Received: 2 December 2020 | Revised: 11 January 2021 | Accepted: 26 January 2021

DOI: 10.1002/sus2.4

SusMat WILEY

PERSPECTIVE ARTICLE

A perspective on sustainable energy materials for lithium batteries

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Funding information
National Natural Science Foundation of China, Grant/Award Numbers: 21825501, 21805161, 21808121, U1801257; National Key Research and Development Program, Grant/Award Numbers: 2016YFA0202500, 2016YFA0200102

Abstract

Lithium ion battery has achieved great success in portable electronics and even recently electronic vehicles since its commercialization in 1990s. However, lithium-ion batteries are confronted with several issues in terms of the sustainable development such as the high price of raw materials and electronic products, the emerging safety accidents, etc. The recent progresses are herein emphasized on lithium batteries for energy storage to clearly understand the sustainable energy chemistry and emerging energy materials. The *Perspective* presents novel lithium-ion batteries developed with the aims of enhancing the electrochemical performance and sustainability of energy storage systems. First, revolutionary material chemistries, including novel low-cobalt cathode, organic electrode, and aqueous electrolyte, are *discussed*. Then, the characteristics of safety performance are analyzed and strategies to enhance safety are subsequently evaluated. Battery recycling is considered as the key factor for a sustainable society and related technologies are present as well. Finally, conclusion and outlook are drawn to shed lights on the further development of sustainable lithium-ion batteries.

KEYWORDS
aqueous batteries, battery recycling, lithium batteries, organic cathode, safety, sustainable energy materials

1 | INTRODUCTION

High-energy density and long service life are the permanent pursuits for rechargeable batteries.[1] Battery technologies have made great progress from the rechargeable lead–acid, nickel–cadmium, nickel–metal hydride batteries to the distinguished lithium (Li)-ion batteries (LIBs). Since the successful commercialization in 1991 by

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Sony Corporation, LIBs have revolutionized the market of portable electronic devices and electric vehicles (EVs), and exhibit great promise in large-scale stationary energy storage systems, such as smart grids, which can efficiently *store* and utilize renewable sources, for example, solar and wind power.[2,3] These applications of LIBs render a possible fossil fuel-free, clear, and sustainable society.[4–6] The award of the 2019 Nobel Prize in Chemistry was jointly granted to John Goodenough, Stanley Whittingham, and Akira Yoshino for their significant contributions to the development of LIBs and the sustainable society.

Current commercial LIBs employ inorganic transition-metal phosphate or oxide cathodes (such as, $LiFePO_4$ [LFP], $LiCoO_2$ [LCO], $LiMn_2O_4$ [LMO], $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ [NCA], and $LiNi_xMn_yCo_zO_2$ [NMC, $x + y + z = 1$]), nonaqueous or polymer electrolytes, graphite or silicon-graphite-mixed anodes.[7–9] With a global sale penetration rate of 2.2% in 2018 according to McKinsey's Electric Vehicle Index (https://www.mckinsey.com/industries/automotive-and-assembly/our-insights/expanding-electric-vehicle-adoption-despite-early-growing-pains), EVs currently still only account for a small percentage of vehicle market. In the long term, if more vehicles and portable electronic devices are shifted to be powered by LIBs, the demand on LIBs will sharply increase. This will lead to the vast consumption of these components in LIBs, such as Li, Co, Ni, Cu, Al, etc., which are high-value metals.[10] Especially, Co and Li are critically rare and very expensive, whose contents in LIBs are 5-15 and 2-7 wt.%, respectively.[11,12] It is forecasted that Li carbonate required by LIB industry will increase from 265 000 tons in 2015 to 498 000 tons in 2025, which can result in a supply shortage in the near future.[13,14] Additionally, the cost of battery accounts for 35-40% of the EV price according to Statista (https://www.statista.com/statistics/797638/battery-share-of-large-electric-vehicle-cost/), leading to the high price of current EVs compared to that of gasoline cars. These scenarios can generate harmful consequence to the sustainable development of LIBs. Advanced electrode design and battery recycling technologies are urgently demanded to handle these problems.[15,16] It should be noted that the ultra-long life is the basic to handle the sustainable dilemma of LIBs, which can reduce the materials requirement and cost in the full-life cycle. Higher energy density and more cycling times are helpful to improve the lifespan of LIBs, which are the research focus of current LIBs and has achieved great success.[17–20]

Another threat to the sustainable development of LIBs is the safety issues of LIBs, which is the bottle neck of the practical applications of batteries.[21] Compared to the fuel vehicles, EVs are still far from mature, and fire accidents are reported occasionally.[22] Compared to the portable devices, the accidents of EVs can generate serious consequence and even personal injury. Materials, cell, and pack design are comprehensively required to guarantee the safe operations of LIBs.[23,24]

In this *Perspective*, the recent progress in energy chemistry of sustainable LIBs is *discussed* (Figure 1). In Section 2, a comprehensive overview of Co-related cathode chemistry during charging and discharging is given. Material chemistries of organic electrodes and aqueous electrolytes are presented in Sections 3 and 4. Characteristics of safety performance for LIBs are introduced in Section 5. Battery recycling technologies are underscored in Section 6. Finally, conclusion and outlook are drawn in Section 7.

