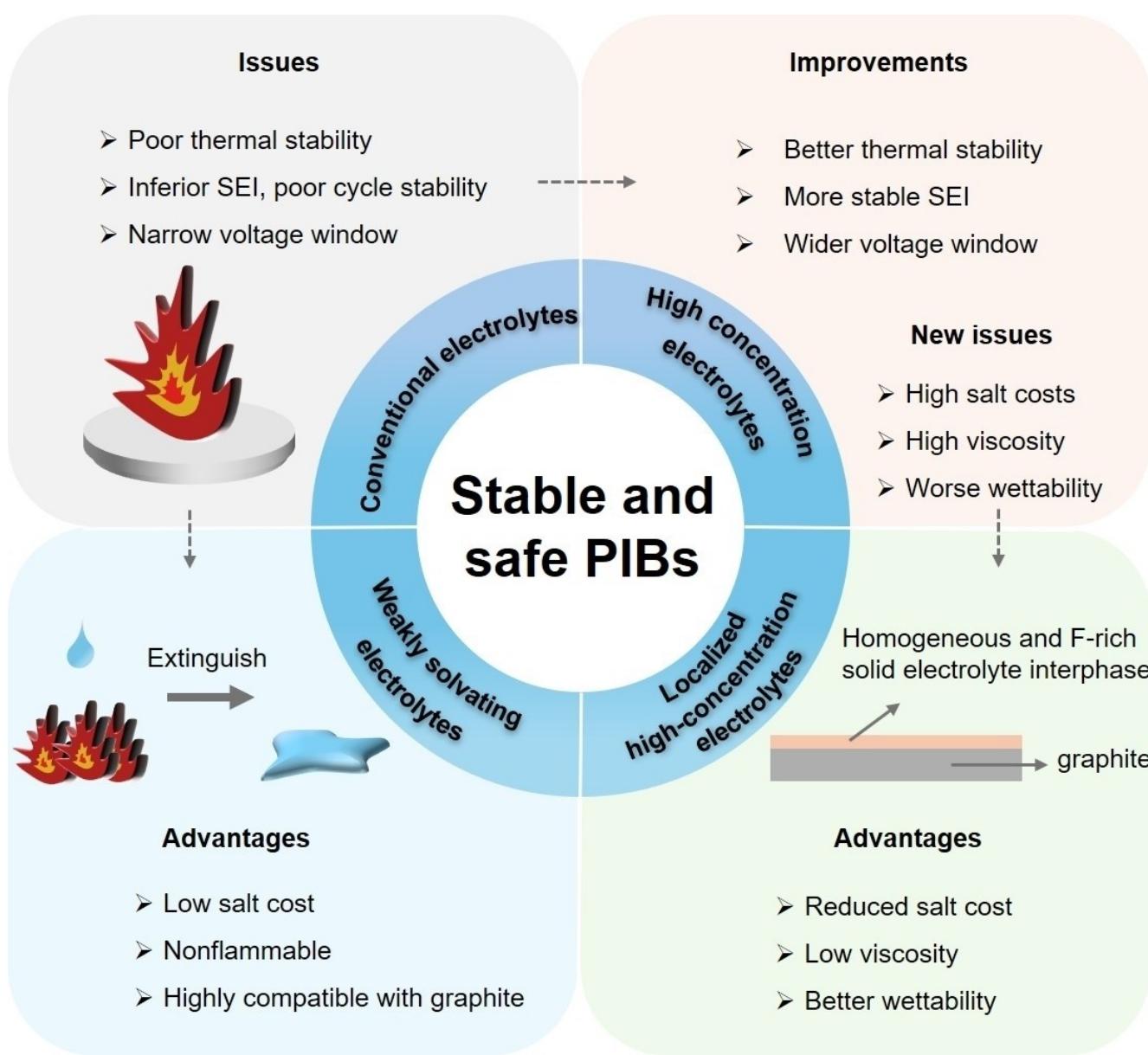


# Organic Electrolytes for Stable and Safe Potassium-Ion Batteries

Shu Xu,<sup>[a]</sup> Xianhui Yi,<sup>[a]</sup> Ling Fan,<sup>\*[a]</sup> and Bingan Lu<sup>\*[a]</sup>



Though lithium-ion batteries (LIBs) are prevalent, the scarcity and uneven distribution of lithium resources have driven the search for complementary battery technologies. Potassium-ion batteries (PIBs) have emerged as a promising contender in this quest due to their low-cost, abundant resources, and potentially high voltage. The frequency of battery-related accidents in recent years has heightened concerns about battery safety. The electrolyte, as a critical component of batteries, plays a pivotal role in determining safety, as it is directly linked to the

formation of the solid electrolyte interphase (SEI), which is crucial for battery stability and security. Initially, enhancing battery safety often came at the cost of performance. However, thanks to the relentless efforts and in-depth research, it is now possible to improve performance without compromising safety. This article provides an overview of organic electrolyte systems and introduces some state-of-the-art electrolyte formulations, aiming to offer guidance on enhancing the safety and stability of PIBs.

## 1. Introduction

The main theme of global development is progress, and energy is one of the key drivers of this development.<sup>[1]</sup> Currently, the world relies heavily on fossil fuels. However, as these resources deplete and the greenhouse effect intensifies, there is an urgent need to identify sustainable energy sources.<sup>[2]</sup> In this context, people have discovered clean energy sources such as solar and wind power, which although sustainable, are often intermittent.<sup>[3]</sup>

Consequently, energy storage devices are indispensable to social development, as they allow us to store energy for use during periods when renewable sources are not producing power.<sup>[4]</sup> Rechargeable lithium-ion batteries (LIBs) are widely used in portable devices like laptops and mobile phones and, due to their long cycle life, high power, and low maintenance costs, have even been applied in transportation and aircraft in recent years.<sup>[5]</sup>

Although lithium-ion batteries have been very successful, their low crustal abundance and uneven distribution may limit their use in future large-scale energy storage applications.<sup>[6]</sup> To address these challenges, researchers are continuously exploring lower-cost alkali metal ion batteries that have the potential to replace lithium-ion batteries in some applications.<sup>[7]</sup> These include sodium-ion batteries (SIBs) and potassium-ion batteries (PIBs), which researchers believe may offer viable alternatives for the next generation of energy storage solutions.<sup>[8]</sup>

SIBs and PIBs significantly outpace LIBs in abundance within the Earth's crust, with sodium at 2.36 wt% and potassium at 2.09 wt% dwarfing lithium's 0.0017 wt%.<sup>[9]</sup> Among these, PIBs are especially promising. The standard electrode potential for K<sup>+</sup>/K ( $-2.93\text{ V}$ ) is lower than that of Na<sup>+</sup>/Na ( $-2.71\text{ V}$ ) and closely aligns with the established Li<sup>+</sup>/Li potential ( $-3.04\text{ V}$ ), offering the PIBs high voltage ability.<sup>[10]</sup> Potassium's weaker Lewis acidity also results in a smaller solvated ion radius, which enhances ionic conductivity and migration velocity.<sup>[11]</sup> This, in turn, significantly improves the performance of PIBs. The ability of potassium ions to form intercalation compounds within graphite, with a theoretical capacity of approximately 279 mAh g<sup>-1</sup>, highlights PIBs' commercial potential.<sup>[12]</sup> Given

these compelling advantages, PIBs are strong contenders as the next generation of energy storage solutions, complement for LIBs, as shown in Figure 1.<sup>[13]</sup>

