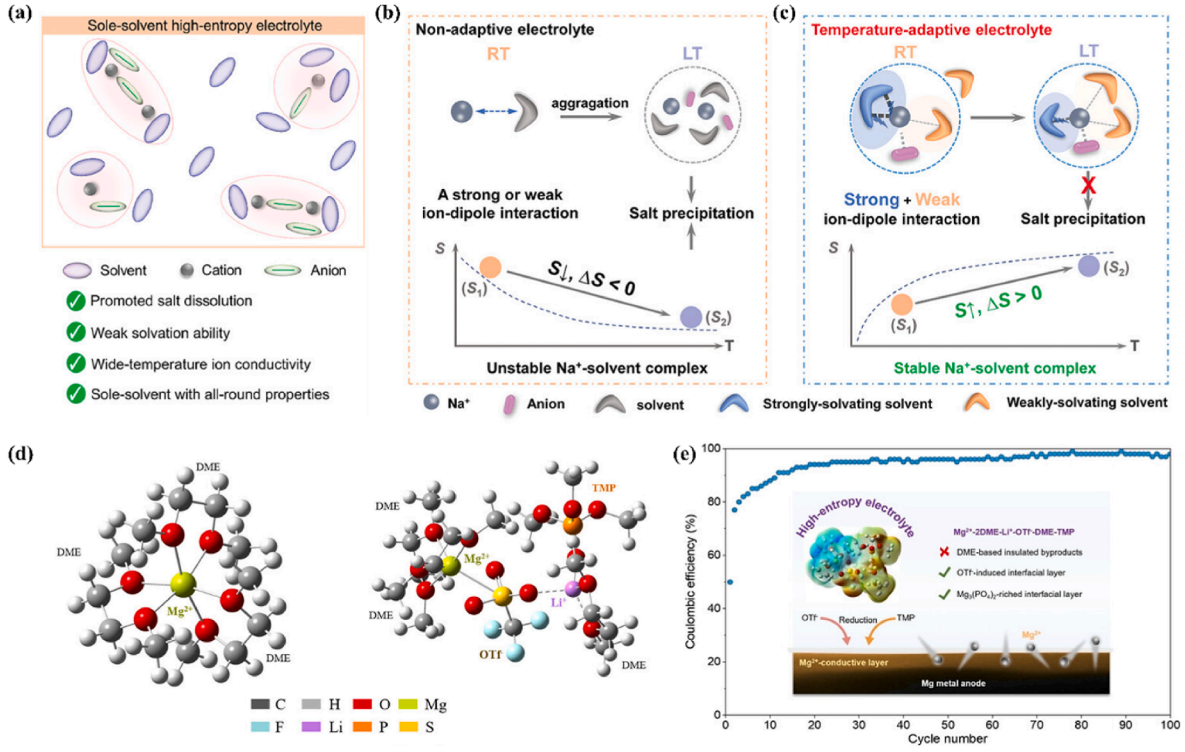


**Fig. 5.** Entropy tuning strategies in non-Li organic batteries. (a) Schematic demonstration of the single-solvent high-entropy Na-ion battery electrolyte. Reproduced with permission from Ref. 58. Schematic illustration of the variation of solvation structure once the temperature changes in (b) conventional electrolytes, and (c) high-entropy electrolytes. (b, c) Reproduced with permission from Ref. 59. (d) Solvation sheath of low-entropy and high-entropy electrolytes in Mg-ion batteries. (e) Cycling performance of the Mg-ion battery with the high entropy electrolyte, the inset shows the Mg anode interfacial reaction. (d, e) Reproduced with permission from Ref. 60.

