#### REVIEW



# Batteries for electric vehicles: Technical advancements, environmental challenges, and market perspectives

Axel Celadon<sup>1</sup> | Huaihu Sun<sup>2</sup> | Shuhui Sun<sup>2</sup> | Gaixia Zhang<sup>1</sup>

#### Abstract

The rapid evolution of electric vehicles (EVs) highlights the critical role of battery technology in promoting sustainable transportation. This review offers a comprehensive introduction to the diverse landscape of batteries for EVs. In particular, it examines the impressive array of available battery technologies, focusing on the predominance of lithium-based batteries, such as lithium-ion and lithium-metal variants. Additionally, it explores battery technologies beyond lithium ("post-lithium"), including aluminum, sodium, and magnesium batteries. The potential of solid-state batteries is also discussed, along with the current status of various battery types in EV applications. The review further addresses end-of-life treatment strategies for EV batteries, including reuse, remanufacturing, and recycling, which are essential for mitigating the environmental impact of batteries and ensuring sustainable lifecycle management. Finally, market perspectives and potential future research directions for battery technologies in EVs are also discussed.

#### **KEYWORDS**

electric vehicles, environmental challenges, market perspectives, rechargeable batteries, technical advancements

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#### INTRODUCTION 1

The widespread adoption of electric vehicles (EVs) is an effective way to promote carbon neutrality, reduce greenhouse gas (GHG) emissions, and combat climate change. In 2021, as depicted in Figure 1A, global GHG emissions were dominated by five key sectors: energy (14.5 GtCO<sub>2</sub>-eq), industry (14.5 GtCO<sub>2</sub>-eq), agriculture,

forestry and land use (10 GtCO<sub>2</sub>-eq), transport (8 GtCO<sub>2</sub>eq), and building  $(3.5 \text{ GtCO}_2\text{-eq})$ .<sup>1</sup> The transport sector, central to this discussion, stands as the fourth-largest emitter of GHGs. Alarmingly, unlike the other sectors, it exhibited a persistent 2% increase in emissions between 2010 and 2019.<sup>2,3</sup> These data alone underscore the critical urgency of addressing emissions from the transport sector in the global effort against climate change. The government, industry, and academia are actively promoting the development of an EV-based transportation system.

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Axel Celadon and Huaihu Sun contributed equally to this work.



FIGURE 1 (A) Global greenhouse gas emissions in 2021. Reproduced under the terms of the CC-BY open access license.<sup>1</sup> Copyright 2021, The Authors. (B) Comparison of global average medium-car lifecycle greenhouse gas (GHG) emissions. BEV, battery electric vehicle; HEV, hybrid electric vehicle; ICEV, internal combustion engine vehicle; PHEV, plug-in hybrid electric vehicle. Tank-to-wheel: emissions associated with burning the fuel to power a vehicle (i.e., tailpipe or end-of-lifecycle emissions); Well-to-tank: emissions that happen between extraction through delivery to a fueling station and transfer to a vehicle or on-site fuel tank). Reproduced under the terms of the CC-BY open access license.<sup>4</sup> Copyright 2024, The Authors. (C) Life-cycle GHG emissions of electric vehicles between 2021 and 2030. Reproduced under the terms of the CC-BY open access license.<sup>5</sup> Copyright 2021, The Authors.

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In 2023, a medium-sized battery electric car was responsible for emitting over 20 t CO<sub>2</sub>-eq<sup>2</sup> over its lifecycle (Figure 1B). However, it is crucial to note that if this well-known battery electric car had been a conventional thermal vehicle, its total emissions would have doubled.<sup>6</sup> Therefore, in 2023, the lifecycle emissions of medium-sized battery EVs were more than 40% lower than equivalent hybrid electric vehicles (HEVs), and about 30% lower than plug-in hybrid electric vehicles (PHEVs) that have been in operation for over 15 years (about 200 000 km).

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When examining the entire lifecycle of an EV (Figure 1C), the results are encouraging.<sup>7,8</sup> Although the figures vary depending on the geographical area considered, we can observe an average halving of GHG emissions between an EV and a combustion vehicle in 2021, and even a threefold reduction in the case of Europe.<sup>5</sup> Whether in the short or long term, it is undeniable that an EV emits far fewer GHGs than its internal combustion equivalent, making it an excellent solution for decarbonizing the transport sector.

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Central to the success and widespread adoption of EVs is the continuous evolution of battery technology, which directly influences vehicle range, performance, cost, and environmental impact. This review paper aims to provide a comprehensive overview of the current state and future directions of EV batteries. This review will delve into the technical advancements, environmental challenges, and market perspectives of EV batteries, providing a holistic understanding of the current landscape and future trajectory of EV technology. Through this comprehensive analysis, we aim to highlight the critical factors driving the transition toward sustainable transportation and the ongoing efforts to overcome the associated challenges.

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**FIGURE 2** (A) Growth in the number of electric vehicles worldwide between 2010 and 2023. (B) Battery demand by mode and region, 2016–2023. Battery demand refers to automotive lithium-ion batteries (LIBs). Note that in the left chart, LDVs represent light-duty vehicles, including cars and vans; the other modes include two/three wheelers, and medium- and heavy-duty vehicles such as buses and trucks. Reproduced under the terms of the CC-BY open access license.<sup>4</sup> Copyright 2024, The Authors.

#### 2 | EV BATTERIES: A GROWING MARKET

When examining a technology, it is crucial to consider its economic impact. These considerations allow us to gauge the dynamism of this prominent technology, particularly through the investments and economic growth it generates.

# 2.1 | Overview of the economic and business environment

Over the past decade, the global number of EVs has experienced exponential growth, surpassing 40 million units by 2023 (Figure 2A).<sup>4</sup> The demand for EV batteries exceeded 750 GWh in 2023, marking a 40% increase compared to 2022, although the annual growth rate was slightly lower compared to the period between 2021 and 2022 (Figure 2B).<sup>4</sup> This surge can be attributed to various

societal factors, including the political measures taken by various countries to meet their carbon targets, increased consumer awareness of environmentally responsible choices, and the ever-increasing price of oil.<sup>9–11</sup> This rise not only reflects a growing demand but also signifies a significant economic opportunity.

By 2022, the distribution between the various electric battery technologies was far from homogeneous, as shown in Figure 3A,B, with  $\text{LiNi}_x \text{Mn}_y \text{Co}_{1-x-y} \text{O}_2$  (NMC) batteries accounting for 60%, LiFePO<sub>4</sub> (LFP) for 30%, LiNi<sub>0.8</sub>Co<sub>0.15</sub>Al<sub>0.05</sub>O<sub>2</sub> (NCA) for 8%, and other technologies for just 2%.<sup>12,13</sup> Although these concepts are discussed in a separate section, it is important to understand that all three acronyms refer to lithium-ion batteries (LIB), and are simply variants of the same process.

The fact that almost 90% of EV batteries rely on lithium (Li) is not without consequences. Similar to oil, whose over-exploitation has led to critical scarcity, basing the future of the world's car fleet on a single metal would only be repeating a mistake.<sup>14</sup> Moreover, Li is known



**FIGURE 3** (A) Graphs illustrating voltage versus capacity are included for positive- and negative-electrode materials presently employed or under serious consideration for the next generation of rechargeable Li-based cells. Reproduced with permission.<sup>18</sup> Copyright 2001, Springer Nature. The depicted output voltage values apply to both Li-ion cells and Li-metal cells. (B) Distribution of different electric battery technologies in 2022. Reproduced under the terms of the CC-BY open access license.<sup>19</sup> Copyright 2023, The Authors. (C) Evolution of the price of the main materials used to build lithium-ion batteries (LIB) compared with the price of a complete battery. Reproduced under the terms of the CC-BY open access license.<sup>19</sup> Copyright 2023, The Authors.

for its lack of abundance within the Earth's crust, ranking as the 33rd most abundant element on Earth out of over 90 elements.<sup>15</sup> It is mainly found in South America, Australia, and North America in the form of salt lake brine (58%), hard rock Li deposits (26%), and Li clay deposits (7%).<sup>16,17</sup> High-quality Li ore and low magnesium content Li ore are more scarce than salt lake brine resources. The global distribution of Li resources is uneven, leading to significant disparities between total Li resources, detectable resources, and immediate production capacity.

From a purely economic point of view, the intensive mining of such a rare element inevitably leads to a rise in its price, which could seriously undermine the accessibility of one of the most viable solutions for decarbonizing the transport sector.<sup>20</sup> Indeed, the price of lithium car-

bonate (as presented in Figure 3C) rose by a factor of 11 between 2021 and 2023, despite previously being as affordable as other LIB materials.<sup>19</sup> All these factors are driving the search for alternative solutions to LIBs, whether by coupling Li with other materials, or replacing it completely with elements with similar properties.

### 2.2 | Major manufacturers of batteries for electric vehicles

When discussing electric cars, it is common to associate them with large automotive companies such as Tesla, GM, Ford, or BMW. Many of these companies are relying on suppliers and subcontractors for the manufacturing of their electric battery packs.