The advancement of battery technology is consistently marked by critical safety considerations. Battery safety issues can be attributed to several key factors. Primarily, batteries are susceptible to overcharging or over-discharging during routine operation, which can result in a rise in internal temperature, initiating exothermic reactions, and culminating in thermal runaway.<sup>[14]</sup> Additionally, under conditions of elevated temperature and voltage, electrochemical reactions become increasingly intricate, encompassing the release of oxygen from the cathode and parasitic side reactions between the electrolyte and electrodes.<sup>[15]</sup> These complex reactions can ultimately precipitate thermal runaway in the battery, leading to rupture and potential explosion. Lastly, the rupture of separators and oxygen evolution at the cathode contribute to thermal runaway, thereby posing significant safety hazards.<sup>[16]</sup> The continuous occurrence of LIBs safety accidents makes the safety performance of batteries face challenges.<sup>[17]</sup> PIBs, similar to lithium-ion batteries, face significant safety challenges. The electrolyte, a vital component of these systems, is crucial for maintaining operational safety and integrity.<sup>[18]</sup> This is because the electrolyte serves as the "bridge" connecting the positive and negative electrodes. A good electrolyte can make the battery more stable by suppressing dendrites and regulating the formation of the solid electrolyte interphase (SEI).<sup>[19]</sup> Choosing non-flammable electrolytes can also greatly ensure the safety of the battery.

Conventional electrolytes have encountered many problems, and with the continuous development of electrolytes, there are also certain solutions, including high concentration electrolytes, and weakly solvating electrolytes with phosphate ester solvents.<sup>[20]</sup> As research into PIBs deepens, the development of safer and more reliable electrolytes has become a prominent focus, the timeline of PIBs electrolytes development is shown in Figure 2. For example, in 2017, Xiao *et al.*<sup>[21]</sup> reported a high concentration potassium bis(fluorosulfonyl)imide (KFSI) - 1,2-dimethoxyethane (DME) electrolyte, which can form a uniform SEI on the surface of potassium metal and endows potassium plating/stripping with high efficiency of ~99%. In 2020, Liu *et al.*<sup>[22]</sup> reported a 1 M KFSI in trimethyl phosphate (TMP) and ethylene sulfate (DTD), this electrolyte can compatible with the graphite anode at a low concentration, and the capacity hardly decays after 100 cycles. In 2023, Fan *et al.*<sup>[23]</sup> reported a non-flammable weakly solvating electrolyte that

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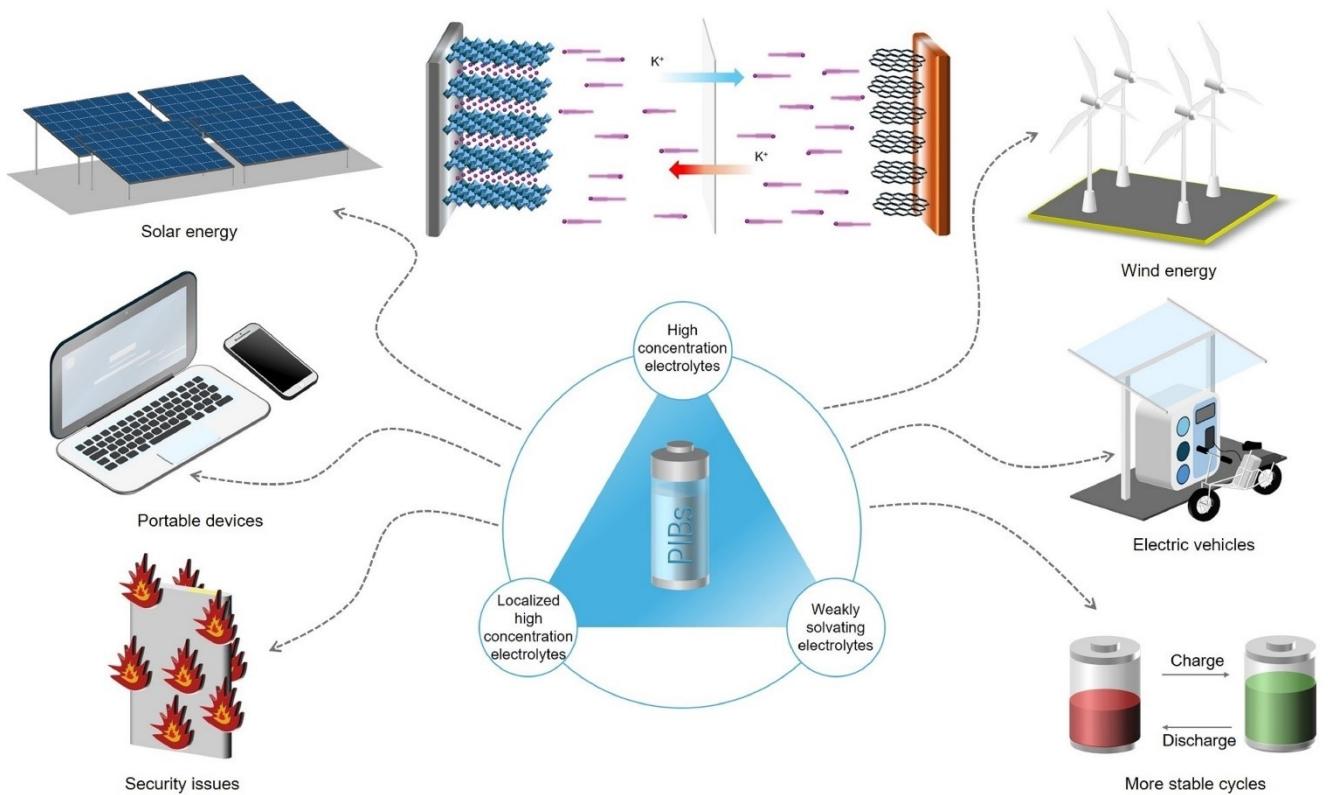


Figure 1. Applications and challenges of potassium-ion batteries.

enables the graphite electrode to operate stably for more than 2 years. This review meticulously examines recent advances and cutting-edge electrolyte formulations.<sup>[24]</sup> It highlights innovations that are setting new standards for the safety and performance of potassium-ion batteries, reflecting a concerted effort to advance the field of energy storage technology.<sup>[25]</sup>

## 2. High Concentration of Electrolytes

Due to the limitations of ionic conductivity, the concentration of electrolytes is generally maintained around 1 M.<sup>[26]</sup> However, traditional electrolytes have several unresolved issues. With the continuous increase in salt concentration, some of these problems have been effectively mitigated, drawing attention to