2 | LOW-COBALT CATHODE

LCO is one of the most important cathodes and was predominantly used in the batteries of electronic devices and even EVs. However, relative to other transition metals, Co is a less abundant mineral product (0.0023% in the earth crust) and difficult to mine because Co is generally a by-product of Cu/Ni mines. Additionally, political issues also raise the mining difficulties because Co is mostly mined in Africa with strong geopolitical instability. All these factors lead to an increasing price of Co from US$ 26 500 per ton in September 2016 to US$ 94 250 per ton in March 2018.[25] Therefore, to obtain a sustainable development of LIBs, Co content in the layered oxide cathode should be much reduced, even to zero.[26,27]

Besides LCO, NCA and NMC are recently equipped in the majority of LIBs for EVs. In terms of cost, Co should be much reduced in the cathode. For example, the cost of LCO batteries can be reduced about two times by these with NCA and NCM811 cathodes, three times by these with the Co-free Ni-rich layered cathode, and 20 times by these with sulfur cathode, without considering the production cost.[28] However, Co is still critically important for the cathode in light of capacity release and cycling stability.[29] Nickel is intrinsically unstable due to its relatively strong magnetic moment in the transition metal layer. When Li intercalates the layer and replaces Ni^{3+}, magnetic frustration can be effectively alleviated, but the replacement can disorder the cathode lattice and lead to the very rapid degradation of cathode structure. When Co is introduced into the cathode structure, it can not only relieve the magnetic frustration, but also maintain the stable cathode structure because Co^{3+} is nonmagnetic (Figure 2).[30] Besides Co, Al can also partly suppress the Li^+/Ni^{2+} mixing to improve the electrode stability, which presents possibility to strengthen the structure stability with the replacement of Co. Mn does not have the ability to suppress the Li^+/Ni^{2+} mixing, while it can increase the thermal stability and reduce the price of the cathode.

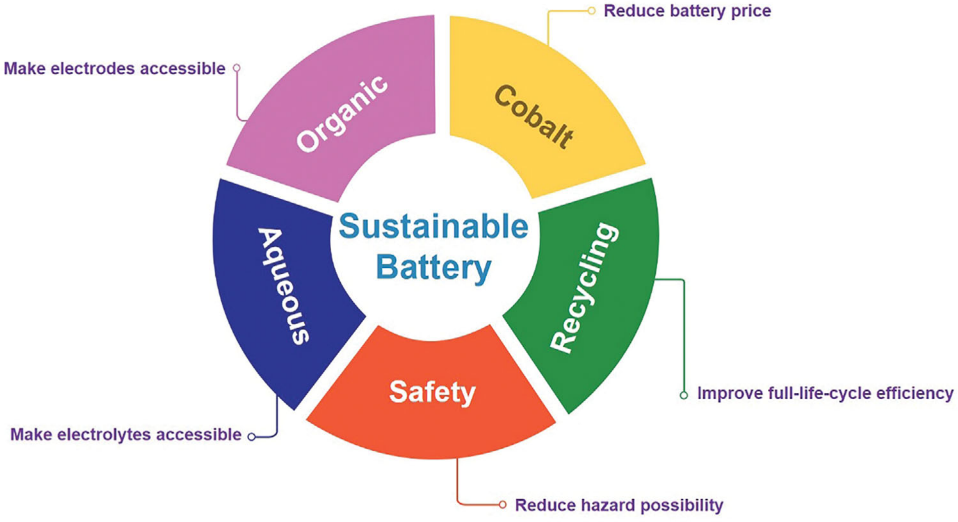

FIGURE 1 Researches for sustainable development of LIBs

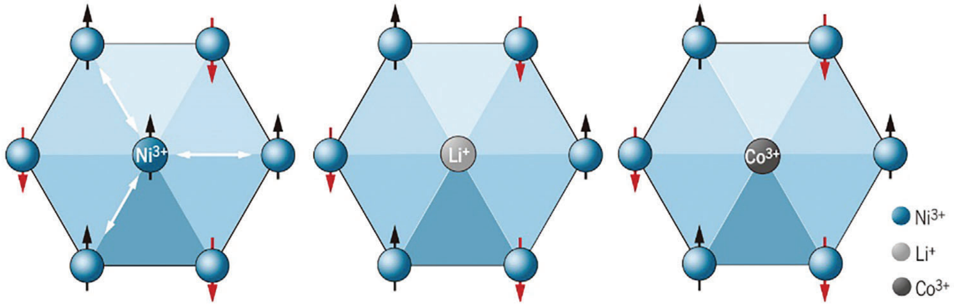

FIGURE 2 Magnetic frustration of Ni, Li, and Co[30]

Extensive research efforts are conducted to reduce or even eliminate the Co content in the cathode of LIBs to lower battery cost while maintaining cycling performance.