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**FIGURE 4** Distribution of the number of electric vehicle batteries used worldwide by manufacturer fabricants. Reproduced under the terms of the CC-BY open access license.<sup>21</sup> Copyright 2023, The Authors.

More than 90% of the companies that have designed the majority of EV batteries in use worldwide are based in the Asia-Pacific region, with CATL, LG Energy Solution, and BYD dominating the market (Figure 4).<sup>21</sup> Although these companies are located in the same geographical area, they adopt different strategies and profiles, as demonstrated by the three leading companies in this ranking.

CATL was officially founded in 2011, although it replaced an earlier version of itself (ATL) dating back to 1999.<sup>22</sup> This relatively new company has experienced rapid expansion since its inception, thanks in particular to strategic contracts with EV giants such as BMW in 2012 and Tesla in subsequent years, as well as acquisitions such as Brump Recycling in 2015.<sup>22</sup> This impressive growth trajectory has enabled CATL to control nearly the entire lifecycle of its batteries, highlighting its expertise and quality credentials. All of CATL's R&D centers and facilities are located in China, with the exception of production plants in Germany.<sup>22</sup> Notably, CATL specializes exclusively in energy storage solutions, spanning from batteries to infrastructure for wind farms. As the world's largest EV battery maker, CATL recently announced an investment of \$1.83 billion to construct an offshore wind farm near its headquarters. This initiative aims to provide clean energy to power its operations, thus reducing its carbon footprint from manufacturing.

LG Energy Solution has a very different profile. Originally a division of the Korean LG Group known as LG Chem, it transitioned into a quasi-independent entity from its parent company in 2020.<sup>23</sup> Its foundation was thus supported by the economic power of LG Group. One of LG Energy Solution's most notable achievements is a major contract with General Motors to produce the Ultium LIBs that power many of its EVs, including those under subsidiaries such as Chevrolet (Chevrolet Silverado EV) and Cadillac (Cadillac LYRIQ).<sup>23</sup> Headquartered in Korea, the company operates facilities across four continents, strategically locating plants close to its key partners such as GMC in Michigan (USA). LG Energy Solution's expertise spans a wide range—from batteries for NASA to energy storage packs sold directly to private individual consumers.<sup>23,24</sup>

Finally, BYD introduces another model that is radically different from those of its two competitors. BYD, founded in 1995, is not primarily a battery manufacturer but an automotive company.<sup>25</sup> At its inception, BYD was visionary in specializing in the construction of electric batteries. Over time, it diversified through strategic acquisitions and officially became a car manufacturer. As an established company, BYD now operates in many countries, with factories in Asia, Europe, and North America, and offices in Australia and South America. One of BYD's advantages is its minimal reliance on subcontractors, as it controls the entire production chain for its vehicles and sells its products directly to consumers. An exception to this direct sales model is its contract to supply electric buses for the decarbonization of some British cities' transport networks in 2015.25

This rapid analysis clearly shows that the EV battery market is a dynamic environment teeming with young companies, many of which were formerly start-ups, as well as companies with diverse profiles. No single entrepreneurial model stands out in this race to electrify the world's vehicle fleet, allowing for varied approaches and greater scope for progress. Advantages

Internal improvemen

TABLE 1 Comparison of internal and external improvement



nternal improvement		External improvement			
dvantages	Challenges	Advantages	Challenges		
Significant performance improvements	- Requires considerable research time	<ul><li>Faster results</li><li>Cheaper process</li></ul>	- No in-depth treatment of issues such as security		
issues such as security	- Often requires substantial resources		- Performance is less advanced		

### 2.3 | The role of companies in the development of batteries for electric vehicles

Companies play a critical role in the development of batteries for EVs, focusing on several key areas: (i) materials innovation and research and development (R&D) to enhance battery performance, extend battery lifetime, and ensure safety; (ii) improving manufacturing efficiency to reduce costs; (iii) securing a reliable supply of raw materials (e.g., lithium, cobalt, and nickel) for battery production; (iv) adapting to market demands and accelerating the global adoption of EVs; and (v) adopting sustainable practices to address the environmental impact by recycling old batteries and developing eco-friendly materials, such as green mining and biodegradable solvents, to reduce the ecological footprint of battery production. In the context of this review, specifically, regarding battery technology development, companies with research and development centers are the driving force behind advancements and progress in EV battery technology. While each company may focus on different aspects, two main strategies can generally be distinguished for improving battery performance: external improvement and internal improvement of the battery.

Before delving into the details of this subject, it is essential to understand the composition of what is commonly referred to as an EV battery (Figure 5A,B). At the most basic level are the cells.<sup>28</sup> These components contain the elements responsible for generating electric current: the electrodes (anode and cathode), the electrolyte, the separator, and the current collector. However, a single cell cannot adequately power a vehicle, which is why multiple cells are connected in a module to increase the overall power.<sup>28</sup> A module contains a number of cells coupled to connectors and an initial cooling system, along with a metal or plastic casing designed to protect the assembly from mechanical impact. Finally, multiple modules are grouped into a pack, which features a second cooling system, sensors, and a battery management system.<sup>28</sup> In simple terms, the cell represents the chemical aspect of a battery, the module represents the mechanical aspect, and the pack represents the electronic aspect (Table 1).

It is challenging to know in detail the advances and improvements of each company. However, based on publicly available information, it is possible to get a general idea of each company's strategy. In the case of CATL, external improvements can be seen in how batteries are organized. For an EV, it is important to maximize power while reducing the vehicle's mass, so that minimal energy is spent moving the vehicle itself. A good external enhancement involves optimizing the battery's components to fit the most cells and the fewest secondary elements. This is one of the promises of cell to pack (CTP) technology, which eliminates modules and directly integrates cells into the pack, a method adopted by CATL.<sup>22,29</sup> This company claims that CTP technology has enabled it to increase the volumetric energy density of its batteries by 55% with the first generation and by 72% with the third generation, reaching 250 Wh kg<sup>-1</sup> for its Qilin NMC battery.<sup>29</sup>

LG Energy Solution is also working on similar processes, such as lamination and stacking, which produce very thin cells and reduce the gaps between them.<sup>30</sup> This concept is used in the Ultium battery.<sup>23</sup> However, the company seems to be focusing on an internal improvement that promises to be the future of batteries: the solid-state battery (SSB). Unlike a conventional battery with a liquid electrolyte, the SSB features a solid electrolyte (SE), making it nonflammable, more resistant, and safer.<sup>31</sup> LGES is working in particular on polymer and sulfide bases, and seems confident enough to announce marketing dates of 2026 and 2030, respectively.<sup>23</sup>

BYD is also working on SSBs but is best known for its LFP batteries, which are based on Li and phosphate. The company claims that this process makes them extremely flame-retardant and resistant to severe shocks, such as those experienced in road accidents, thanks in particular to the absence of cobalt, which reduces the risk of overheating.<sup>32</sup>

It is worth noting that the development of EVs is significantly influenced by the cost, weight, and volume of their batteries. These factors directly impact the overall efficiency, affordability, and performance of EVs, thus playing a crucial role in their widespread adoption. Currently, lithium batteries are paramount in EVs, comprising a substantial portion of the EV's cost, weight, and volume. Typically, the battery pack accounts for about 30%-40%



**FIGURE 5** (A) Composition of an electric vehicle battery pack. Reproduced under the terms of the CC-BY open access license.<sup>26</sup> Copyright 2020, The Authors. (B) States of battery integration. Reproduced under the terms of the CC-BY open access license.<sup>27</sup> Copyright 2023, The Authors.

of the total cost of an EV. This underscores the importance of efficient battery recycling; we will talk about recycling in a later section. On the other hand, developing low-cost batteries, such as low-material-cost lithium batteries and other metal-based batteries, is important. For instance, sodium-ion batteries are estimated to be 30% cheaper than their Li-ion counterparts. The battery pack's weight can range from 20% to 30% of the vehicle's total weight, and it occupies a significant portion of the vehicle's volume. Lighter batteries can improve vehicle efficiency and increase driving range; compact batteries allow for more flexible vehicle designs and can free up space for passengers and cargo. Innovations in battery chemistry, such as the use of silicon in anodes, are aimed at increasing energy density and reducing weight (equal to smaller battery). Advances in SSB technology are expected to reduce the weight and volume of batteries, making them more compact without compromising on energy capacity. In summary, reducing the cost, weight, and volume of batteries through advancements in battery chemistry and materials is essential. Innovations in battery design aim to increase energy density, allowing more energy to

be stored in smaller, lighter packs. This not only improves the range and efficiency of EVs but also enhances the vehicle's overall usability and comfort. This is particularly important for integrating batteries into various types of vehicles, from small city passenger cars to large trucks and buses. These improvements, combined with other factors mentioned earlier in this section, such as manufacturing optimization, supply chain management, market strategies, and addressing environmental impact, are crucial for enhancing overall vehicle performance.