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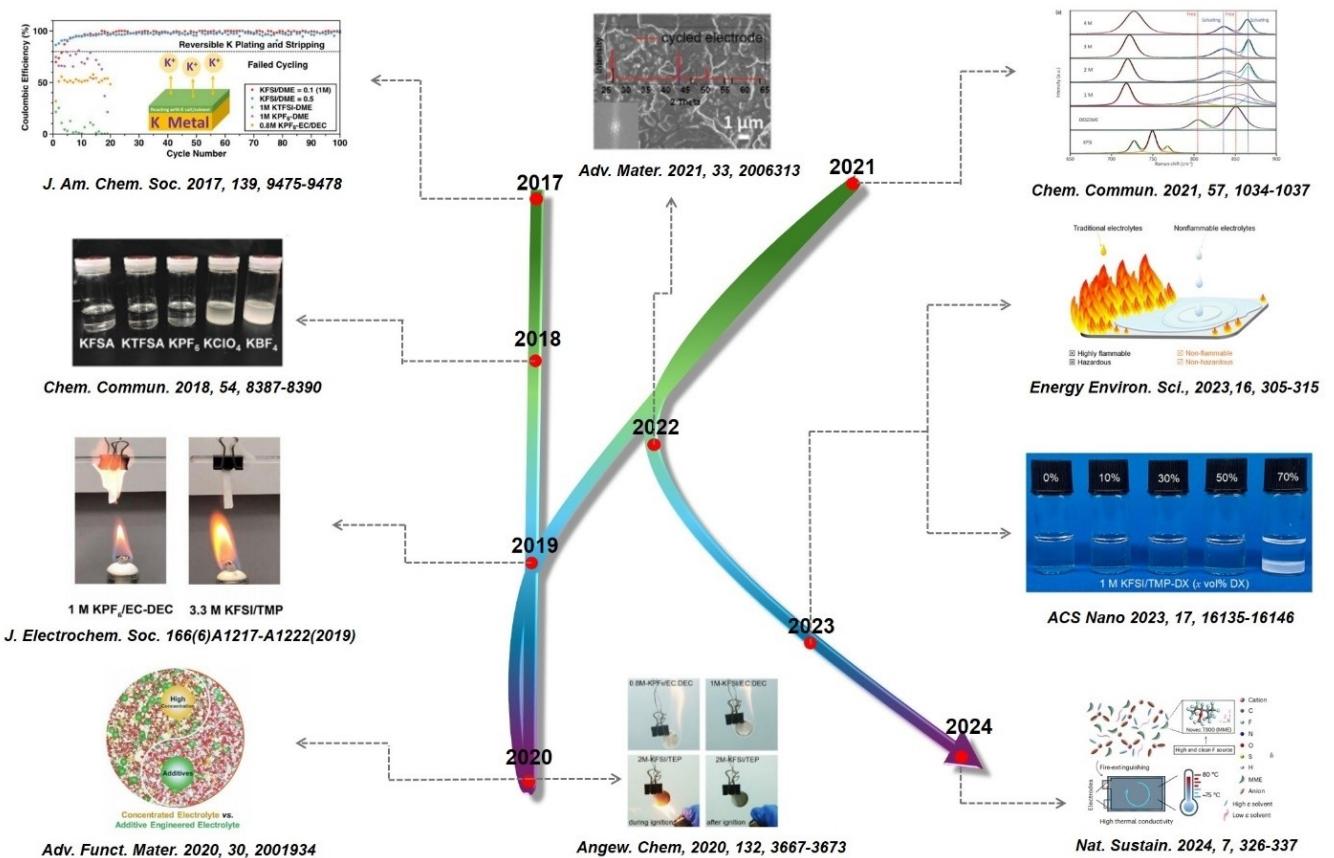


Figure 2. Timeline of electrolyte development for potassium-ion batteries.

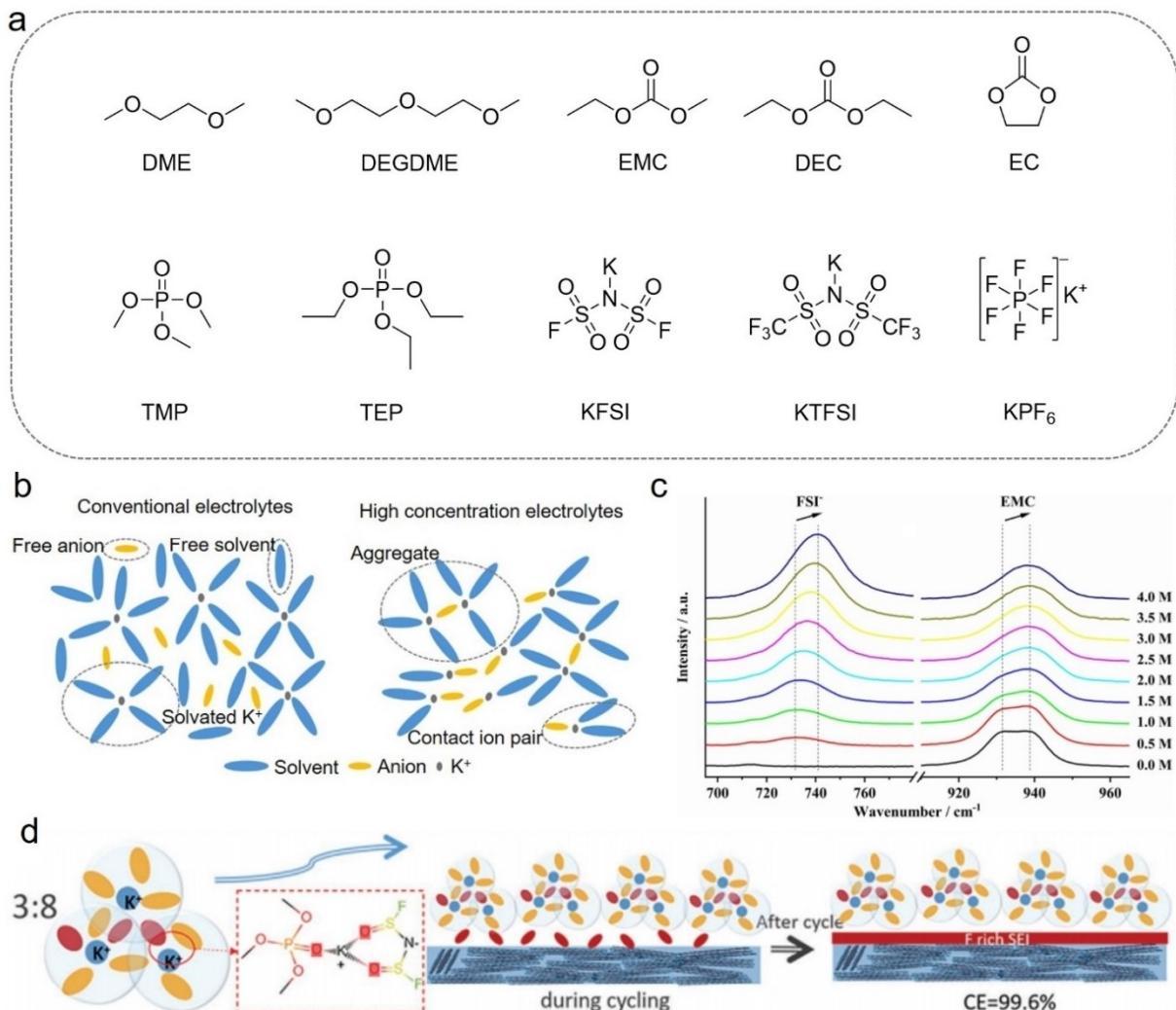
high-concentration electrolytes (HCEs).<sup>[27]</sup> The solvents and salts commonly used in high-concentration electrolytes are shown in Figure 3a. The essence of HCEs lies in their unique solvation environment, which reduces the number of free solvent molecules and enhances cation-anion interactions.<sup>[28]</sup> In conventional electrolytes, potassium ions form solvation shells with solvent molecules, which undergo complex changes during battery charging and discharging.<sup>[29]</sup> As shown in Figure 3b, as the salt concentration increases, the number of free solvent molecules in the electrolyte decreases, resulting in the formation of aggregates and contact ion pairs.<sup>[30]</sup> This transition significantly alters the physicochemical properties of the electrolyte, including improved thermal stability, expanded electrochemical windows, reduced corrosion of aluminum current collectors and alleviative dendrite growth.<sup>[31]</sup>

In PIBs, the uncontrollable growth of K dendrites in commercial potassium hexafluorophosphate (KPF<sub>6</sub>) electrolytes results in poor battery cycle performance, low coulombic efficiency, and even short circuits caused by dendrites piercing the separator, posing safety hazards.<sup>[32]</sup> In contrast, HCEs, enable more uniform deposition/stripping of K, thus suppressing dendrite formation and achieving more stable cycle performance.<sup>[33]</sup> Compared with conventional electrolytes, HCEs conducive to forming anion-derived electrode-electrolyte interphase due to the reduced number of free solvent molecules and enhanced cation-anion interactions, thereby improving the

oxidative stability of the battery.<sup>[34]</sup> Thus, the HCEs are conducive to widening the voltage window, allowing the battery to have a higher energy density.

