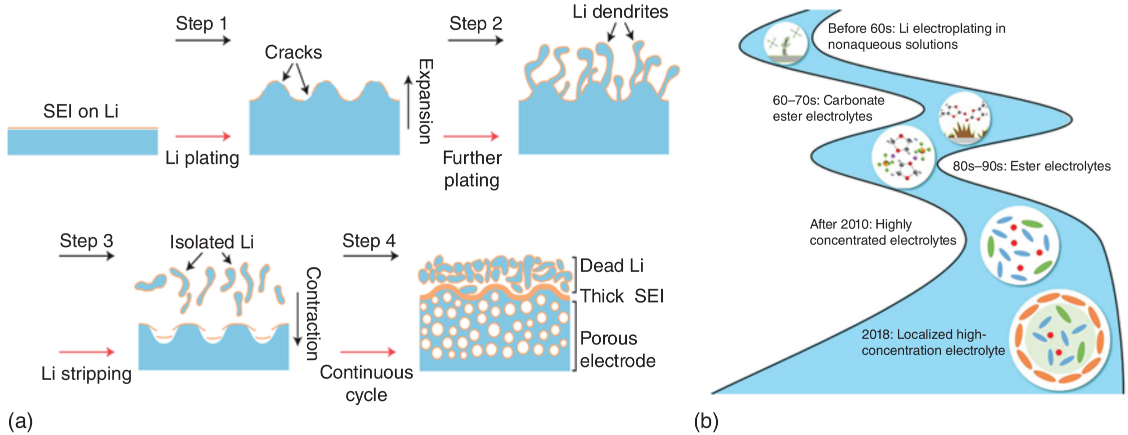

Figure 1.25 (a) Schematic showing the problems related to Li metal during the Li stripping/plating process. Source: Reproduced with permission Lin et al. [200]. (b) Schematic diagram for the development progress of liquid electrolytes in lithium metal batteries. Source: Reproduced with permission Jie et al. [201].

As shown in Figure 1.25b, the investigation of electrolyte for Li plating/striping can be dated back to the nineteenth century, using a nonaqueous electrolyte with LiCl and pyridine. Then, different salts and solvents were studied [208, 209]. After preliminary studies, aprotic solvents were proved to be useful to inhibit the reaction between Li metal and electrolytes. In the 1960s, due to the demand for high-energy-density storage systems, the electrolytes used in LMBs started to get attention. Electrolytes with alkyl carbonate esters such as propylene carbonate (PC) as solvents and $LiPF_6$ or $LiClO_4$ as Li salts were used in rechargeable batteries due to their good ionic conductivity [210]. However, carbonate ester-based electrolytes showed low coulombic efficiency, which hindered their further application [211].

In the late 1970s, ether electrolytes were reported, which could obtain high CE and stable reversibility of the Li electrode due to the formation of more suitable SEI layers with both organic and inorganic contents [212]. With the commercialization of Li–ion batteries with graphite as anode, the research on electrolytes for LMBs was limited. Recently, as graphite anodes are approaching their theoretical capacity, there has been a renaissance in LMBs. At this stage, excellent strategies have been applied to improve the cycling stability of both anode and cathode through electrolyte modifications. Adding suitable electrolyte additives is a simple and promising way to in situ form stable solid electrolyte interfaces on both anode and cathode, leading to enhanced cycling stability of LMBs [213]. For example, gradient SEI layer was formed by adding bisfluoroacetamide (BFA) into the electrolyte, consisting of a C–F rich surface and a LiF rich bottom layer (Figure 1.26a,b), which can contribute to the preferential reduction of BFA on Li metal surface due to higher reductive potentials (Figure 1.26c) [214]. As a result, the Li||Li batteries can stably cycle for 400 cycles at a current density of $1\,mA\,cm^{-1}$ (Figure 1.26d). The solvation structures of Li ions can also be changed by adding electrolyte additives, such as alkyl-triphenyl-phosphonium bromides (alkyl-TPPB) [215]. The presence of Br^- ions in the electrolyte can enlarge the reaction distance between Li ions with solvents (Figure 1.26e). Thus, the coordinate number of EC in solvation structures decreases, which can suppress the reduction of solvents and improve the stability of electrolytes. HCEs were also reported to improve the cycling stability of Li metal anodes [216, 217]. In addition, HCEs exhibit good oxidation stability, which can simultaneously improve the stability of cathode materials, such as $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC811). However, high cost and viscosity of HCEs make them unsuitable for practical applications. Therefore, diluents are added into electrolytes to form localized high-concentration electrolytes (LHCEs) [218]. LHCEs exhibit the advantages of HCEs, as well as decrease the cost and viscosity of HCEs, which are regarded as advanced liquid electrolytes for Li metal batteries.