#### 3 | EV BATTERIES: AN IMPRESSIVE RANGE OF TECHNOLOGIES

### 3.1 | Lithium batteries

# 3.1.1 | Lithium-ion batteries and general overview

Awarded the Nobel Prize in Chemistry in 2019, LIBs are the best-known and most widely used batteries by the gen-



**FIGURE 6** Structure of lithium-ion batteries (LIB). Reproduced with permission.<sup>33</sup> Copyright 2010, Elsevier.

eral public. They also serve as good representation of the overall functioning of today's electrochemical batteries.

To generate electricity, a LIB relies on a chemical reaction.<sup>34,35</sup> First of all, it is important to note that in the scenario depicted in Figure 6, Li is not present in its pure form. Instead, it is combined with graphite on the anode side (the negative pole of the battery), and reacts with a metal oxide, such as cobalt, on the cathode side (the positive pole of the battery). At the atomic level, lithium is intercalated, meaning that a layer of lithium atoms is sandwiched between two layers of other materials. In the case of the anode, the structure alternates between layers of lithium and graphite (LiC<sub>6</sub>), while in the case of the cathode, the structure alternates between layers of lithium, cobalt, and oxygen (LiCoO<sub>2</sub>).<sup>36</sup>

The anode and cathode are in contact with an electrolyte, a liquid solution containing solvents and salts.<sup>37</sup> When in contact with the electrolyte, the anode tends to oxidize, meaning that the Li atom within the  $\text{LiC}_6$  loses an electron, and becomes a positive  $\text{Li}^+$  ion. Conversely, at the cathode, a reduction occurs, meaning that the lithium atoms in  $\text{LiCoO}_2$  try to gain an electron. However, these reactions remain minor because the electrons released by the anode cannot escape, and the cathode does not have access to a sufficient quantity of electrons to initiate its reduction. It is also important to note that, at the start of the reaction, the cathode has a very low lithium content, with a composition closer to  $\text{CoO}_2$ .

When the anode and cathode are connected by a conducting wire, electrons flow between them. The anode, seeking to release electrons, transfers them through the wire to the cathode, which accepts them. This flow of electrons constitutes the electric current that is utilized at the battery's terminals. Consequently, the cathode becomes negatively charged (due to the electrons it gains), while the anode becomes positively charged (due to the electrons SusMat WILEY 18 of 30

it loses).<sup>38</sup> Naturally, these components strive to achieve a neutral charge by balancing their positive and negative charges. To accomplish this, the anode releases positively charged Li ions into the electrolyte as it loses electrons, while the cathode draws Li ions from the electrolyte as it gains electrons (Table 2). The quantities of electrons and ions lost by the anode equal those gained by the cathode, ensuring a balanced reaction.

In the case of a Li-based battery, the electrolyte solvent must not be aqueous under any circumstances. This is crucial because lithium reacts vigorously upon contact with water, producing dihydrogen ( $H_2$ ), a highly flammable gas that contributes significantly to the risks associated with Li batteries. Even atmospheric humidity can trigger this reaction. Moreover, opting for a non-aqueous electrolyte introduces effective but flammable alternatives, adding another layer of safety concern.

However, the most severe danger arises during battery overheating, which can lead to deformation, ignition, or even explosion. This phenomenon, known as thermal runaway (Figure 7A), poses the greatest risk.<sup>39–41</sup>

As mentioned previously, Li is a rare and increasingly expensive element. In the case of the Li-ion cobalt/graphite battery, cobalt is also known for its toxicity.<sup>42</sup> Moreover, the supply of cobalt may begin to deplete as there are no new mines opening to further sustain the growing demand. At present, cobalt ore reserves stand at 6.68 million tons, with countries such as the Democratic Republic of Congo, Indonesia, and Australia being the most abundant.<sup>43</sup> The shortage and price increase of cobalt may occur within approximately the next 30 years. If no alternative is found, it is likely to become a bottleneck in the automotive market. Despite these drawbacks, this type of battery offers proven technology, long service life, and unparalleled performance in terms of energy density due to lithium's characteristics.<sup>44</sup> Additionally, the graphite anode provides a unique balance with its relatively low cost, abundance, and high energy density.

To maintain the competitiveness of the cathode while reducing overall battery costs, a widely adopted technique involves incorporating nickel (Ni) and manganese (Mn) into the lithium (Li) and cobalt (Co) cathode structure.<sup>45</sup> Instead of solely atomic layers of Li, Co, and O (LiCoO<sub>2</sub>), layers of Li, Ni, and O (LiNiO<sub>2</sub>), and Li, Mn, and O (LiMnO<sub>2</sub>) are utilized. Each of these additional elements plays a crucial role in the cathode's behavior: cobalt stabilizes the layered structure, reduces impedance, and enhances electrode conductivity, while nickel has been shown to increase cathode energy density.<sup>46</sup> However, due to their similar atomic sizes, excessive nickel can replace lithium during battery recharge cycles, leading to degradation over time.<sup>47</sup> Similarly, Mn contributes to cathode stability but in excess can disrupt the layered structure.<sup>44</sup>

#### TABLE 2 Redox reaction in lithium-ion batteries (cobalt/graphite).

Reduction (cathode)	Oxydation (anode)	Complete equation
$CoO_2 + Li^+ + e^- \rightarrow LiCoO_2$	$\text{LiC}_6 \rightarrow \text{C}_6 + \text{Li}^+ + \text{e}^-$	$\text{LiC}_6 + \text{CoO}_2 \rightleftharpoons \text{C}_6 + \text{LiCoO}_2$



**FIGURE 7** (A) Evolution of a thermal runaway. Reproduced under the terms of the CC-BY open access license.<sup>41</sup> Copyright 2017, The Authors. Comparison of  $\text{LiNi}_x \text{Mn}_y \text{Co}_{1-x-y} \text{O}_2$  (NMC) and  $\text{LiFePO}_4$  (LFP) batteries in terms of energy density (B) and cost (C). Reproduced under the terms of the CC-BY open access license.<sup>49</sup> Copyright 2019, The Authors. (D) Advantage of LFP battery over NMC battery in terms of number of charge cycles. Reproduced under the terms of the CC-BY open access license.<sup>50</sup> Copyright 2020, The Authors.

The challenge for NMC batteries lies in achieving a balance among cost, performance, and stability, although they do not fully mitigate the safety issues inherent to lithium, despite improved thermal stability.<sup>48</sup>

As previously mentioned, while NMC batteries dominate the EV market, another competitive technology is LFP batteries. Like their NMC counterparts, LFP batteries utilize iron (Fe) and phosphate in their cathodes (LiFePO<sub>4</sub>). Despite slightly higher costs and lower performance (Figure 7B,C), LFP batteries are distinguished primarily by their structural stability, resulting in significantly longer service lives compared to NMC batteries around 7000 charge cycles versus 1000 under typical conditions (Figure 7D).<sup>49,50</sup> Furthermore, their superior thermal resistance enhances safety during operation.<sup>51</sup>

Regarding anode, graphite offers numerous advantages and has become the preferred choice for practical applications of LIBs. However, non-carbon-based anode materials have also been widely explored and developed in research and the market. For instance, the commercialized spinel  $Li_4Ti_5O_{12}$  anode materials exhibit significant advantages over graphite anodes in terms of safety, cycle life, and rate performance.<sup>52</sup> The high Li removal potential (~1.55 V vs.  $Li/Li^+$ ) of  $Li_4Ti_5O_{12}$  material prevents Li deposition and solid electrolyte interphase (SEI) formation, ensuring high safety. Additionally,  $Li_4Ti_5O_{12}$  has zero strain characteristics, and its volume change remains minimal even after incorporating three Li ions in the lattice, which enhances its cyclic stability.<sup>53</sup> Moreover, the Li-ion diffusion coefficient of Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> is as high as  $2 \times 10^{-8}$  cm<sup>2</sup> s<sup>-1</sup>, an order of magnitude higher than that of graphite. Consequently, Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> also demonstrates excellent rate performance. Although the capacity of Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> material is only ~175 mAh g<sup>-1</sup>, it can serve as a substitute material for graphite anode in battery systems that require fast charging, long lifespan, and high safety.<sup>54</sup>

In addition to commercial graphite and Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> materials, silicon (Si) is a promising anode material, which can react with Li to form alloys, with a theoretical capacity of up to 4200 mAh g<sup>-1</sup>.<sup>55,56</sup> However, significant volume changes occur before and after alloy formation, severely damaging the material structure, which negatively impacts cyclic performance and renders it impractical for use.<sup>57,58</sup> To improve the performance of Si materials, several strategies have been developed, including preparing nano silicon, silicon oxides, and synthesizing composites of Si and carbon and other substances.<sup>59,60</sup> For example, Si nanospheres with a pomegranate-like structure were assembled using double-layer carbon coating.<sup>61</sup> This hierarchical structure effectively stabilizes and limits the growth of SEI films, resulting in extremely high stability during the cycling process. After 1000 cycles, the capacity of the anode material remains at 1160 mA h  $g^{-1}$ , with a capacity retention rate of 97%. Furthermore, using alginate salts to replace carboxymethyl cellulose and polyvinylidene difluoride as electrode binders for nano Si can obtain highly stable Si anodes.<sup>62</sup> After 100 cycles at a current density of 4200 mA  $g^{-1}$ , the capacity remains basically unchanged at approximately 2000 mAh  $g^{-1}$ . This approach results in significantly higher capacity and reversibility compared to using the original binders under the same conditions.