1. High-Ni NMC: The NMC chemistry includes several forms, such as NMC111 (Figure 3A), NMC532/622, and most recent NMC811. The high Ni and low Co can lead to a high capacity and low price, meanwhile a poor cycling stability and high safety hazard. Electrolyte modification,[31–33] interfacial design,[34–36] postsynthesis annealing,[37] and morphology regulation,[38] can efficiently relieve these issues.
2. LFP: Reported firstly in 1997 by Goodenough et al., LFP has become an important electrode material (Figure 3B),[39] which has excellent long-term cycling ability, thermal stability, environmental friendliness, and low cost. Compared with LCO and NMC, the theoretical capacity of LFP is relatively low (170 mAh/g at room temperature), which limits the practical application of LFP, especially in EVs.[40] By size reduction and carbon coating[41] to improve the rate performance[42] and technology improvement in the pack design, such as cell to pack (CTP) strategy by CATL Corporation and Blade Battery strategy by BYD Corporation, the energy and power density can be largely increased to satisfy the demand of EVs.
3. Lithium- and manganese-rich cathodes: Lithium- and manganese-rich layer-structured cathodes with reduced Co content (such as $Li_{1.2}Ni_{0.15}Co_{0.1}Mn_{0.55}O_2$, Co content: 6.9 vs. 60.2 wt.% for LCO) can deliver high reversible capacities of over 280 mAh/g, which is nearly double of those of LCO and LFP cathodes.[43,44] However, the high capacity is at the cost of severe phase transitions and oxygen release, limiting the cathode voltage caused by activating the lower-voltage redox couples of Mn^{3+}/Mn^{4+} and Co^{2+}/Co^{3+} besides the pristine Ni^{2+}/Ni^{3+}, Ni^{3+}/Ni^{4+}, and O^{2-}/O^{-} redox couples.[45,46] Surface coating and introducing foreign elements can be efficient in inhibiting the voltage fade.

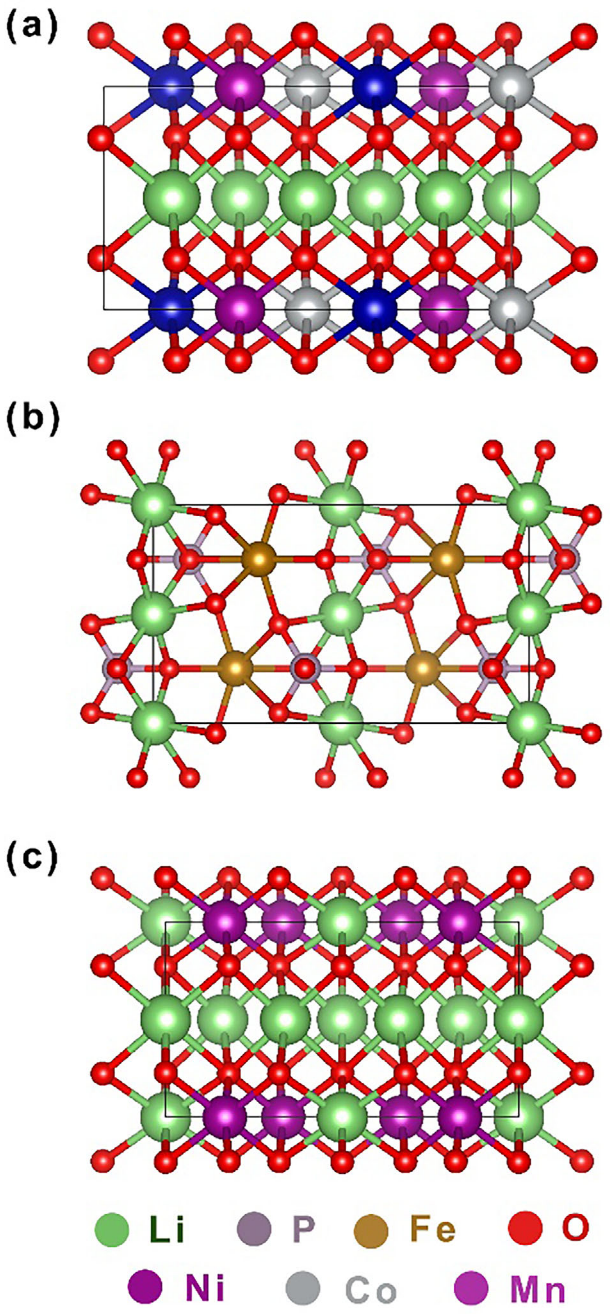

FIGURE 3 Structure of several selected cathode materials. (A) NMC111; (B) LFP; (C) Li_2MnO_3