Safety is also a critical indicator for batteries. In HCEs, the high concentration of salt significantly reduces the number of free solvent molecules.<sup>[35]</sup> These free solvent molecules in lower-concentration electrolytes may promote the volatility and decomposition of the electrolyte, but this impact is greatly reduced in HCEs.<sup>[36]</sup> Meanwhile, the anion-derived SEI has good stability and can effectively suppress side reactions at the interface, thus inhibiting dendrite growth and effectively preventing short circuit situations.<sup>[37]</sup> Therefore, HCEs have unique advantages in terms of safety and cycling stability compared to low concentration electrolytes.

In order to investigate the impact of high-concentration electrolytes on the performance of potassium-ion batteries, Hosaka *et al.*<sup>[38]</sup> designed an electrolyte solution with 7 mol kg<sup>-1</sup> KFSI in DME. This electrolyte solution exhibits a wider potential window (0–4.3 V vs. K<sup>+</sup>/K) than the traditional KPF<sub>6</sub>/carbonate ester electrolyte and does not cause aluminum corrosion. The graphite || K<sub>2</sub>Mn[Fe(CN)<sub>6</sub>] full-cell pretreated in the high-concentration KFSI/DME electrolyte shows reversible charge and discharge behavior, with a capacity retention rate of over 85% after 101 cycles and an average coulombic efficiency of 99.3%, demonstrating the superiority of HCEs. Münster *et al.*<sup>[39]</sup> explored the effect of high-concentration electrolyte of 3.5 M



**Figure 3.** High concentration electrolytes. a) Molecular structure of commonly used solvents and salts in electrolytes. b) Comparison of the solvation structure of conventional electrolytes and high-concentration electrolytes. c) Raman spectra in KFSI/EMC electrolytes as a function of salt concentration. Reprinted with permission from ref.39. Copyright 2019 The Authors. Published by Wiley-VCH Verlag GmbH & Co. KGaA. d) F-rich SEI layer generated by a high concentration electrolyte of KFSI:TMP with a molar ratio of 3:8. Reprinted with permission from ref.42b. Copyright 2020 Wiley-VCH GmbH.

KFSI in ethyl methyl carbonate (EMC) on the performance of graphite electrode. The linear sweep voltammetry (LSV) revealed that the high-concentration electrolyte had better oxidative stability compared to the low-concentration counterpart. Subsequently, the coulombic current (CC) test revealed that the dissolution of aluminum current collectors was significantly reduced in the high-concentration electrolyte. They also analyzed the solvation structures at high salt concentrations using Raman spectroscopy as shown in Figure 3c. Moreover, in a 3.5 M KFSI/EMC electrolyte, a potassium-ion dual-graphite battery (DGB) demonstrated good charge-discharge cycling, with a capacity retention of 76% after 200 cycles and a coulombic efficiency between 90–95%. Xu *et al.*<sup>[40]</sup> developed a high-performance electrolyte consisting of 4 M KFSI in diethylene glycol dimethyl ether (DEGDME). In this electrolyte, K||Cu coin cells can achieve stable cycling with a coulombic efficiency of 98% and remain stable over 400 cycles. Moreover, the K||Prussian blue (PB) cells demonstrated a good

capacity retention of 99.7% after 500 cycles. X-ray photoelectron spectroscopy (XPS) analysis revealed that this electrolyte can form a highly stable SEI on the potassium metal surface, which is composed of inorganic compounds derived from the decomposition of FSI<sup>-</sup> and oligomers resulting from the decomposition of DEGDME.

Although ether-based and carbonate ester-based HCEs can effectively improve thermal stability, there is still a risk of flammability. Using inherently non-flammable or flame-retardant solvents as electrolytes is an economical way to address this issue. Low molecular weight phosphate esters have similar physicochemical properties to carbonate esters and have broad application prospects. However, at low concentrations, phosphate esters have difficulty forming a stable SEI film on the graphite surface. The HCE system solves this problem, making the use of non-flammable solvents as electrolytes possible.

In order to develop a safer electrolyte that is also compatible with graphite anodes, Zeng *et al.*<sup>[41]</sup> integrated

results from tests such as ionic conductivity, flammability, and X-ray diffraction (XRD) to ultimately select an electrolyte composed of 3.3 M KFSI and TMP. Compared to the low-concentration KFSI/TMP electrolyte, this electrolyte demonstrated excellent compatibility with the graphite anode. At a current density of  $60 \text{ mA g}^{-1}$ , the graphite electrode maintained a reversible capacity of  $214 \text{ mAh g}^{-1}$  after 80 cycles with a coulombic efficiency of 99.6%. In this electrolyte, the Prussian blue cathode also showed an initial coulombic efficiency of 84% and a reversible capacity of  $46 \text{ mAh g}^{-1}$  with a capacity retention rate of 95%.

Liu *et al.*<sup>[42]</sup> developed an electrolyte composed of 2 M KFSI in triethyl phosphate (TEP) by considering ionic conductivity, viscosity, and cost. In this electrolyte, the potassium metal anode achieved stable potassium plating/stripping for over 500 cycles with a coulombic efficiency of 99.6%. Additionally, the graphite electrode demonstrated excellent cycling performance in this electrolyte, showing a reversible capacity of  $275 \text{ mAh g}^{-1}$  at a current density of 0.2 C, and maintaining a capacity retention of 90% after 500 cycles. Using this electrolyte, a full cell with perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) as the cathode and pre-potassiated graphite as the anode also exhibited good cycling performance, which provides a reversible charge capacity of  $127 \text{ mAh g}^{-1}$  and maintaining a coulombic efficiency of 96.6% over 50 cycles. In another work of them,<sup>[42]</sup> an electrolyte composed of flame retardant TMP and KFSI with a salt-to-solvent molar ratio of 3:8 showed exceptional cycling stability, maintaining a capacity retention of 74% after 2000 cycles (two years), superior to the 0.8 M  $\text{KPF}_6$ /ethylene carbonate (EC): diethyl carbonate (DEC) and 1 M KFSI/EC: DEC electrolytes. A full cell with PTCDA as the cathode and graphite as the anode demonstrated a specific capacity of  $75 \text{ mAh g}^{-1}$  in the 3:8 (KFSI: TMP) electrolyte and stably cycled for 200 cycles. The excellent performance is benefit from the formation of a F-rich SEI (Figure 3d) on the electrode, which is highly related to the unique solvation structures that rich in aggregates in HCEs.

In general, HCEs offer significant benefits such as the suppression of potassium dendrite formation, which is crucial for safety, the enhancement of battery cycling stability, and the improvement of electrolyte oxidative stability. Additionally, HCEs exhibit superior thermal stability and an expanded electrochemical window, which are desirable for high-performance battery systems. Despite these advantages, HCEs encounter several challenges that impede their broader adoption. Notably, the high salt concentration leads to increased economic costs, while the high viscosity of these electrolytes results in reduced ion mobility, lower conductivity, and compromised membrane wettability. These factors collectively limit the advancement and practical application of HCEs in the field of potassium-ion batteries.