**Table 1**
Performance comparison of different high-entropy electrolytes.

| | Electrolyte formula | Anode | Cathode | Performance summary | Ref. |
|---|---|---|---|---|---|
| **Aqueous electrolyte** | LiCl and $ZnCl_2$ in water with a molar ratio of 2:1 | Zn | $Zn_xVOPO_4{\cdot}2H_2O$ | Capacity retention of 90.0% and 81.1% after 200 cycles at −70 °C and −80 °C | 43 |
| | $Zn(ClO_4)_2$, $NaClO_4$, $LiClO_4$, and $H_2O$ were mixed with a molar ratio of 0.2:0.4:0.4:7 | Zn | NaVO | No capacity decay after 1500 cycles at 1 A $g^{-1}$ | 44 |
| | | Zn | PANi | No capacity decay after 1500 cycles at 1 A $g^{-1}$ | 44 |
| | $ZnSO_4$, $Zn(Ac)_2$, $Zn(OTf)_2$, $Zn(ClO_4)_2$ and $Zn(BF_4)_2$ dissolved into water with a concentration of 50 mM | Zn | PANi | Capacity retention of 70.4% at 15 A $g^{-1}$ after 10,000 cycles | 45 |
| | Different Zn salts in EG/PG/glycerol/NMA/SN/glyme/diglyme with designated salts, organics, and water molecules at a molar ratio of 1.5:4:24 | Zn | NVO | Capacity retention of 80% at 0.5C after 400 cycles in the pouch cell at −40 °C | 46 |
| | 4.5 M LiTFSI, 0.1 M KOH, and 0.1 M $CO(NH_2)_2$ in water | $Li_4Ti_5O_{12}$ | $LiMn_2O_4$ | Capacity retention of 92% after 470 cycles at an areal capacity of 2.5 mAh $cm^{-2}$ | 47 |
| **Non-aqueous electrolyte** | 0.1 M LiFSI, 0.1 M LiDFOB, 0.1 M LiTFSI, 0.1 M $LiNO_3$ in the commercial 1.0 $LiPF_6$ EC/DMC (1:1 in volume) electrolyte with 5% FEC | Li | NCM811 | Capacity retention of 82% after 400 cycles at 0.5C | 53 |
| | | Graphite | NCM811 | Capacity retention of 90% after 1000 cycles at 2.0C | 53 |
| | 0.2 M LiFSI, 0.2 M $LiPF_6$, 0.2 M LiDFOB, 0.2 M LiTFSI, 0.2 M $LiNO_3$ in PC with 5% FEC | Graphite | NCM811 | Capacity retention of 94.0% after 600 cycles at 1.0C | 54 |
| | | Si/Graphite | NCM811 | Capacity retention of 94.5% after 300 cycles at 1.0C | 54 |
| | 1 M LiFSI in DME, DEE, DEGDME, TTE, and BTFE mixture (volume ratio = 5:2.5:2.5:45:45) | Cu (anode free) | NCM532 | Capacity retention of 70% after 110 cycles at 0.2C charging and 0.5C discharging | 57 |
| | | Cu (anode free) | NCM532 | Capacity retention of 70% after 80 cycles at 2C in the pouch cell | 57 |
| | 1.2 M $NaFP_6$ and 0.6 mM $NaBF_4$ in D2 | Al/C (anode free) | NFM | Capacity retention of 83% after 100 cycles in the pouch cell | 58 |
| | 0.5 M LiDFOB, 0.2 M $LiPO_2F_2$, and 0.3 M $LiPF_6$ in EMC and DMC (volume ratio = 7:3) | Li | NCM811 | Capacity retention of 80% after 500 cycles | 62 |
| | 0.15 M LiFSI, 0.10 M LiTFSI, 0.10 M LiBFTI, 0.10 M LiDFOB and 0.15 M $LiNO_3$ in DME | Li | NCM811 | Capacity retention of 80% after 600 cycles at 6C | 63 |
| | 1 M $NaPF_6$ into DIG: THF (DME: THF) binary solvents with a volume ratio of 2:8. | Hard carbon | NNCM | Capacity retention of 90.6% over 400 cycles at −40 °C | 59 |
| | 0.2 M $Mg(TFSI)_2$ in DME mixed with 1 M LiOTF in TMP with a volume ratio of 9:1 | Mg | $Mo_6S_8$ | Capacity retention of 98.4% after 200 cycles | 60 |
| | 2 M KFSI in TMP/TEP-TTE-HFME-OTE | K | KFeHCF | Capacity retention of 87.5% after 2000 cycles at a current density of 0.5 A $g^{-1}$ | 61 |

chaotic. As a result, the desolvation kinetics were boosted, and the electrode stability was improved. The resulting superior battery performance provided strong proof of the effectiveness of the entropy tuning strategy in chlorine-free magnesium metal batteries (Fig. 5e).

In summary, entropy tuning in non-aqueous electrolytes can be achieved through introducing diverse solvents, anions, or cations. Similar to aqueous electrolytes, the stability of batteries can be effectively improved (Table 1). Furthermore, the solvation structure is also modified. As a result, suitable solvation clusters can be achieved to reach a high ionic conductivity, and the SEI formation process can also be tuned due to the altered interfacial reactants. Meanwhile, the low-temperature performance can also be improved. Besides the full crystallization of solvents, which is commonly considered in aqueous electrolytes, the mitigation of salt precipitation is the main reason for low-temperature performance optimization. However, because of the much lower inherent freezing point of organic solvents, the bottom line of working temperature in non-aqueous high-entropy electrolytes is very low, which may not match any possible applications. Therefore, the trade-off between the cost of the electrolyte formula and maximum performance is necessary to make the non-aqueous high-entropy electrolytes more practical.