#### 3.1.2 | Lithium metal batteries

Unlike LIBs, which benefit from established technology and decades of experience, lithium metal batteries (LMBs) are still in the research and development stage.<sup>63–66</sup> However, their immense potential suggests that once matured, this technology could secure a significant position in the EV battery market.

LMBs differ from LIBs in that they feature an anode composed solely of metallic lithium, rather than aiming to intercalate lithium into a layered matrix such as graphite. This design allows lithium to be utilized to its fullest potential without performance compromise from other materials. Consequently, LMBs offer impressive energy density, owing to lithium's high specific capacity of 3862 26924552 2024, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/sus2.234 by Ecole De Technologie Superieur, Wiley Online Library on [28/10/2024], See the Terms and Conditions (https://onlinelibrary.wiley.com/enorthic) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

mAh g<sup>-1</sup> compared to graphite's 372 mAh g<sup>-1</sup>, and its extremely low anode electrode potential (-3.04 V vs. the standard hydrogen electrode [SHE]).<sup>46,67</sup> However, achieving this level of performance greatly depends on the optimal choice of the cathode. Despite these advantages, LMBs face several challenges, including the high cost of lithium, thermal runaway risks, and a critical issue that hinders their competitiveness with LIBs: dendrite formation (Figure 8).<sup>68,69</sup>

In theory, the Li ions released by the cathode should deposit uniformly on the anode to maintain a flat anode/electrolyte interface. However, observations from the visualized cell indicate that Li does not distribute uniformly on the electrode surface. Instead, it forms through the nucleation ("nuc") and growth ("grow") of dendrites (Figure 8A-D).<sup>70</sup> Nucleation itself involves additional energy barriers, resulting in growth kinetics  $(k_{\text{grow}}^0)$  being significantly faster than nucleation kinetics  $(k_{nuc}^0, k_{nuc}^0)$  $k_{grow}^0$ ). As dendrites nucleate, the primary reaction pathway of the cathode shifts from nucleation to growth, where additional Li preferentially deposits on the surface of the dendrites rather than forming new nucleation sites. Following a polarity switch, the main reaction pathway at the anode involves the rapid dissolution of dendritic Li. As the amount of active Li in the dendrites approaches zero, a characteristic increase in the battery's polarization voltage occurs due to the slow dissolution transition from the surface to the kinetics. The electro-dissolution of the body leads to the formation of pits. During polarity changes, Li will preferentially nucleate and form dendrites on these uneven surfaces. These issues that are detrimental to battery life are still being studied to find feasible solutions.<sup>71</sup> As shown in Figure 9, several strategies exist to address such challenges, including designing 3D structured anodes, fabricating artificial SEI layers, introducing electrolyte additives, and employing solid-state electrolytes (SSEs).72-75 Our team has employed novel electrolyte additives that are simple and low-cost and can potentially extend the battery operating temperature range.<sup>64</sup> Importantly, without changing the current manufacturing process of batteries, adding a small amount of additives can improve the safety and energy density of the batteries.

On the other hand, one of the cathodes of LMBs is inexhaustible: the atmosphere. Indeed, the Li-air battery exploits the oxygen present in the atmosphere ( $O_2$ ) to create a redox reaction with Li.<sup>77</sup> The virtual absence of a physical cathode makes this battery extremely lightweight, which could be of great interest in the transport sector. However, the development of a protective membrane that allows oxygen to pass through while blocking any contact between Li and water in the atmosphere (which could lead to a violent reaction) needs significant improvement.<sup>78</sup>





**FIGURE 8** Changes in cell polarization (top) are correlated with a schematic representation of morphology (middle; color-coded to match the appropriate reaction pathway as described in Scheme) and energy barrier diagrams (bottom). EL-a and EL-b are the top electrode and the bottom electrode respectively. This phenomenon is observable at four distinct points in the voltage trace: (A) Commencement of the half-cycle, characterized by the kinetically slow process of dendrite nucleation. (B) Minimum cell polarization during the half-cycle, showcasing dendrites on both electrodes and representing kinetically fast reaction pathways. (C) Maximum cell polarization, signifying the removal of "active" Li from dendrites; electro-dissolution transitions to kinetically slow bulk dissolution. (D) Second decrease in cell polarization, marking the point where pitting becomes the kinetically slow process. Reproduced under the terms of the CC-BY open access license.<sup>70</sup> Copyright 2016, The Authors.

Similarly, the cathode can also be composed of sulfur (S), creating a Li-sulfur (Li-S) battery. One of the advantages of this choice is the moderate atomic mass of this element, which, combined with the energy density of Li, gives a very attractive total battery energy density of around 550 Wh kg<sup>-1</sup>.<sup>79</sup> Moreover, the price of sulfur is quite low compared to cobalt. Nevertheless, the greatest challenge for Li-S batteries is the change in cathode volume during use.<sup>79</sup> When discharged, this electrode can increase its volume by up to 80%, resulting in high mechanical stress and possible rupture of the battery envelope. The low conductivity of sulfur is also a concern, but this disadvantage can be mitigated with a carbon coating.<sup>80</sup>

Despite all our efforts and technical expertise, the use of Li in these batteries does not address the intrinsic problems associated with this metal, such as its cost, scarcity, and behavior at high temperatures. Consequently, alternative elements are being considered to replace this precious material while maintaining its unparalleled performance.

#### 3.2 | Alternative materials to lithium

There are numerous candidates for replacing Li in batteries (Figure 10). Although developing alternative battery

meet the needs of energy storage devices. These systems must serve as viable substitutes or supplements to Li battery systems. Based on practical requirements such as cost, environmental protection, service cycle, and performance, batteries should possess at least five basic characteristics: low cost, low hazard potential, high energy density, long cycle life, and high-power density. Specifically, the selection and matching of cathodes, anodes, and electrolytes should meet the following criteria: (i) use of inexpensive, abundant, and easily synthesized materials, (2) employ environmentally friendly and safe materials that are stable in air, non-flammable, and low in toxicity, (3) ensure materials have high capacity and reasonable operating voltage, (4) achieve good reversibility of anode/cathode reactions and acceptable electrolyte electrochemical stability, and (5) enable rapid electrochemical reaction kinetics to reduce polarization, minimize energy loss during charging/discharging, and shorten charging time. Currently, in addition to LIBs, batteries using different reaction cations that meet the above conditions have been developed, including those based on Na (sodium), K (potassium), Mg (magnesium), Ca (calcium), Zn (zinc), and Al (aluminum) ions. As shown in Figure 10, the high abundance

systems is necessary and urgent, it is crucial to first clarify the desired characteristics of such battery systems to



FIGURE 9 Schematic diagram of different strategies to inhibit dendrite formation. Reproduced with permission.<sup>76</sup> Copyright 2021, Elsevier.

of these metals in the Earth's crust makes their potential applications very promising.

#### 3.2.1 Aluminum

Al is positioning itself as an interesting alternative due to its low-cost, abundance, safety, and scalability.<sup>82</sup> This battery uses a graphite cathode that stores ions through intercalation, similar to LIBs, and an Al metal anode.<sup>83</sup> However, the reaction that powers this battery is quite different from that of a LIB. There is no unidirectional flow of Al ions from one electrode to another, making the term "Al-ion" somewhat misleading.

The electrolyte in this battery is made up of aluminum chloride ions, such as  $AlCl_4^-$  or  $Al_2Cl_7^-$ . It is the movement and transformation of these ions that drive the reactions needed to create an electric current (Figure 11A).<sup>83</sup>

Compared with the most relevant battery technologies for non-aqueous and aqueous media, the working principle of Al-ion battery (AIB) is slightly different. However, AIBs can meet the practical requirements for new batteries, such as high power density (4 kW kg<sup>-1</sup>), cycle

life (20 000 cycles), and high safety (due to ionic liquids and Al), which shows promising prospects (Figure 11B).<sup>84</sup> Some AIBs boast an energy density of 40 Wh kg<sup>-1</sup> (partly due to the lightness of Al) and up to 7500 cycles without any decline in overall battery capacity.<sup>85</sup> Furthermore, Al is much more abundant and affordable than Li. However, the ionic electrolyte used is still very expensive, and the larger diameter of Al atoms compared to Li atoms can lead to breakage of the graphite matrix during intercalation.83

Similar to LMBs, it is possible to use ambient air as a cathode. Coupled with Al's low mass density, this approach presents a battery of incomparable lightness, possibly providing an impressive energy density of 8100 Wh kg<sup>-1</sup> and almost total recyclability.85-87 However, the major drawbacks in this scenario are the corrosion of Al and self-discharge (e.g., due to hydrogen evolution) in the alkaline electrolyte, which makes the battery difficult to recharge, and the expensive cathode membrane required to filter the oxygen required for the reaction.<sup>86,87</sup> The issue of Al corrosion can be addressed by using various Al alloys (e.g., Al-0.5Mg-0.02Ga-0.1Sn-0.5Mn and Al-5Zn-1Mg-0.02In-0.05Ti-0.1Si).86-89 Nevertheless, like most alternative materials to LIBs, Al batteries are free from the specific safety issues associated with Li.