### 3. Localized High-Concentration Electrolytes

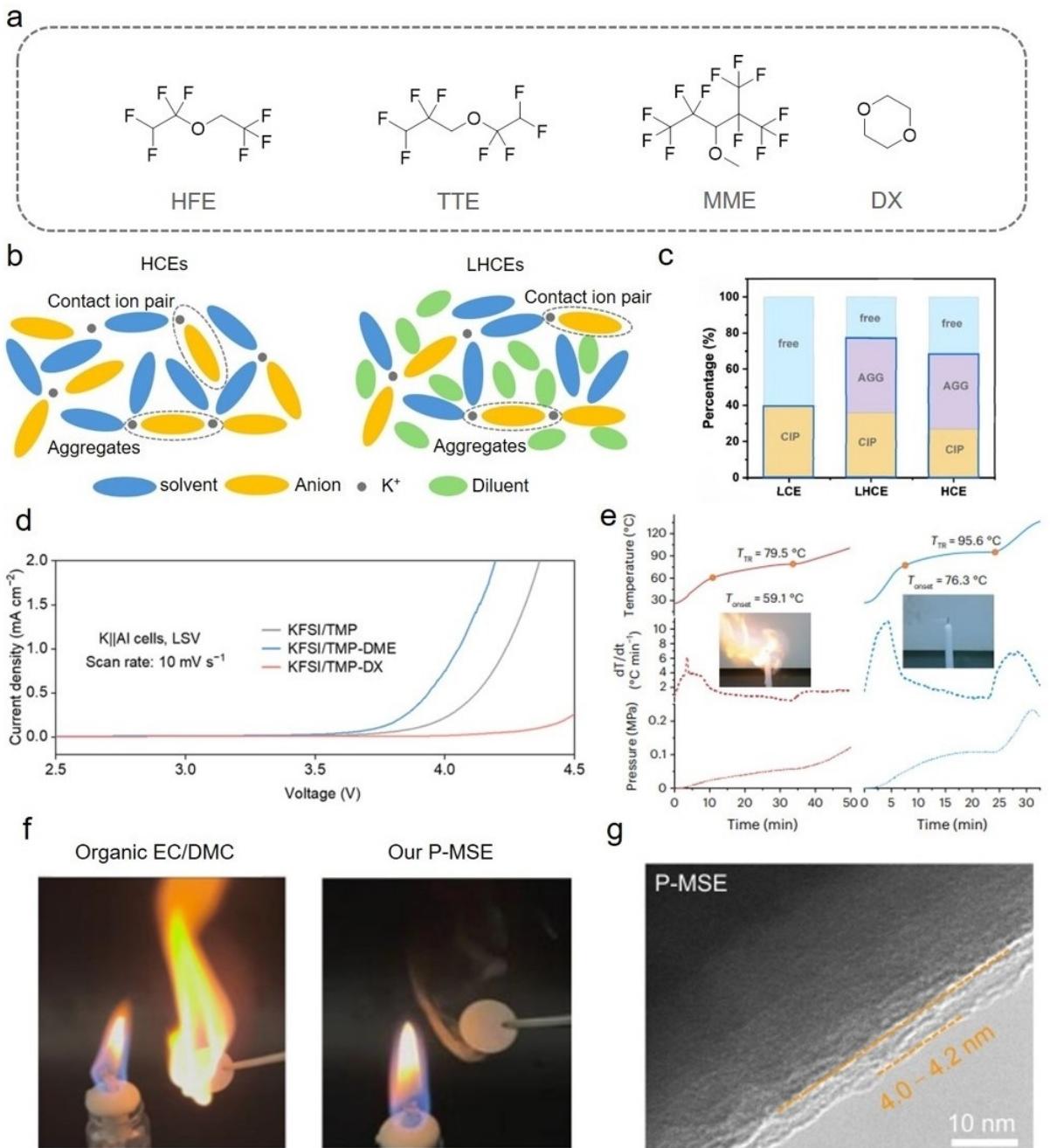
Although HCEs have significant advantages and broader application prospects compared to traditional electrolytes, they

still face many challenges.<sup>[43]</sup> Firstly, the substantial increase in salt concentration leads to economic burdens.<sup>[44]</sup> For example, the price of KFSI salt is much higher than the electrolyte solvents. This makes the cost of the electrolyte in the HCEs system greatly increased, hindering its large-scale development. Secondly, the high concentration of salt also results in increased viscosity of the electrolyte, which reduces the ionic conductivity and decreases the wetting degree of the separator. This can lead to "dry areas" during the cycling process, shortening the battery life.<sup>[45]</sup>

To address these issues in the HCEs system, scientists have proposed a new local high-concentration electrolyte system (LHCEs).<sup>[46]</sup> The diluents commonly used in localized high-concentration electrolyte are shown in Figure 4a. The so-called LHCE is obtained by adding an "inert" diluent added to the HCEs system.<sup>[47]</sup> This diluent should be miscible with the solvent but unable to dissolve the salt in the HCEs, ensuring that the unique solvation environment of the original HCEs is not affected while greatly reducing the viscosity and the amount of salt in the unit electrolyte.<sup>[48]</sup> It can be understood as a mixture of HCEs and diluent. In the LHCEs system, since the unique solvation environment of HCEs is retained, as shown in Figure 4b and c, after the addition of the diluent, this system not only retains the unique advantages of the HCEs system but also significantly reduces costs and viscosity, making the LHCE have broad development prospects.<sup>[49]</sup>

In the selection of diluents, there are also some clear principles. Firstly, it should be compatible with the original solvent and unable to dissolve the salt in it.<sup>[50]</sup> Secondly, to ensure that the voltage window of the electrolyte does not narrow after the addition of diluent, the selected additives should have a similar or wider voltage window than the original electrolyte.<sup>[51]</sup> Thirdly, to reduce the viscosity of the original HCEs, the diluent should have lower viscosity and better wettability.<sup>[52]</sup> Then, since the purpose of adding the diluent is to reduce the cost of using salt, a low-cost diluent is more preferred.<sup>[53]</sup> Lastly, to ensure that a stable F-rich SEI can still be formed, the choice of diluent should also consider whether it helps in the formation of a stable SEI.<sup>[54]</sup> In terms of safety, considering the LHCE greatly retains the characteristics of HCEs, and non-flammable solvents can also be chosen as diluents, it not only alleviates the dilemma of high viscosity and high cost of HCEs but also provides more safety guarantees.<sup>[55]</sup> For inherently non-flammable phosphate ester-based high-concentration electrolytes, the LHCEs system not only solves the problem of incompatibility with graphite at low concentrations but also reduces the original cost, giving this type of electrolyte a broader development prospect.<sup>[56]</sup> Therefore, the LHCEs system also has unique advantages in terms of safety and stability.

In order to further improve the compatibility of phosphate-based electrolytes with graphite, reduce the cost of electrolytes, and improve the viscosity of electrolytes, Liang *et al.*<sup>[57]</sup> dissolved 1.13 mol  $\text{kg}^{-1}$  of KFSI in a mixed solvent of TEP and 1,1,2,2-tetrafluoroethyl 2,2,3,3-tetrafluoropropyl ether (TTE) with a volume ratio of TEP to TTE of 1:1 as the electrolyte for potassium-ion batteries. It showed excellent cycling perform-



**Figure 4.** Localized high-concentration electrolyte. a) The molecular structures of commonly used diluents. b) Comparison of solvation structure of high-concentration electrolyte and local high-concentration electrolyte. c) Comparison of ionic speciation present in LHCE and HCE. Reprinted with permission from ref.62. Copyright 2023 Elsevier B.V. d) Comparison of thermal stability of KFSI/TMP, KFSI/TMP-DME and KFSI/TMP-DX. Reprinted with permission from ref.59. Copyright 2023 American Chemical Society. e) Electrolyte thermal stability test. Reprinted with permission from ref. [60]. Copyright 2024, The Author(s), under exclusive licence to Springer Nature Limited. f) P-MSE electrolyte flammability test. g) TEM image of FeHCF. Reprinted with permission from ref.61. Copyright 2024 Wiley-VCH GmbH.

ance in half-cells with graphite as the anode, with a capacity retention of 92.4% after 1400 cycles at a current density of  $0.1 \text{ A g}^{-1}$ . The  $\text{K}_{0.5}\text{MnO}_2 \parallel$  graphite full cell exhibits a good stability of 50 cycles, with a capacity retention of 75%. The flammability test of the electrolyte also showed excellent flame resistance.