O_2-type Li-rich cathode of a single-layer Li_2MnO_3 is a completely Co-free system and it can release an ultrahigh capacity of 400 mAh/g, which is induced by the anionic oxygen redox processes (Figure 3C).[47] Spinel $LiNi_{0.5}Mn_{1.5}O_4$ cathode is another promising Co-free electrode with a high voltage of 4.7 V and, thus, an ultrahigh energy density of 650 Wh/kg, 162.5 and 131.3% higher than those of $LiMn_2O_4$ and LFP, respectively.[27] The problems of these similar cathode are the rapid capacity decay and severe electrolyte decomposition, which can be effectively handled by interfacial coating and morphology regulation, etc.[48–51] Additionally, sulfur is also a Co-free cheap electrode material with a high theoretical capacity of 1675 mAh/g, delivering a high theoretical energy density of 2600 Wh/kg when matching Li metal anode.[52–55]

Low-Co cathode is theoretically beneficial for the low cost and sustainable development of LIBs. However, there are still many technical problems such as the complicated and costly production technology of these novel low-Co cathodes and the difficulty in large scalability. Great innovations in science and engineering are critically required for the mass production of low-Co batteries.

3 | ORGANIC ELECTRODE

Since the first commercialization of LIBs, cathodes of LIBs are generally inorganic components, mainly produced from ores, rather than renewable resources.[56–60] In the long term, if all vehicles and electronic devices are powered by LIBs, the cathode materials will suffer from severe resource crisis.[61,62] Therefore, renewable electrode materials are compelled to develop for LIBs. Recently, a new category of organic electrode materials has attracted wide attention due to their potentially sustainable nature and low carbon footprint, which can be facilely produced from biomass.[63–68] Actually, tricarbonyl compound was employed as an organic electrode for LIBs as early as the 1960s.[61] Since that, various organic cathodes have been developed such as polyacetylene, organosulfur compounds, nitroxyl polyradical, etc.[69–71]

The organic electrodes can be generally categorified as three types of p-, n-, and bipolar-type based on their redox mechanisms (Figure 4).[72,73] (1) p-type electrodes must firstly lose electrons from their original neutral state forming a positively charged state. (2) n-type electrodes must accept electrons initially from their origin neutral state forming a negative state. The electrochemical potential of p-type electrode is higher than that of n-type electrode. Therefore, p-type electrode is usually adopted as the cathode, while n-type electrode can be either adopted as cathodes or anodes, decided by its practical redox potentials and its counterparts. (3) Bipolar-type electrodes are able to both lose and accept electrons from the neutral state, thus, can be either charged or discharged initially. The category method is decided by the redox center of organic cathodes in the molecules.

Sustainability is the most significant feature for organic cathode relative to the routine inorganic cathode materials. However, as an electrode potential for practical applications, it must satisfy many other demands.[74] (1) Energy

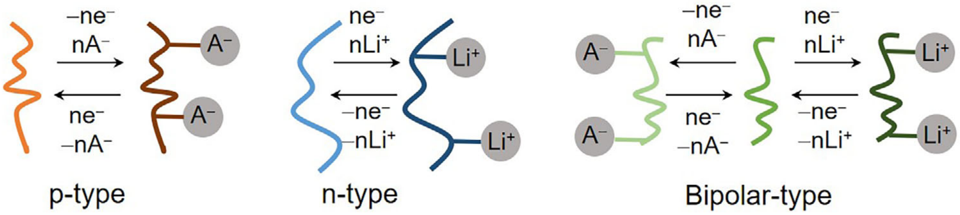

FIGURE 4 Three typical mechanisms for the redox chemistry of organic electrodes

density: the organic electrodes normally cannot present a wide voltage window (higher cathode voltage or lower anode voltage), while they can release a high capacity by overlithiation, such as 720 mAh/g for dimethyl trisulfide[75] and 902 mAh/g for cyclohexanehexone[76] as the cathode, 2000 mAh/g for 1,4,5,8-naphthalenetetracarboxylic dianhydride as the anode.[77] An energy density of 500-750 Wh/kg on the material level can be obtained.[78–82] (2) Power density: it is adopted to indicate the ability of fast charge and discharge. There exists a balance between the energy and power density. A high power density usually corresponds to a low energy density. Because of the fast kinetics and porous structure of organic electrodes, they are potential to have a high power density. (3) Cycling stability: compared to the above two features, this feature of organic electrodes is not good due to the dissolution of organic electrode into the electrolyte, phase transformation, and side reactions. Fortunately, polymer cathode and high-polarity organic salt, etc. are proposed to enhance the cycling lifespan to 1000 cycles with more than 85% of their initial capacity.[83–87]

The current researches of organic electrodes are generally conducted at the material level with a low mass loading and ratio of active materials. More experiments are required to be conducted at practical conditions, such as high areal loading (mAh/cm), low electrolyte amount, low negative/positive ratio, etc. Besides, the detailed redox mechanisms of many organic electrodes are still not clear, which requires further comprehensive investigations. Therefore, before the practical applications, more researches and co-operations from the scientific and engineering communities on organic electrode materials are still demanded.