Except these electrolyte modifications, some functional electrolytes were also studied, such as nonflammable electrolytes [219], high-voltage electrolytes [220], electrolytes operated at extreme conditions (such as high or low temperature) [221]. Significant advances from both the fundamental understanding and practical engineering have been made, but the reaction between Li metal and electrolyte is

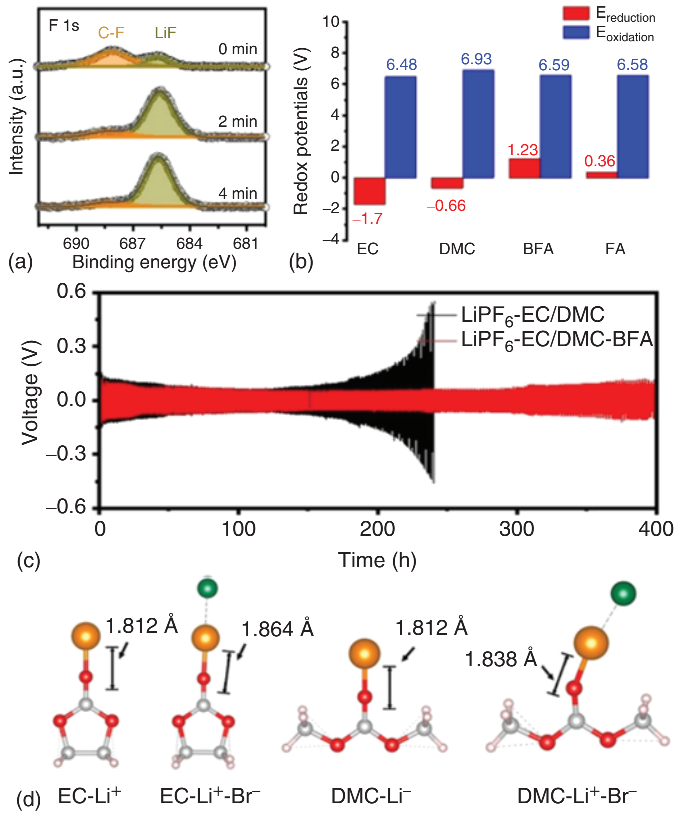

Figure 1.26 (a) XPS spectra of F 1s in Li||Li cells after 10 cycles in $LiPF_6$-EC/DMC-BFA electrolyte. (b) Theoretical calculation of redox potentials of EC, DMC, BFA, and FA molecules; (c) cycling performance of Li||Li cells using $LiPF_6$-EC/DMC-BFA electrolyte at a current density of 1 mA cm^{-2} with a cutoff capacity of 0.5 mAh cm^{-2}. Source: Reproduced with permission Li et al. [214]. (d) The comparison of the distance between Li^+ ion and O atom of the EC and DMC molecules with and without the Br^- ion coordinator. Source: Reproduced with permission Qi et al. [215].

not inhibited, which will affect the practical applications of LMBs with long-term requirements.

1.4 Prospects

Over the past several decades, LMBs have been regaining intensive research attention due to their high energy density compared with Li-ion batteries. Until now, practical applications of LMBs with different cathodes is not realized due to their inherent drawbacks. Therefore, to solve these detrimental issues and realize the practical deployment of LMBs, many problems need to be addressed:

(1) Mechanism: Clear understanding of the reaction mechanism is vital to further improve electrochemical performance of batteries. The mechanism of Li_2O_2

decomposition on a cathode catalyst in Li–O_2 batteries is not elucidated yet. Due to the sluggish kinetics of O_2, designing cathodes with high surface area as well as catalysts with high OER catalytic activity is important. In addition, for practical applications of Li–O_2 batteries, the effect of CO_2 in air should be considered, which can react with Li_2O_2 to produce Li_2CO_3. For Se cathodes, the redox reaction pathway in carbonate electrolytes is not clear. Some works indicate a single-conversion process, while others indicate a multiple-reaction process. In addition, due to the insolubility of polyselenides in carbonate electrolytes, in-depth research needs to be done on carbonate electrolytes rather ether-based electrolytes.

(2) Shuttle effect: The shuttle effect is a big problem in Li–S, Li–Se/Te, Li–I_2/Br_2 batteries. To solve this problem, designing cathode host materials with both physical and chemical adsorption properties is a promising way, such as heteroatom doping on carbonaceous materials and introducing metal-based polar species. Inserting a block interlayer is the simplest way. For practical applications, lean electrolytes and high mass loading should be considered, which are generally are paid less attention. Exploring SSEs with high ionic conductivity is the direct way to inhibit the shuttle effect.

(3) Li metal anodes: Due to random deposition of Li ions, the dendrite growth is the biggest challenge with Li metal batteries. More works on anode protection need to be done, such as electrolyte additives, SSEs, or artificial SEI layer on Li metal. To further understand anode and electrolyte degradation as well as the components and structures of electrolyte interfaces, new analytical tools and in situ techniques should be applied.

If these problems are solved, Li metal batteries will take great strides toward their practical applications.

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