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**FIGURE 10** Elemental abundance, ionic radius, standard electrode potential (vs. the standard hydrogen electrode [SHE]), along with gravimetric and volumetric capacities of distinct metal anodes. Reproduced with permission.<sup>81</sup> Copyright 2019, Wiley.



FIGURE 11 (A) Movement of Al ions within an Al-ion battery during the charge cycle. Reproduced under the terms of the CC-BY open access license.<sup>83</sup> Copyright 2017, The Authors. (B) The performances of Al-ion, Li-ion, and lead-acid batteries are compared across five key parameters: energy density, specific power, cost, cycle life, and safety. Reproduced with permission.<sup>84</sup> Copyright 2019, Royal Society of Chemistry.

### 3.2.2 | Sodium

While Al draws its strength from its lightness, Na draws it from its abundance.<sup>86</sup> Not only is it the sixth most abundant element in the Earth's crust, but its presence in seawater increases its abundance and accessibility 10-fold.<sup>15,90</sup> Moreover, Na shares many properties with Li, making the Na-ion battery (SIB) process virtually identical to that of the LIB.<sup>91</sup> Battery giants such as NorthVolt and BYD have recently announced their ambitious SIB plans. Northvolt's validated cell uses a hard carbon anode and a Prussian White-based cathode, rather than Li, Ni, Co, mak-

ing it safer, more cost-effective, and more sustainable. On November 18, 2023, BYD announced a partnership with Huaihai Holding Group to jointly build an SIB factory with an annual capacity of 30 GWh. The main challenge for SIBs is their low energy density, about 100 Wh kg<sup>-1</sup>, due to the low conductivity of Na (Figure 12A).<sup>92,93</sup> To address this, as shown in Figure 12B, all materials used in this technology are selected to enhance performance. This includes using a graphite cathode (despite its tendency to form agglomerates that impact long-term performance) and adding elements such as Ni, Fe or Mn to boost the Na cathode (with a composition modeled on NMC batteries).<sup>82,94</sup> SIBs





**FIGURE 12** (A) Comparison of energy density of Na-ion battery with lead-acid battery and lithium-ion battery (LIB). Reproduced with permission.<sup>92</sup> Copyright 2021, Wiley. (B) Electrode materials and corresponding electrochemical performances in current Na-ion battery technologies. Reproduced with permission.<sup>95</sup> Copyright 2013, Royal Society of Chemistry.

can operate with an aqueous electrolyte, making them very low toxicity, but this choice reduces their electrical potential due to the electrochemical stability window of water. To enhance performance, a non-aqueous electrolyte with esters as solvents can be employed, but this comes at the expense of the environmental aspect.<sup>93</sup>

#### 3.2.3 | Magnesium

To conclude this list of materials that could potentially replace Li, magnesium (Mg) stands out due to its balanced attributes.<sup>96</sup> While it may not excel in terms of abundance, cost, or energy density, it also does not exhibit significant weaknesses in these areas, making it a compelling compromise (Figure 13A,B). This is primarily attributed to its high theoretical capacity and high standard reduction potential, which is slightly lower than that of lithium and sodium.<sup>97</sup>

The first reports on rechargeable Mg batteries appeared in the early 1990s. Following investigations into Mgion conductive electrolytes by Gregory et al., Aurbach et al. achieved a significant technological breakthrough by establishing a prototype for Mg batteries.<sup>98,99</sup> In 2008, Aurbach and his team further advanced the field by exploring all-phenyl-complex electrolytes, which expanded the potential window-an essential step marking the second breakthrough in rechargeable Mg batteries.<sup>100</sup> Since 2010, the development of Mg-based batteries has seen explosive growth. Researchers have developed non-nucleophilic electrolytes and high-voltage electrolytes compatible with sulfur cathodes and oxide cathodes, respectively.<sup>101–103</sup> In 2019, Davidson et al. identified the formation of dendrites in Mg metal anodes within specific electrolytes.<sup>104</sup> Up to this point, innovative material design concepts have

been extensively employed, emphasizing the importance of understanding Mg dendrites. However, research in this field is still in its early stages. For Mg-ion batteries, a more promising cathode has not yet been identified, even though the anode performs well in a two-dimensional layered matrix such as zinc or lithium, and the electrolyte is often aqueous.<sup>97</sup> The primary research focus remains on preventing the corrosion of the anode by the aqueous electrolyte, which reduces the battery's overall efficiency to 60%.

Mg-metal batteries are also under investigation, though suitable cathodes are still lacking. Moreover, an insulating film tends to form on the surface of the Mg anode, severely limiting the life cycle of this battery despite the absence of dendrites.<sup>97</sup>

### 3.2.4 | Other alternatives

As previously mentioned, several alternatives to Li exist. Therefore, this section concludes by highlighting a few candidates that, while less recognized, merit inclusion in this discussion. Firstly, the potassium-ion battery (PIB) presents an intriguing alternative in terms of performance. Its theoretical energy density closely rivals that of LIBs, attributed to the small Stokes radius of K ions (K<sup>+</sup>: 3.6 Å, Li<sup>+</sup>: 4.8 Å, Na<sup>+</sup>: 4.6 Å) in organic electrolytes.<sup>106,107</sup> This characteristic facilitates efficient ion transport within the electrolyte, offering a high potential energy density due to lower redox potential. Moreover, unlike Li, which has uneven and limited geographical distribution, potassium resources are abundant and cost-effective.<sup>108</sup> However, significant technological challenges hinder commercialization, particularly due to the larger size and molecular





**FIGURE 13** (A) Current design strategies for rechargeable Mg-based batteries. (B) The price, volumetric capacity, and charge density of various metal materials for energy storage technologies. Reproduced with permission.<sup>105</sup> Copyright 2021, American Chemical Society.

weight of  $K^+$  ions, leading to volume fluctuations in the electrode. This impacts the reversibility of chemical reactions and transport efficiency, resulting in relatively modest overall performance. Currently, the energy density based on the K cathode remains below 500 Wh kg<sup>-1</sup>, while for K<sup>-</sup> ion full batteries, PIB achieves below 400 Wh kg<sup>-1</sup> (based on the total weight of positive and negative electrode materials).

Calcium metal batteries (CMBs) have recently garnered attention as a promising option due to their ideal theoretical redox potential (-2.87 V vs. SHE), which is slightly higher than that of lithium (-3.04 V vs. SHE) and potassium (-2.37 V vs. SHE). Additionally, CMBs boast a high capacity of 1337 mAh g<sup>-1</sup>/2072 mAh cm<sup>-3</sup>.<sup>109-114</sup> The Ca ion, with twice the number of charges compared to alkaline metal ions, experiences significantly lower polarization than other multivalent metal ions (Mg<sup>2+</sup>, Al<sup>3+</sup>), facilitating fast and efficient charge transfer.<sup>111,115</sup> Furthermore, given that Ca is the third most abundant metallic element in the Earth's crust, the cost of Ca resources is only

one-eighth that of Li.<sup>115</sup> However, despite their attractive theoretical energy density and relatively low cost, CMBs are still in the initial research stage and face numerous challenges. One primary challenge arises from variations in the oxidation-reduction potential of Ca metal anodes, which has been measured by different research groups using different electrolytes or even the same electrolyte, indicating poor stability in the oxidation-reduction potential of Ca metal anodes.<sup>116</sup> Another challenge stems from early investigations by Aurbach et al., who explored various organic electrolytes, including acetonitrile, tetrahydrofuran,  $\gamma$ -butyrolactone, and propylene carbonate as solvents, and  $Ca(ClO_4)_2$ ,  $Ca(BF_4)_2$ , etc., as salts in CMBs. It was discovered that Ca metal fails to form an effective passivation layer in these organic electrolytes, leading to non-reversible electrodeposition. Although recent efforts have identified feasible electrolytes achieving reversible deposition/dissolution of Ca metal, the persistent issue of serious polarization hysteresis during Ca plating/stripping remains.<sup>117-120</sup> Addressing these challenges requires the



**FIGURE 14** (A) Zn-ion battery with Zn metal anode and intercalative cathode along with Zn dendrites and corrosion in Zn metal anode. Reproduced with permission.<sup>137</sup> Copyright 2020, Wiley. (B) Schematic illustration of battery configurations for Zn-air batteries. Reproduced with permission.<sup>138</sup> Copyright 2023, Wiley.

design of numerous reliable experimental devices under unified technical parameters, specifications, and standards to elucidate the composition/function of the SEI formed in the electrolyte. This effort aims to understand the energy storage mechanism of intercalated or converted cathodes, ultimately advancing CMB technology toward practical realization.