4 | AQUEOUS BATTERY

Nonaqueous electrolyte is generally adopted in the commercial LIBs due to their wide electrochemical window and relatively high stability against electrode materials. However, the nonaqueous electrolytes also have several defects such as high price leading to the increase in the battery cost and high flammability resulting in safety hazards. Aqueous electrolyte can perfectly handle these two problems of nonaqueous electrolyte due to its intrinsically safe and easily available nature.[88–90] Additionally, aqueous electrolyte also has the features of environmental benignity (nonvolatility and nontoxicity) and capability of fast charging and, thus, high power density based on the potentially high ionic conductivity of aqueous electrolyte.[91–93]

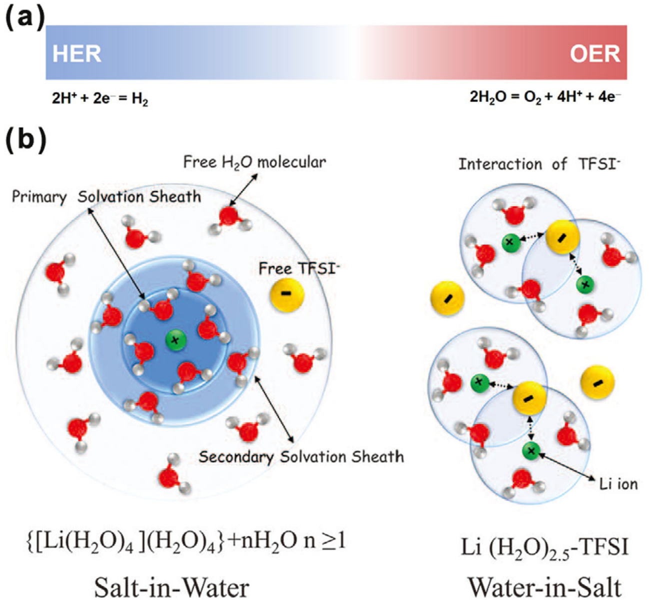

FIGURE 5 (A) HER and OER issues of aqueous electrolyte result in a low voltage; (B) solvation structure of salt-in-water and water-in-salt electrolytes[95]

Except for the obvious advantages, the practical applications of aqueous electrolyte are limited by its theoretically narrow electrochemical window of 1.23 V (Figure 5A),[94] resulting in a low voltage and insufficient energy density of practical batteries. The risk of oxygen evolution reaction (OER) occurs at the cathode part and hydrogen evolution reaction (HER) at the anode part, resulting in electrolyte consumption, side reactions, and severe capacity decay.

To realize the practical applications of aqueous electrolyte, the most urgent issue is to widen its working voltage and improve the battery energy density. Some progress has been achieved: (1) Water-in-salt electrolyte: due to the coexistence of its oxygen site (Lewis basicity) and hydrogen site (Lewis acidity), water has a strong capability to dissolve most of Li salts with a high concentration. Suo et al. successfully dissolved 20 mol lithium bis(trifluoromethanesulfonyl)imide in 1 kg water and obtained highly concentrated aqueous electrolyte (Figure 5B).[95] In the water-in-salt electrolytes, the direct contact of water electrolyte and electrode materials can be potentially prevented due to the unique solvation structure and the absence of free water molecules.[96] Therefore, the oxidation and reduction of water molecules on the electrode surface can be potentially avoided and a stable potential window is substantially extended.[97–99] (2) Hydrate melt electrolytes: similar to the water-in-salt electrolyte, this electrolyte is to obtain a eutectic molten salt in the presence of aqueous electrolyte and efficiently trap water to the ions, maintaining the system with the absence of free water.[100] (3) Interface regulation: similar to the nonaqueous electrolyte, interfacial films are observed as well in the aqueous electrolyte, which can prevent the contact of electrolyte and electrode to widen the electrochemical stable window. Tris(trimethylsilyl) borate additive was demonstrated to form a stable solid electrolyte interface (SEI) on the cathode surface, which can stabilize water molecules at a wide voltage window.[101] By rational designs on the electrolyte and electrode structure, a high voltage is potentially achieved for a practical battery and then a revolution can be realized for LIBs.

5 | BATTERY SAFETY

The safety performance of power batteries is generally characterized by thermal runaway behaviors, a phenomenon that is induced by exothermic reactions in the cells, largely increasing the cell temperature and potentially resulting in fire and even explosion.[102,103] As an energy carrier, there are no absolutely safe batteries and high energy density usually means high potential security risks.[104–106] Take Tesla Model S as an example, its failure rate is about 1 in 10 000, while 7.6 fire accidents in 10 000 vehicles occur in USA.[107,108] Therefore, the self-induced thermal runaway failure of EVs exists based on the probability, but is not higher than that of fuel vehicles. However, due to the highly focused attentions on EVs, the accidents of EVs are much easier to be concerned by the customers. Therefore, the challenged safety performances in particular for the high-energy-density energy storage systems have to be further improved.