Qin et al.<sup>[58]</sup> prepared a solution with a molar ratio of KFSI:DME:1,1,2,2-tetrafluoroethyl-2,2,2-trifluoroethyl ether (HFE)=

1:1.90:0.95 as the electrolyte, which showed strong high-voltage characteristics. LSV tests found no obvious anodic current generation at voltages higher than 5.3 V (relative to  $\text{K}^+/\text{K}$ ). It also showed excellent cycling stability in half-cells with high-loaded graphite as the anode (about  $8 \text{ mg cm}^{-2}$ ), with a reversible capacity of  $200 \text{ mAh g}^{-1}$  after 300 cycles and an coulombic efficiency of 99.5%. The flammability test of the electrolyte also showed that it had better thermal stability than

low-concentration ether-based electrolytes, with only a small blue flame after ignition. In another work, Wen *et al.*<sup>[59]</sup> developed an electrolyte where 1 M KFSI was dissolved in a mixed solvent of TMP and non-fluorinated anti-solvent 1,4-dioxane (DX) with a volume ratio of 1:1. This electrolyte has a wide voltage window, as shown in Figure 4d, and enable the K||graphite cells to cycle 1000 cycles without obvious capacity decay, and the K||K cell to cycle for over 2200 hours. Also, the K||PTCDA full cells exhibited a stability of 100 cycles with a capacity retention of 80% and a high coulombic efficiency of 99.8%.

In order to balance the relationship between battery safety, cost, and performance, Yi *et al.*<sup>[60]</sup> developed a safe, economical, and high-performance electrolyte with 1 M KFSI in DME/3-methoxyperfluoro(2-methylpentane) (MME)/1,1,2,2,5,5,6,6-octafluoro-3-oxahexane (TTE(HFE)) (with a molar ratio of DME:MME:OOE=3.5 : 0.875:3). Under this electrolyte, the K||K cell stably cycled for over 12 months. And the K||graphite cell also showed good cycling stability, with a battery retaining 93% of its initial capacity after 2400 cycles. This electrolyte has also been proven to have a wide operating temperature window, and due to the non-flammability of the electrolyte (Figure 4e), the safety of the battery is also guaranteed. To improve the safety and cycle life of potassium-ion batteries, Zhang *et al.*<sup>[61]</sup> dissolved 0.6 M of KFSI into a mixed solvent composed of tris(1-propyl) phosphate (TPP), tris(2,2,2-trifluoroethyl) phosphate (FTEP), and OOE with a volume ratio of TPP:TFP:TTE=4:1:5 as the electrolyte (P-MSE). The electrolyte is non-flammability (Figure 4f), reveals its excellent safety. Also, this electrolyte showed a wide electrochemical window, allowing K||PB half-cell to cycle over 2000 times at 4.2 V high voltage with a capacity retention of 80%. It also matched well with graphite, with the graphite anode not experiencing capacity decay after 1300 cycles in this electrolyte. The pouch cell also showed excellent stability under this electrolyte, with the battery retaining over 81% of its initial capacity after 1400 cycles and an average coulombic efficiency of 99.6%. The excellent stability is due to the generation of thin and uniform SEI (Figure 4g).

In order to develop a non-flammable localized high-concentration electrolyte to improve the stability and safety of potassium-ion batteries based on aluminum collectors, Chen *et al.*<sup>[62]</sup> developed a TMP\_LHCE electrolyte with a molar ratio of KFSI:TMP:HFE=1:1.7:2. The K||Al half-cell using the TMP\_LHCE electrolyte showed stable potassium metal cycling for over 3400 hours (over 800 cycles) with a high reversibility of 98%. At a current density of 0.2 C, the graphite anode in the TMP\_LHCE electrolyte maintained over 300 cycles with a reversible capacity of 257.6 mAhg<sup>-1</sup> and a capacity retention of 92.6%. The SEI formed in this electrolyte has better stability and mechanical strength, effectively inhibiting the corrosion of the aluminum collector and reducing the dissolution of the SEI, thereby improving the cycling stability of the anode. And this electrolyte also has excellent flame resistance. It shows self-extinguishing characteristics in ignition tests, significantly improving the safety of the battery system compared to traditional flammable carbonate ester-based electrolytes.

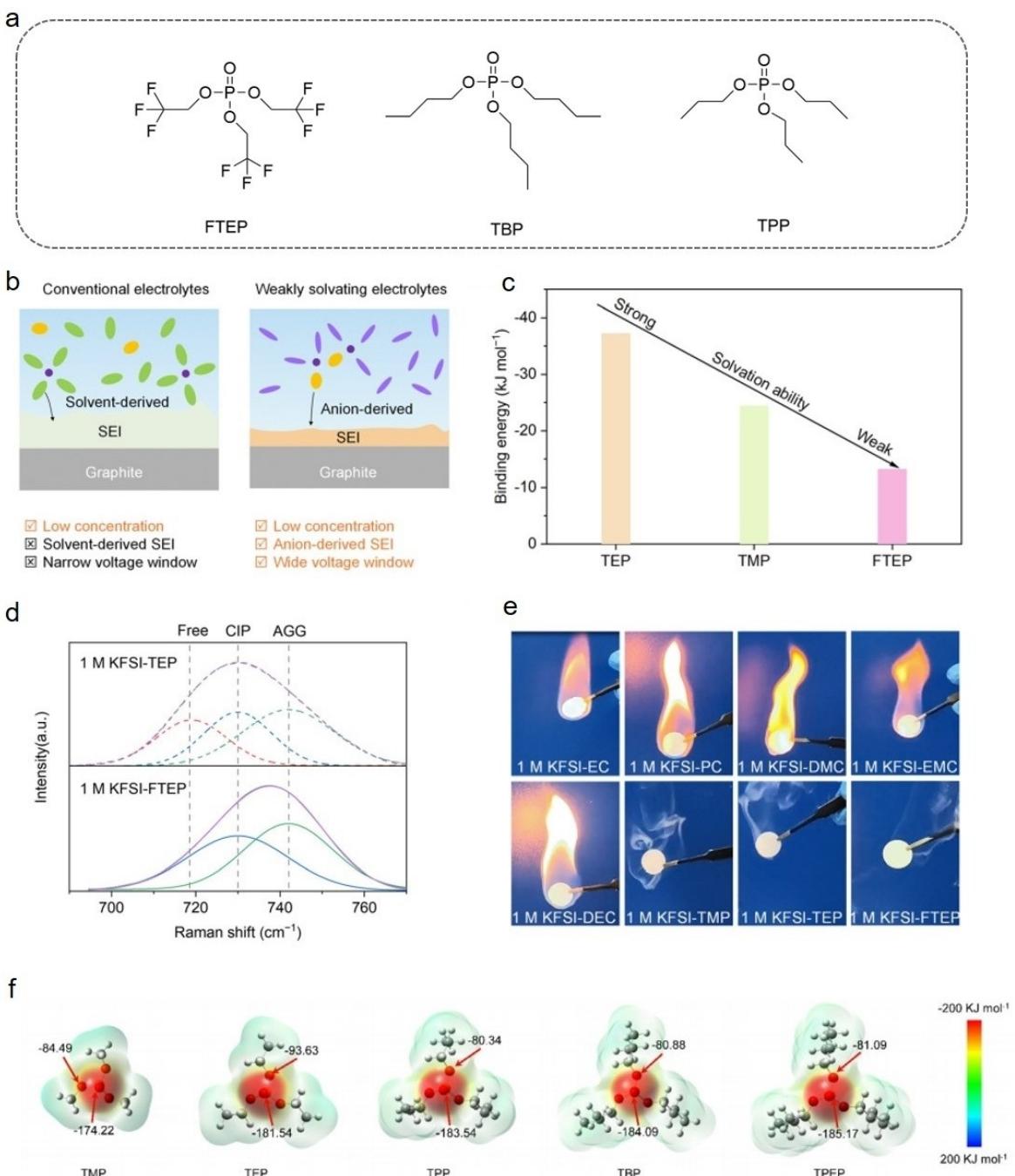
In general, LHCEs have adeptly inherited the merits of HCEs, while mitigating the issues of high viscosity and elevated salt cost associated with HCEs. Despite the excellence of LHCEs as electrolytes, they are still face some challenges. Key concerns include the environmental friendliness and cost-effectiveness of additives, as well as the complexity of the preparation process. Addressing these issues requires further discussion and practical experimentation to foster the ongoing development and application of LHCEs.