Another promising alternative is Zn, which is again based on the same criteria of cost and safety when compared to Li.<sup>121</sup> Recently, novel materials have been developed to serve as electrode components, which demonstrate excellent battery cycling and rate performances.<sup>122,123</sup> However, the Zn-ion battery (Figure 14A), which uses Zn<sup>2+</sup> ions for its operation and a Zn metal anode, has yet to reach a breakthrough for wide application.

Nonetheless, similar to the other metals mentioned earlier, the cathode can be composed of dioxygen, resulting in a Zn-air battery (Figure 14B).<sup>124–126</sup> This combination shows impressive results, such as an energy density of 200 Wh kg<sup>-1</sup> with an aqueous electrolyte.<sup>123,127–129</sup> In recent years, the cycling performance has also been significantly improved by developing various stable bifunctional cathode materials to enhance the reactions of the oxygen reduction reaction (for discharging) and oxygen evolution reaction (for charging process).<sup>125,130–134</sup> However, to promote the technology for commercialization, special attention needs to be paid to the formation of dendrites, the charging voltage being higher than the discharging voltage causing an overall efficiency of 70%, and a progressive deformation of the anode, which never returns exactly to its initial state.<sup>130,132,135</sup> Despite these challenges, the theoretical energy density of the Zn-air battery, at 1218 Wh kg<sup>-1</sup>, remains impressive, prompting further research.<sup>134–136</sup>

Dual-ion batteries (DIBs) store families of cations and anions separately during charge and discharge cycles. The characteristics of dual ion insertion and dissolution in these batteries provide several advantages: (1) the high potential for anionic insertion into graphite results in ultra-high voltage (graphite/graphite and Li/graphite types exceeding 4.4 V). (2) Substituting the metal-rich positive electrode in traditional LIBs (e.g., LiCoO<sub>2</sub>/graphite batteries) with graphite materials, which constitutes about half of the total cost, can substantially reduce costs by approximately 30%.<sup>139,140</sup> (3) Replacing cathodes containing non-renewable elements such as Ni, Co, and Mn contributes significantly to sustainability and a reduced  $CO_2$  footprint. (4) Due to the absence of available oxygen in the graphite cathode, DIBs inherently ensure safety, markedly decreasing safety risks.

However, the insertion and extraction of anions within the graphite structure primarily occur at high voltages, especially when compared to  $\text{Li}/\text{Li}^+$  exceeding 4.5 V.<sup>141,142</sup> This leads to more pronounced oxidative degradation reactions within the DIB, resulting in a decrease in coulombic

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efficiency (CE). Utilizing an excessively high cutoff voltage can exacerbate electrolyte oxidation, leading to the formation of a substantial layer at the top of graphite, and hindering effective anion transfer at the interface.<sup>143</sup> Additionally, according to the DIB mechanism, graphite and corresponding electrolytes undergo significant volume changes during the charging and discharging processes. While graphite anodes exhibit a relatively moderate volume enhancement of about 10% during Li-ion insertion, introducing more voluminous anions can promote graphite expansion to nearly 140%.<sup>144</sup> Therefore, a careful balance between higher capacity and higher CE may be necessary to explore a new DIB system that meets the requirements of higher capacity energy storage systems.

Finally, the anode-free LMB is a compromise. Given that lithium is a precious resource, this concept proposes using only a lithium cathode and a simple collector on the anode side, rather than two electrodes both made of lithium metal. Although theoretically feasible, the CE of an anode-free battery determines its reversibility and actual cycling performance due to the absence of excess lithium in the battery system. Meanwhile, the reaction between the electrolyte and highly active lithium metal or dead lithium during the cycling process still restricts the CE of the anode-free battery.<sup>145–148,150</sup> In recent years, researchers have explored various strategies to improve CE, mainly focusing on the following aspects: (1) optimizing electrolytes, including SEs and liquid electrolytes, to achieve uniform nucleation Li metal; (2) constructing new 3D fluid collectors or applying functional coatings to regulate Li deposition; (3) designing favorable solutions (such as circulation, temperature, external pressure) to extending the cycling life.<sup>148,149,151-160</sup> Although these strategies have significant effects in extending battery life, recent progress has not yet been summarized and the internal mechanism for extending battery life remains unclear. Currently, there is a surge of interest in anode-free Li batteries, but there is a lack of systematic overviews.

#### 3.3 | Solid-state battery

As mentioned in Section 2.1.1, the electrolytes used in LIBs are the primary cause of their significant safety issues, particularly their flammability and the potential for thermal runaway. SSBs address these problems fundamentally by introducing a non-flammable SE that is more resistant to mechanical stress compared to its liquid counterpart.<sup>161–164</sup> Within the SE family, inorganic solid electrolytes (ISEs) are typically single-ion cationic conductors, which lead to higher charge transfer efficiency compared to traditional liquid electrolytes with low transfer numbers.<sup>165</sup> Specifically, the Li conductivity of ISEs



**FIGURE 15** Unresolved challenges for solid-state Li metal batteries. Reproduced with permission.<sup>170</sup> Copyright 2021, Wiley.

(sulfides) usually ranges between  $10^{-6}$  and  $10^{-4}$  S cm<sup>-1</sup>, which is 2-4 orders of magnitude lower than that of liquid electrolytes (10<sup>-2</sup> S cm<sup>-1</sup>).<sup>166</sup> Various structural tuning methods, from atomic to mesoscale, can effectively elevate ion conductivity to an acceptable level of  $10^{-3}$ S cm<sup>-1</sup>. Notably, the  $Li_{10}GeP_2S_{12}$  family, a sulfide electrolyte, demonstrates increased ionic conductivity of Li up to  $10^{-2}$  S cm<sup>-1</sup>, comparable to liquid electrolytes at room temperature.<sup>167</sup> Furthermore, the solid-state treatment process can employ bipolar electrodes to stack batteries within a single package, resulting in reduced package volume and increased energy density.<sup>168</sup> This trend is further accelerated by the emergence of wearable technology. Finally, this technology holds the promise of fully utilizing the potential of Li metal, with its specific capacity of 3862 mAh g<sup>-1</sup>, potentially leading to much higher energy densities than those offered by current Li batteries.63,169

However, there are several new challenges to developing a marketable SSB (Figure 15).<sup>170,171</sup> Although less severe than those of liquid-electrolyte lithium metal batteries, the non-lithium anodes and cathodes of SSBs are composed of composite materials with particle sizes ranging from nanometers to micrometers. These materials, such as cathode active material (CAM), SSE, conductive additives, and binders, typically have various solid–solid interfaces, including CAM–SSE, CAM–binder, CAM–conductive additives, SSE–binder, and SSE–conductive additive interfaces.<sup>172</sup> The chemical and electrochemical reactions, poor contact, and mechanical instability at these solid-solid interfaces pose significant challenges for the development of SSBs. These challenges include harmful interface reactions, contact losses that hinder ion transport, and the fracture of active materials.<sup>173</sup> Currently, vehicles containing Li metal polymer batteries, such as Bolor é Bluecar, are on the market.<sup>174</sup> However, the poor oxidation stability of polyethylene oxide-based polymers limits the selection of positive electrodes, particularly layered transition metal oxides with high operating voltages (>3.8 V). As a result, these polymers can only be matched with low-voltage materials such as LFP. Consequently, even though lithium metal anodes are used, the energy density of these battery systems is usually lower than that of state-of-the-art LIBs. Ceramic or glass inorganic electrolytes have also been incorporated into small lithium batteries with thin-film structures. However, the vacuum deposition process used in their manufacturing is expensive and not easily scalable. To achieve the cost, energy density, and performance goals required for EVs, it is necessary to redesign these devices using low-cost manufacturing processes that incorporate thick (~100 µm) composite cathodes to enhance energy density.

# 3.4 | Status of different types of batteries in EV applications

Electric vehicles use a variety of battery types, each with its own set of advantages and disadvantages. Table 3 provides an overview of the different types of batteries used in EVs, highlighting their advantages, disadvantages, and specific applications.

Overall, each of these battery types offers unique benefits and poses certain challenges. The choice of battery for an EV depends on factors such as cost, energy density, safety, and the specific application requirements of the vehicle. Currently, LIBs and SSBs are the most popular choices for EV applications due to their superior energy density, long cycle life, and relatively low weight. However, ongoing research and development are exploring the potential of other battery types to improve safety, and performance while reducing costs in EVs. In particular, battery technologies are evolving to enhance the efficiency and sustainability of EVs.