Battery temperature abnormally rises under the mechanical, electrical, or thermal abuse conditions, causing the chemical reactions continuously. This results in chain reactions, and leads to thermal runaway.[109] During the entire process of temperature rise in a typical NMC/graphite battery with polyethylene membrane, the decomposition of SEI (80-120°C),[110] reaction between anode and electrolyte, melting of polyethylene, decomposition of NMC and electrolyte, etc., are initiated sequentially (Figure 6A).[107,111] Usually, thermal runaway temperature is defined as where the heat generation rate exceeds 1°C/s. Membrane melting-induced short circuit is the most frequent reason for thermal runaway,[112] while the reactions between plated Li and the electrolyte, the released oxygen from the cathode and the anode are also the reasons under some abuse conditions, such as high rate, overcharge, and overdischarge, etc. (Figure 6B). Even in some systems with solid-state electrolyte (SSE), the side reactions between Li metal and SSE can generate a great deal of heat and result in thermal runaway.[113] By comparing the thermal runaway temperatures, heat generation rates, and total generated heat of different SSEs, the thermal stability of four SSEs matching Li was identified with the order of $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3 < Li_{1.4}Al_{0.4}Ti_{1.6}(PO_4)_3 < Li_{3x}La_{2/3\text{-}x}TiO_3 < Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$. The generated oxygen from SSEs at the increased temperatures is considered to account for the thermal runaway of SSEs matching Li metal anodes.

Methods in materials and pack scale, and safety detections are required to reduce the hazard caused by thermal runaway (Figure 6C).[114] (1) Battery materials modifications: surface coating of the cathode[115–118] and anode,[119–121] stable electrolyte system,[122–124] and stiff separator[125–128] are designed to enhance the intrinsic safety of batteries. (2) Pack design: the battery pack must be mechanically strong to bear unanticipated mechanical destruction. Some unique electronics, such as fuse, thermistor, etc., are beneficial to diminish the continuous damage of the external short circuit, overcharge, overdischarge, etc. (3) Safety detection: each battery pack must be equipped with a battery management system (BMS) to detect the state of batteries, such as state of charge (SOC), state of energy (SOE), state of health (SOH), cell inconsistency, etc., which are accurately estimated online. When BMS detects the potential safe hazards, early warning must be provided to avoid the secondary hazard. As indicated by Ouyang and coworkers, the evacuation time required for a light vehicle must be larger than 30 s and 5 min for a 12 m-length bus to guarantee the passenger safety during the thermal runaway accident.[107] An enhanced safety performance is promising to achieve through the multiscale strategies.

6 | BATTERY RECYCLING

According to EV Sales Blog, the sales of EVs exceeded 2.2 million worldwide in 2019. If the average weight and volume of battery pack are conservatively assumed to 250 kg and 0.5 cubic meter, the resultant pack wastes can be around 550 000 tons and 1.1 million cubic meter after