#### 4. Weakly Solvating Electrolytes

In addition to localized high-concentration electrolytes, there are weakly solvating electrolytes that can also meet the requirements of low salt cost, highly compatibility with graphite and other electrodes.<sup>[63]</sup> The weakly solvating electrolytes that we are familiar with typically include fluorinated solvents and some phosphate ester solvents such as FTEP, tributyl phosphate(TBP), and TPP. Their chemical formulas are shown in Figure 5a. Weakly solvating electrolytes can utilize the non-flammable or even fire-retardant solvents, which greatly ensures the battery's safety.<sup>[64]</sup> Weakly solvating electrolyte can help form more stable and anion-derived SEI with fewer defects, as shown in Figure 5b, which can improve the battery's ability to operate at high voltages without degradation.<sup>[65]</sup> This characteristic helps to improve the electrolyte's antioxidant capacity, thereby broadening the battery's electrochemical window.<sup>[66]</sup> Moreover, weakly solvating electrolytes can also promote the formation of an inorganic-rich SEI, effectively protecting the electrode surface and reducing the decomposition of the electrolyte.<sup>[67]</sup> Weak solvation means that the interaction between the solvent and the potassium ions is weak, as shown in Figure 5c, while the interaction between the anions and the potassium ions is strong, which is also conducive to forming a stable SEI on the electrode surface, reducing the continuous decomposition of the electrolyte, thereby extending the battery's cycle life and improving the battery's coulombic efficiency.

Compared with localized high-concentration electrolytes, weakly solvating electrolytes have a simple formulation.<sup>[68]</sup> The removal of diluents further increases economic benefits. Thanks to their unique solvation structure, as shown in Figure 5d, they not only achieve the purpose of reducing free ions in high-concentration electrolytes but also reduce the amount of salt used.<sup>[69]</sup> Moreover, they also have unique advantages in terms of safety. In other words, weakly solvating electrolytes enable the electrolyte to achieve excellent electrochemical performance at a lower salt concentration.<sup>[70]</sup> They are an economical and promising type of electrolyte with a broad development prospect.

In order to improve the safety and long cycle performance of potassium-ion batteries, Fan *et al.*<sup>[23]</sup> reported a weakly solvating electrolyte composed of 1 M KFSI in FTEP. The design of this electrolyte utilizes the weak solvation ability of the FTEP solvent to promote the formation of an anion-derived SEI on the anode surface and reduce the decomposition of the



**Figure 5.** Weakly solvating electrolytes. a) The molecular formula of the solvent mentioned in the text. b) The illustrations of SEI formation in low-concentration conventional electrolytes and weakly solvating electrolytes. c) The binding energies between various solvents and  $\text{K}^+$ . d) Raman spectra of the 1 M KFSI-TEP and 1 M KFSI-FTEP. e) Flame tests of various electrolytes. Reprinted with permission from ref.23. Copyright 2023 Royal Society of Chemistry. f) Electrostatic potential (ESP) maps. Reprinted with permission from ref. [71]. Rights managed by AIP Publishing.

electrolyte. This electrolyte has demonstrated excellent performance and enable  $\text{K}||\text{Cu}$  cell to stably cycle for over 200 days. Also, when using this electrolyte, the  $\text{K}||\text{graphite}$  cell can stably operate for over 1700 cycles with a capacity retention of 82.2%, corresponding to a long-last operation time for over 26 months (more than two years). In the 1 M KFSI-FTEP electrolyte, the Mn-based Prussian blue analogue (Mn-PBA) cathode material demonstrates good charge and discharge performance

and cycle stability, with a capacity retention of 77.3% after 200 cycles and an average coulombic efficiency of 99.5%. The Mn-based oxides (KMO) cathode material also shows stable cycle performance, with a capacity retention of 83.2% after 100 cycles. As shown in Figure 5d, its non-flammability greatly improves the safety of the battery.

In another work, Geng *et al.*<sup>[71]</sup> developed an electrolyte of 1 M KFSI in TPP. In order to evaluate the effect of the terminal

functional groups in the phosphate-based solvent on the electron density of the oxygen atom, the electrostatic potential energy calculation was performed on it, as shown in Figure 5f. The weak solvation ability and steric hindrance effect of the TPP solvent induces an anion-rich solvation structure. The unique solvation structure further conducive to forming a stable SEI on the electrode surface, which helps to protect the electrode and reduce the decomposition of the electrolyte, thereby improving the battery's cycle performance. The K||graphite half-cell using the 1 M KFSI-TPP electrolyte shows good capacity retention and high coulombic efficiency. The K||PB half-cell undergoes 300 cycles at a high current density of  $500 \text{ mA g}^{-1}$  with a capacity retention of 93.38% and an average coulombic efficiency of 98.79%. Furthermore, the K||PB full-cell showed negligible capacity decay after 300 cycles with an average coulombic efficiency of 99.08%.