#### 4 | EV BATTERIES: END OF LIFE TREATMENT

As with large-scale energy production, a technology cannot be considered truly sustainable unless its entire life SusMat WILEY 18 of 30

cycle is virtuous and sustainable. As shown in Figure 3, the production and use of an EV does emit a certain amount of  $CO_2$ -eq, but this is much less than that of a combustion vehicle.<sup>5,175</sup> However, this figure does not account for the end-of-life phase of the vehicle, particularly the battery. There are three primary ways to reduce the environmental impact of a battery at the end of its useful life: reuse, remanufacturing, and recycling (Figure 16).<sup>176,177</sup>

#### 4.1 | Reuse

Before discussing the principle of reuse, it is important to understand that an EV battery is typically considered unfit for its original purpose when it loses between 20% and 30% of its initial capacity.<sup>178</sup> This means that if the battery is dismantled directly, 70%-80% of its capacity remains unused. For example, in 2019, a Canadian household consumed an average of 90.5 GJ of energy, equivalent to 2.87 kW in 1 h. A small EV, such as the Mini Cooper SE, has a 29 kWh battery, while a larger vehicle such as the GMC Hummer EV pickup has a 200 kWh battery.<sup>179</sup> This means that the battery of the Mini Cooper SE could power a Canadian home for approximately 10 h, while the battery of the Hummer EV could do so for around 70 h. Although these calculations are based on the performance of new batteries, they highlight that repurposing EV batteries for national energy production is a viable option. Additionally, using these batteries as energy storage systems capable of supplying power grids during short time windows aligns well with the intermittent nature of renewable energy sources.180

In a Nature Communications article published in January 2023 by Xu et al., a complete simulation of the introduction of the second-life system for EV batteries was presented (Figure 17A).<sup>178</sup> This study estimates global energy consumption in 2050 to be between 3.4 and 19.2 TWh, depending on the scenario. It demonstrates that second-life EV batteries alone could meet this demand by delivering between 15 and 32 TWh of energy. The study considers four scenarios for the evolution of battery technology, the gradual replacement of the global car fleet with EVs, and battery degradation over time. However, Figure 17A assumes a 100% participation rate in the second-life system for batteries.

To be effectively implemented, this system relies on a series of classification stages based on the condition of the various battery components. When a battery pack is deemed unsuitable for vehicle operation, it is tested, and its performance is measured. If the results are favorable (around 80% of the initial capacity), the



TABLE 3 Overview of the different types of batteries used in electric vehicles (EVs).

Battery types	Advantages	Disadvantages	Applications in EVs
Lead-acid batteries	<ul><li>Low cost</li><li>High power output</li><li>Mature technology</li></ul>	<ul> <li>Heavy and bulky</li> <li>Low energy density</li> <li>Fast discharge rates</li> <li>Limited life cycle, especially with deep discharge cycles</li> </ul>	Mostly used in starter motors and auxiliary applications in EVs rather than as the primary power source.
Nickel-cadmium batteries	<ul><li>Robust and durable</li><li>Long life cycles</li><li>Higher depth of discharge</li><li>Recyclable</li></ul>	<ul> <li>Environmental hazards (toxic Cd)</li> <li>Memory effect</li> <li>Less favorable energy density</li> </ul>	Due to their disadvantages, nickel-cadmium batteries are less favored in modern EVs.
Nickel-metal hydride batteries	<ul> <li>Cost-effective</li> <li>Long life cycles</li> <li>High safety (resistant to overcharge/discharge)</li> <li>Tolerance to unfavorable conditions</li> </ul>	<ul> <li>High self-discharge rate</li> <li>Memory effect</li> <li>Less favorable energy density</li> </ul>	Previously popular used in hybrid electric vehicles due to their safety (i.e., less prone to overheating and thermal runaway compared to LIBs); however, they are largely being phased out, due to their limitations.
Li-ion batteries	<ul> <li>High energy density</li> <li>Long cycle life</li> <li>High charge and discharge efficiency</li> <li>Mature technology</li> <li>Low self-discharge rates and minimal memory effect</li> </ul>	<ul> <li>High cost</li> <li>Potential safety concerns</li> <li>(risk of overheating and fire)</li> <li>Environmental issues</li> </ul>	Widely used in most modern electric cars, including models from Tesla, Nissan, and Chevrolet, etc., due to their high energy density, good power-to-weight ratio, and long cycle life.
Na-ion batteries	<ul> <li>Cost-effective</li> <li>Smaller carbon footprint versus LIBs</li> <li>High safety</li> <li>Robust capacity retention</li> </ul>	<ul> <li>Lower energy density compared to LIBs</li> <li>Still in the early stages of development</li> </ul>	Offer a cost-effective, environmentally friendly alternative to LIBs, especially for short-range and compact EVs, due to the abundance and low cost of sodium.
Solid-state batteries (including Li-metal batteries)	<ul> <li>Higher energy density</li> <li>Long lifespan</li> <li>Improved safety (less prone to leakage, overheating, or catching fire)</li> <li>Faster charging</li> </ul>	<ul> <li>High production costs</li> <li>Difficulty in manufacturing scaling up</li> <li>Still in the development stage</li> <li>Not widely commercialized</li> </ul>	Considered the future of EV batteries due to their potential for higher energy density and improved safety, current development by different car makers, such as QuantumScape, Toyota, Nissan BMW, etc.
Aluminum-ion batteries	<ul> <li>Low cost</li> <li>High capacity and energy density</li> <li>Faster charging</li> <li>High safety and stability</li> <li>Super long lifespan</li> </ul>	<ul> <li>Corrosion</li> <li>Still in the development stage</li> <li>Scaling up for big manufacturing</li> <li>Market adoption</li> </ul>	Present a promising and potentially game-changing technology, due to low cost, high safety, fast charge, long cycle life, etc.
Other batteries (K, Mg, Ca, Zn, etc.)	<ul><li>Low cost</li><li>Improved safety</li><li>High energy density</li><li>Long lifespan</li></ul>	<ul> <li>Material design</li> <li>Still in the development stage</li> <li>Scaling up for big manufacturing</li> <li>Market adoption</li> </ul>	Emerging as promising alternative for next-generation EVs, in consideration of cost-effectiveness, high safety, and environmentally friendly. However, they encounter many challenges.

Abbreviation: LIB, lithium-ion battery.

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FIGURE 16 Life cycle of an electric vehicle battery, including reuse, reconditioning, and recycling. Reproduced under the terms of the CC-BY open access license.<sup>176</sup> Copyright 2021, The Authors.



FIGURE 17 (A) Projections of the capacity delivered by all electric vehicle batteries in second use over the year 2050. Reproduced under the terms of the CC-BY open access license.<sup>178</sup> Copyright 2023, The Authors. (B) Selection process for end-of-life batteries. Reproduced under the terms of the CC-BY open access license.<sup>176</sup> Copyright 2021, The Authors.



 TABLE 4
 Conflicting goals for battery production and reconditioning/recycling.<sup>176</sup>

Objectives for producing a battery	Objectives for reconditioning/ recycling a battery
<ul><li>High safety</li><li>Lightness</li></ul>	<ul> <li>Good accessibility of components for rapid disassembly</li> </ul>
<ul> <li>Low cost</li> <li>Minimization of empty spaces</li> </ul>	<ul> <li>Use of easily removable components so as not to damage the assembly</li> </ul>

pack is directly reused for energy storage at national, urban, or private network scales. The remaining packs are dismantled into modules, which are then individually tested and either assimilated into smaller storage units, such as electric bicycles, or reassembled into a new pack if they are in good condition. Finally, the rest of the modules are completely dismantled and recycled, if possible. Figure 17B schematically summarizes this process.<sup>176</sup>

### 4.2 | Remanufacturing

The principle of remanufacturing is based on a simple observation. As explained in Section 1.3, an EV battery comprises numerous components, including cells, modules, sensors, and cooling systems. When an entire pack can no longer perform its functions, it is usually due to a few defective components affecting the operation of the whole system. Many other parts, however, may still be in perfect condition. Therefore, it would be imprudent to completely dismantle the pack and recycle its elements without first attempting to recover components that are still suitable for use in a new pack.<sup>181</sup>

Although seemingly straightforward, this process presents significant technical challenges.<sup>182,183</sup> Reconditioning is intricate, time-consuming, and highly expensive due to the wide variety of battery packs on the market.<sup>176</sup> Additionally, EV batteries are not currently designed to be fully disassembled with easy access to all components. It is important to understand that the specifications of pack producers differ from those of entities seeking to recondition or recycle them, as shown in Table 4.

To achieve full operational efficiency and viability, battery disassembly should eventually be fully automated.<sup>176,184</sup> However, the wide variety of existing designs makes it challenging to develop a universal disassembly protocol and process.<sup>185</sup> To make reconditioning more feasible, a standard imposed on battery designers could facilitate dismantling. Additionally, incorporating artificial intelligence to handle the diverse disassembly requirements appears to be a promising solution.

## 4.3 | Recycling

Recycling is widely recognized as a key method for enhancing the sustainability of a product's life cycle. This is especially true for EV batteries, given the high cost of the materials used in their production (Figure 18A).<sup>176</sup>

The challenges of recycling are multifaceted. According to a 2020 article by Beaudet et al., the primary challenge is to manage the risks associated with storing unusable components, particularly due to their toxicity and flammability.<sup>186</sup> Additionally, burying these components can lead to soil and groundwater contamination. The second challenge is to reduce the carbon footprint of EVs, a significant portion of which is attributed to the mining of rare earth elements. The third challenge focuses on decreasing the dependence of EV production on mining operations, which, as previously noted, exploit rare metals such as lithium without adequately addressing their gradual depletion. Moreover, the environmental impact of these mines is inconsistent with the energy transition goals supported by EVs. The fourth challenge is more practical: reducing vehicle costs, such as achieving a 30% reduction in battery pack prices.<sup>187</sup> The penultimate challenge highlights the potential for recycling batteries to foster a new local economy, benefiting states through increased tax revenues, providing jobs for workers, and reducing transportation costs for materials.<sup>188</sup> Finally, the last challenge addresses a geopolitical aspect: reducing reliance on specific suppliers whose monopolies pose a risk to the supply chain if they fail to meet growing demand.