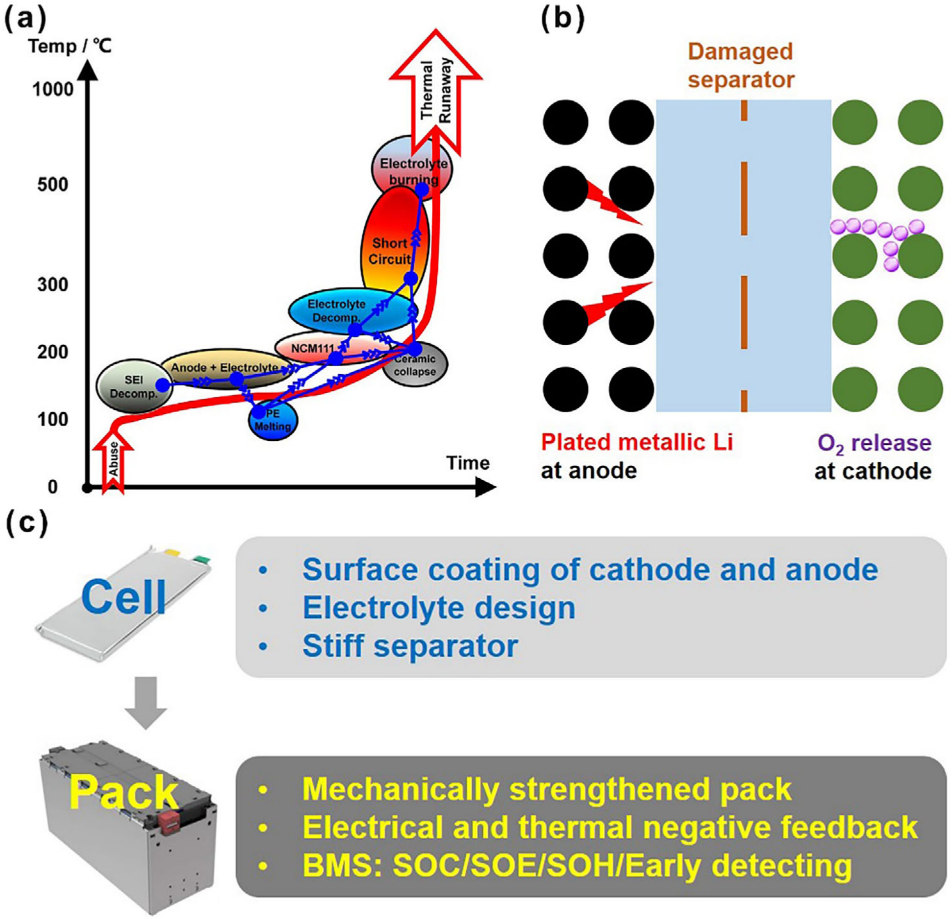

FIGURE 6 (A) Temperature-time curves during thermal runaway.[107] (B) Thermal runaway sources for LIBs. (C) Methods to avoid thermal runaway

the scrappage of these vehicles.[129] Additionally, spent LIBs are classified as hazardous wastes, which will pollute the environment and harm the animals and humans. Therefore, recovery of the components of cycled LIBs is valuable from an economical view and critically significant from the perspective of a sustainable development.[130] It is critically pressing to develop battery recycling technologies in terms of environment and economic considerations.

In the waste management hierarchy, there are two levels of recycling options: reuse and recycling. "Reuse" means that power batteries of EVs can have a second life in the low-grade scenarios, while "recycling" means that battery materials should be recovered as much as possible with preserving high quality. Therefore, the spent batteries must be carefully assessed. The detailed recycling processes are very complicated (Figure 7).[129] Three technology routes are usually employed. (1) Pyrometallurgical recovery: pyrometallurgical methods adopt a high-temperature furnace to reduce the metal oxide components to an alloy of Co, Cu, Fe, and Ni, etc. Preferential recovery of Co, Li_2CO_3, and graphite from LCO/graphite battery,[131] Li_2CO_3 from spent LCO/LMO/NMC,[132] Li_2CO_3 and Mn_3O_4 from LMO/graphite battery[133] has been successfully realized by this method. (2) Hydrometallurgical recovery: hydrometallurgical methods adopt aqueous solutions to retain the desired metals from cathodes and the most frequently adopted aqueous electrolyte is H_2SO_4/H_2O_2 system.[134,135] This method can be facilely conducted at room temperature, while may generate large quantities of waste water and require associated additional costs. Based on this strategy, Mn separation from others,[136] Co of high purity,[137] Li and Co separation with high efficiency[138,139] can be achieved. (3) Biological recovery: this is an emerging method for LIB materials recovery and can be potentially supplementary to the other two methods for metal recovery. For example, Co and Ni are very difficult to separate by the pyrometallurgical and hydrometallurgical methods, while specific microorganisms can digest the metal oxides selectively from the cycled cathodes and reduce them to generate Co and Ni particles.[140]

There are several key challenges for the current LIB recycling technologies. (1) Technical aspects: the disassembly of large capacity power batteries has lots of difficulties due