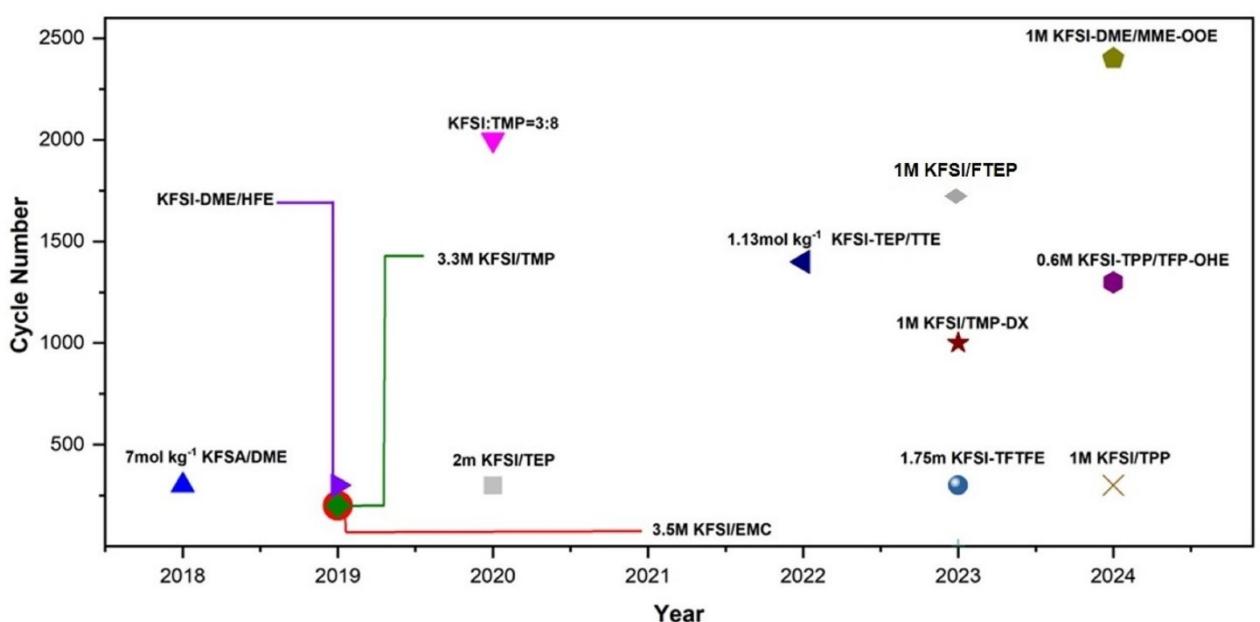
Weakly solvating electrolytes are particularly adept at facilitating the formation of thinner, more homogeneous inorganic-rich SEI, which significantly enhances the cycling

stability of batteries. Additionally, these electrolytes exhibit superior thermal stability and an expanded electrochemical window, contributing to the overall safety of the batteries. Despite these notable advantages, weakly solvating electrolytes confront certain challenges, including the need to achieve a delicate balance between solution concentration and ionic conductivity, as well as the development of more cost-effective weakly solvating solvents. Overcoming these hurdles is pivotal for the advancement and broader application of weakly solvating electrolytes in the future of battery technology.

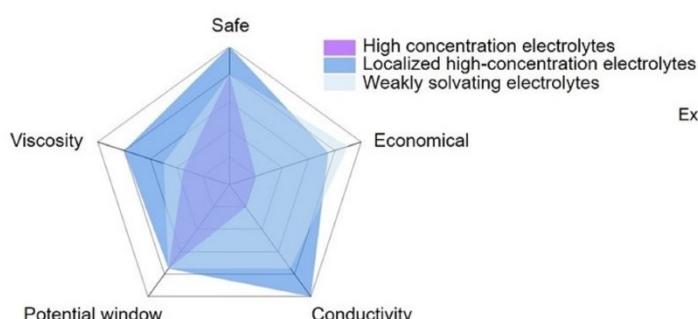
## 5. Summary and Outlook

The graphite anode performance of the various formulations in the article is shown in Figure 6a. In recent years, battery safety and stability issues have gradually gained public attention.<sup>[72]</sup> This article discusses the advantages and disadvantages of different types of electrolytes. Compared to traditional electro-

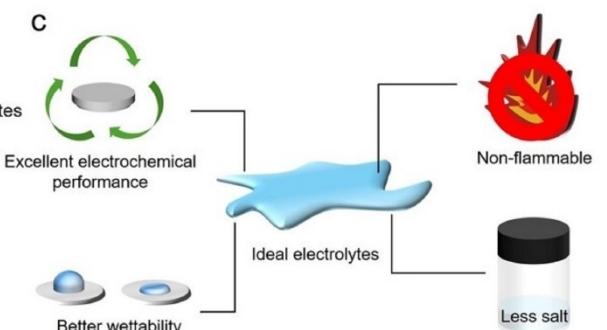
a



b



c



**Figure 6.** Summary of the three kinds of electrolytes. a) Summary of the performance of graphite anodes in various electrolytes. b) Radar diagram of high-concentration electrolyte, localized high-concentration electrolyte and weakly solvating electrolyte. c) The direction of developing novel electrolytes in PIBs.

lytes, high-concentration electrolytes have sacrificed viscosity at the cost of improving thermal stability and cycle stability.<sup>[73]</sup> To address the viscosity and cost issues of high-concentration electrolytes, scientists have begun to use localized high-concentration systems. This system greatly retains the unique solvation environment of high-concentration electrolytes, reduces the salt content per unit volume by adding diluents, and lowers the viscosity of the high-concentration electrolyte.<sup>[74]</sup> Similarly, weakly solvating electrolytes, which achieve the effects of high-concentration electrolytes at low concentrations, rely on their weak interaction with potassium ions to enhance cycle stability.<sup>[75]</sup> Moreover, due to their non-flammability, they improve battery safety. A more intuitive comparison can be observed from the radar plot in Figure 6b.

From a safety perspective, non-flammable electrolytes are more conducive to maintaining battery safety. Taking phosphate esters as an example, although they are not compatible with graphite anodes at low concentrations, this compatibility issue has been resolved through high-concentration electrolyte strategies.<sup>[76]</sup> However, this approach leads to a sharp increase in salt costs, and the electrolyte has high viscosity and poor wettability, which hinders its development.<sup>[77]</sup> The emergence of local high-concentration electrolytes has effectively addressed this issue.<sup>[78]</sup> Weakly solvating electrolytes with nonflammable solvents, benefiting from their weak interaction with potassium ions, it can effectively control the amount of salt used in the electrolyte and are nonflammable or even flame-retardant, ensuring battery safety and stability.<sup>[79]</sup> In summary, an excellent electrolyte should possess characteristics such as being non-flammable, low salt costs, low viscosity, and excellent electrochemical performance.<sup>[80]</sup> In the future development, electrolytes that are more cost-effective, have better wettability, and are non-flammable will have a broader future, while ensuring performance, as shown Figure 6c. It is hoped that this review will provide corresponding assistance for the designing of stable and safe potassium-ion batteries. For ease of reference, Table 1 describes the full names and abbreviations in the text.

**Table 1.** A table of full names and abbreviations that appear in the article.

Full name	Abbreviation
Lithium-ion batteries	LIBs
Sodium-ion batteries	SIBs
Potassium-ion batteries	PIBs
Solid electrolyte interphase	SEI
Potassium bis(fluorosulfonyl)imide	KFSI
1,2-dimethoxyethane	DME
Trimethyl phosphate	TMP
Ethylene sulfate	DTD
High-concentration electrolytes	HCEs
Ethyl methyl carbonate	EMC
Linear sweep voltammetry	LSV
Coulombic current	CC
Dual-graphite battery	DGB
Diethylene glycol dimethyl ether	DEGDME
Prussian blue	PB
Mn-based Prussian blue analogue	Mn-PBA
Mn-based oxides	KMO
X-ray photoelectron spectroscopy	XPS
X-ray diffraction	XRD
Triethyl phosphate	TEP
Perylene-3,4,9,10-tetracarboxylic dianhydride	PTCDA
Potassium hexafluorophosphate	KPF <sub>6</sub>
Ethylene carbonate	EC
Diethyl carbonate	DEC
Local high-concentration electrolyte system	LHCEs
1,1,2,2-tetrafluoroethyl 2,2,3,3-tetrafluoropropyl ether	TTE
1,1,2,2-tetrafluoroethyl-2,2,2-trifluoroethyl ether	HFE
3-methoxyperfluoro(2-methylpentane)	MME
1,1,2,2,5,5,6,6-octafluoro-3-oxahexane	OOE
Tris(1-propyl) phosphate	TPP
Tris(2,2,2-trifluoroethyl) phosphate	FTEP
Tributyl phosphate	TBP

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## Conflict of Interests

The authors declare no conflict of interest.

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