## 5. Conclusion

As one of the most versatile energy storage devices, rechargeable batteries require continuous improvements in stability and energy efficiency. Thus, many efforts have been made to design advanced electrolytes, which may solve issues for both electrodes. Among diverse electrolyte design strategies, entropy tuning has shown potential. The entropy can be elevated by introducing more components, including solvents, salts, and additives, into the electrolytes. Especially for liquid electrolytes, which exhibit inherent disorderliness, entropy tuning can be easily achieved. Based on the solvent type, liquid electrolytes are divided into aqueous and non-aqueous electrolytes. For aqueous electrolytes, entropy can be increased by adding multiple salts, solvents, and additives. Due to their different binding forces, the free water can be reduced, and the H-bond network is regulated. As a result, the fast ion transportation kinetics can be ensured with a suppression of water-induced parasitic reactions and a decrease in the freezing point. For non-aqueous electrolytes, entropy tuning can be achieved through similar strategies. Besides the advantages mentioned above, diverse components in non-aqueous electrolytes can also be employed to optimize the formation of the SEI layer. However, multiple challenges still remain for the practical application of high-entropy electrolytes. The first one is that the complicated components make the synthesis and characterization of the electrolytes extremely challenging. Meanwhile, the exploration of their individual function is also difficult. Furthermore, computational methods for high-entropy electrolytes are also complex. Unlike computations for traditional electrolytes, which involve three or four distinct molecules, the diverse components make the modeling and computing process more complicated. Secondly, the homogeneity of electrolytes cannot be ensured. For organic electrolytes, different organic solvents may cause microphase separation because of their compatibility. This may cause the inhomogeneous distribution of electrolytes on electrodes. This issue may be more pronounced for aqueous electrolytes. Many aqueous high-entropy electrolytes introduce organic

contents, which show poor compatibility with water. As a result, water-in-oil emulsions may form. Despite reported successful cases, incompatible contents can damage the solubility or ionic conductivity. Thirdly, high-entropy electrolytes may sacrifice some merits inherent in traditional electrolytes. For both aqueous and organic electrolytes, high-entropy tuning may result in a rise in cost. Furthermore, for aqueous ones, a large amount of organic co-solvents may make the electrolyte flammable. Herein, based on these challenges, we proposed several perspectives about the future standardization of high-entropy liquid electrolytes design.

1. Machine learning can be employed for filtering the proper compositions of high-entropy electrolytes. The experimental evaluation for high-entropy electrolytes is inconvenient and complicated due to their diverse constituents. However, some theoretical characteristics, like the coordination number or electronegativity, can be obtained through modeling and calculations. Thus, machine learning can efficiently process these properties to identify suitable combinations. This also consolidates the logic for designing the high-entropy electrolyte.
2. Compatibility between high-entropy electrolytes and current battery production lines is crucial for overcoming their practical application bottlenecks. Even though the synthesis of a high-entropy electrolyte is straightforward, the mass production still requires careful consideration of the capital cost related to the production line adjustment. If the order of adding different constituents is critical, it will be difficult to just supply the pre-mixed materials in the current manufacturing line. As a result, the cost for supplementing parallel beltlines will further damage the cost-effectiveness of high-entropy electrolytes. Meanwhile, if additional compositions make the storage or transportation of high-entropy electrolytes more challenging, the practical value of such a formula is also undermined.
3. One specific successful formula of a high-entropy electrolyte cannot be viewed as an improvement on the entropy tuning strategy. For one high-entropy electrolyte, the substitution of each component is necessary to confirm the proposed mechanism. For example, if multiple solvents are chosen depending on their molecular structure, other solvents with similar structures should also be tested to ensure that the synergetic improvement comes from integration of structures. Meanwhile, a thorough study with the constituent part subtraction is also necessary to determine the necessity of each component. Both studies can be combined with machine learning to advance efficiency.

Overall, the ongoing development of rechargeable batteries puts a higher demand on their stability and energy efficiency. The electrolyte engineering has been confirmed effective for both key characteristics. Meanwhile, by introducing diverse salts, solvents, and additives, the high-entropy liquid electrolytes seem a promising and convenient solution among the electrolyte engineering strategies. However, they are still far from the practical application unless a systematic sorting of components can be established and the cost performance can be carefully evaluated. In this review, we summarized recent research on high-entropy liquid electrolytes and analyzed their design strategies and underlying mechanisms. We sincerely hope that this review can provide some insights into the future design of high-entropy electrolytes and motivate their advancement to industrial applications.

## CRediT authorship contribution statement

**Mingcong Tang:** Writing – original draft, Conceptualization. **Xiaohong Zou:** Writing – review & editing, Investigation. **Lizhen Wu:** Writing – review & editing, Investigation. **Gang Liu:** Writing – review & editing. **Rong Chen:** Writing – review & editing. **Liang An:** Writing – review & editing, Supervision, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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