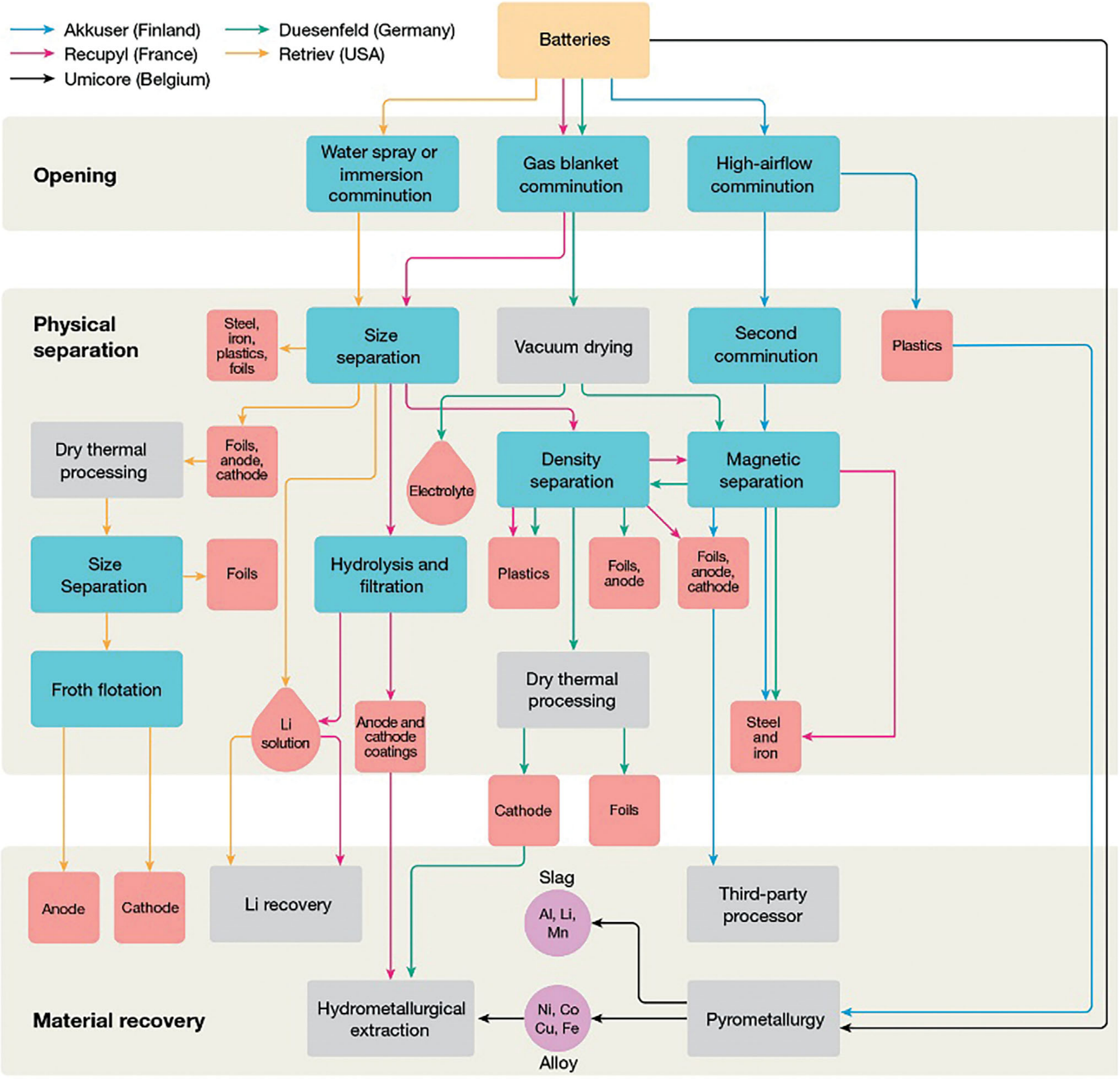

FIGURE 7 The typical routes for battery recycling[129]

to the variations in the cell structure and chemistry. Complicated multiple operations are usually required with a well-established automatic processing line. The disassembly process also has great potential hazards induced by short circuit. (2) Economic aspects: based on cost-benefit calculations, the current recycling technologies are generally conducted on the cell components with high price, such as Li, Co, and Ni,[10] while some less expensive components are not much considered. Advanced recycling technologies with economic feasibility at least cost-neutral are urgently required to recover other battery components, which is required by a sustainable development. (3) Environmental aspects: the recycling technology involves high temperature or large quantities of water treatments. It must be noted that the recycling process must not generate additional pollutions to the environment. Therefore, based on the great prospects of battery energy storage systems and the immaturity on the battery recycling technology, there is plenty of opportunities in this field.

7 | CONCLUSIONS AND OUTLOOK

Energy production is the permanent motivation to the society development, while energy storage systems, especially LIBs, can make the energy utilization more efficient.

Sustainable development of LIBs has become a worldwide objective and received more attention than ever before due to the vast production and extensive applications of LIBs. Sustainability should be regarded as an additional dimension besides morphology, composition, and structure when designing next-generation batteries. In the *Perspective*, the significance of sustainable development of LIBs is emphasized, and the energy chemistry of sustainability is discussed including low Co content, organic electrode, and aqueous battery, battery safety, and battery recycling. By clearly analyzing these aspects, novel materials and energy chemistry, advanced technologies are required to realize the sustainable development of LIBs.

The combination of high electrochemical performance and sustainability has become crucial for the further development of advanced LIBs. Much efforts have been made during previous researches in designing novel electrodes, electrolytes, and separators to achieve batteries with high performance in energy density, power density, lifespan, safety, and low cost. Further remarkable breakthroughs in battery sustainability in theory and application are required and several aspects can be considered in the future researches on sustainable LIBs.

Sustainability of LIBs involves the full-life cycle, and is a set of raw materials, synthesis of battery components, battery assembly, use, and recycling. The cases discussed in the *Perspective* are just some pivotal issues during the practical applications of LIBs. Clearly analyzing the full-life-cycle feature, developing novel energy chemistry, and realizing the efficient recovery of spent battery materials of LIBs are of vital importance for the sustainable LIBs, which are beneficial to build a harmonious and sustainable society.

ACKNOWLEDGMENTS

This work was supported by National Natural Science Foundation of China (21825501, 21805161, 21808121, and U1801257) and National Key Research and Development Program (2016YFA0202500 and 2016YFA0200102).

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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How to cite this article: Cheng X-B, Liu H, Yuan H, et al. A perspective on sustainable energy materials for lithium batteries. *SusMat*. 2021;1:38–50. https://doi.org/10.1002/sus2.4