Recycling can take various forms, and its outcomes can be integrated at different stages of a product's life cycle.<sup>190–193</sup> However, three main approaches can be isolated: recovery, resynthesis, and refunctionalization.<sup>189</sup> When evaluated against 10 techno-economic criteria (Figure 18B), refunctionalization shows the highest average score (3.4/5). Nonetheless, it faces technical constraints and feasibility issues. For instance, battery components suitable for this approach must be in relatively good condition, which is not always guaranteed. Both resynthesis and recovery methods have identical scores (2.8/5), but recovery is rated at the extreme end of the scale, while resynthesis shows more variability. On a more technical note, three industrial processes are currently employed for battery recycling: pyrometallurgy, hydrometallurgy, and direct recycling. Table 5 compares these processes in terms of their respective strengths and weaknesses.<sup>194</sup> Table 5 compares them in terms of their respective strengths and weaknesses.



FIGURE 18 (A) Breakdown of the total cost of an electric vehicle battery. Reproduced under the terms of the CC-BY open access license.<sup>176</sup> Copyright 2021, The Authors. (B) Three main forms of battery recycling, compared on the basis of 10 criteria. Reproduced under the terms of the CC-BY open access license.<sup>189</sup> Copyright 2020, The Authors.

In summary, there are various recycling and reuse technologies for waste LIBs, each with its specific application scenarios and advantages and disadvantages. At present, the wet process is the predominant method used by most recycling enterprises of Li batteries, while technologies such as pyrometallurgical and mechanochemical methods are difficult to use alone. However, these methods can serve as effective pre-leaching strengthening techniques to promote metal extraction and reduce reagent consumption. Therefore, the selective leaching of valuable metals, which selectively extracts Li from the host material while preserving the internal structure of the cathode material, is an ideal technology for simplifying subsequent separation processes. However, conventional pre-treatment

steps are still needed to collect cathode materials from different types of waste LIBs. Based on their reducibility, two recovery systems, low acid liquid-phase reaction and solid-phase reaction under mechanochemical forces, were established to efficiently, mildly, and cost-effectively recover materials, and the synergistic mechanisms of these technologies were analyzed. It is worth highlighting the role of green solvents in battery recycling. Green solvents such as organic acids, ionic liquids, deep eutectic solvents, and supercritical fluids are characterized by minimal toxicity and biodegradability. Their use significantly reduces environmental hazards compared to traditional solvents. Utilizing a green solvent mixture system can effectively integrate the advantages of various solvents. For instance,

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### TABLE 5 Comparison of the three main industrial recycling techniques.

Recycling method	Advantages	Disadvantages
Pyrometallurgy	<ul> <li>Adaptable; applicable to various battery chemistries and configurations</li> <li>No sorting or additional mechanical pre-treatment is required</li> <li>Achieving a high recovery rate for metals, such as Co, Ni, and Cu</li> <li>Established technology; can be employed with existing pyrometallurgical facilities</li> </ul>	<ul> <li>Incapable of recycling Li, Al, or organic materials</li> <li>Unable to process LFP batteries</li> <li>Requires costly gas clean-up to prevent toxic air emissions</li> <li>Energy intensive</li> <li>Capital intensive</li> <li>Further refinement is essential to obtain elemental metals from metal alloys produced in the smelting process</li> </ul>
Hydrometallurgy	<ul> <li>Suitable for any battery chemistry and configuration</li> <li>Versatile in separation and recovery processes to selectively target metals</li> <li>High recovery rates, for example, for Li</li> <li>High purity of products (suitable for cathode precursors, etc.)</li> <li>Energy efficient</li> <li>No emission of air pollutants</li> </ul>	<ul> <li>Crushing of battery cells is required (raising safety concerns)</li> <li>Cathode structure is broken down by acid</li> <li>Results in a significant volume of process effluents to be managed through treatment, recycling, or disposal</li> <li>Not economical for LFP batteries</li> <li>No recovery of anode materials (e.g., graphite and conductive additives)</li> <li>High operating cost</li> </ul>
Direct recycling	<ul> <li>Retains valuable cathode structure</li> <li>Allows for the retrieval of practically all battery components, such as the anode, electrolyte, and foils</li> <li>Well-suited for LFP batteries</li> <li>Energy efficient</li> <li>Convenient for recycling manufacturing scraps</li> </ul>	<ul> <li>Demands intricate mechanical pre-treatment and separation processes</li> <li>The reclaimed material might not match the performance of virgin material or could become outdated before its market introduction</li> <li>Amalgamation of cathode materials could diminish the value of the recycled product</li> <li>Regeneration processes are yet to be developed</li> <li>Direct recycling has not expanded to industrial level</li> </ul>

Abbreviation: LFP, LiFePO<sub>4</sub>.

organic acids can be effectively incorporated into the acidleaching process of industrial production, demonstrating significant developmental potential. A specific example is the combination of 20 g  $L^{-1}$  of oxalic acid with 2.5 mol  $L^{-1}$ H<sub>2</sub>SO<sub>4</sub>, which achieves leaching rates of 99.26%, 98.41%, 96.95%, and 97.54% for Li, Ni, Co, and Mn, respectively.<sup>199</sup> Emerging organic solvents such as palm oil, palm fatty acid distillate, and 1-octanol offer low solubility and temperature flexibility, making them suitable for various extraction conditions. By leveraging these green solvents, battery recycling can become more efficient and environmentally friendly.

## 5 | CONCLUSION AND PERSPECTIVES

Battery-powered EVs are one of the key technologies of the 21st century. It is crucial to ensure that these new vehicles do not replicate the path of combustionpowered vehicles—efficient but environmentally harmful technologies reliant on non-renewable resources.

1. The upstream activities involved in battery material production and battery manufacturing consume a large amount of energy. Particularly for certain battery materials, the energy required for mining and other processes (such as primary extraction and refining) is often supplied by power grids that rely heavily on fossil fuels or by direct combustion of fossil fuels (e.g., natural gas, diesel). This situation needs to change, with renewable energy sources being utilized for mining and upgrading activities. To achieve this, it is essential to evaluate the entire lifecycle of these batteries from multiple perspectives (e.g., consumers, car manufacturers, battery designers) throughout the production, manufacturing, and usage phases of EVs. Additionally, finding environmentally friendly and sustainable solutions that balance factors such as manufacturing costs, driving performance, safety, and recycling is crucial for making informed decisions.

- 2. Although EV manufacturers are spread worldwide, the majority of production and refining activities are concentrated in the Asia-Pacific region. Companies in this region range from leading suppliers of lithium battery systems, such as CATL, to established car manufacturers such as BYD. The dynamic environment in this region rewards advanced and sophisticated technologies, pushing industry leaders to explore new research and development areas. To achieve efficient and sustainable goals for the global EV industry, these industry leaders need to share and exchange their advancements with EV companies in other regions. Through collaboration and joint efforts, they can drive progress toward a more sustainable future.
- 3. Despite various types of batteries, LIBs still dominate the market. Li, a crucial battery material, has significant safety risks and performance issues, which can lead to battery fires or even explosions. This has led to the development of substitutes using metals such as aluminum, sodium, zinc, or magnesium, as well as entirely different technologies such as SSBs. While the results from these alternatives are promising, their current maturity is low, and there are still implementation challenges to overcome. Therefore, it will take considerable time and development for these technologies to achieve commercial competitiveness with lithium-based batteries.
- 4. Designing EV batteries with modularity and ease of recyclability in mind is crucial for balancing economic feasibility and environmental protection. By making batteries modular and easily removable, manufacturers can facilitate the recycling process and enhance the efficiency of recovering valuable materials. Integrating principles such as second life, reconditioning, and comprehensive recycling strategies into battery design can significantly reduce the environmental impact of EVs over their entire lifecycle. This approach not only supports sustainability goals but also promotes a circular economy where materials are reused and recycled, minimizing waste and resource depletion.
- 5. With the popularization of EVs, the role of Li batteries has become increasingly significant, making battery recycling a critical issue. Achieving high recovery rates for essential materials such as lithium, cobalt, and nickel is crucial for effective battery recycling but remains challenging due to the complex composition of batteries, which includes various metals, electrolytes, and separators. The harmful environmental impact and high cost of cobalt have led to efforts to gradually phase it out from cathode chemistry. However,

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considering the environmental impact of disposing of end-of-life batteries and the risks associated with material demand in the market, significant efforts are required to develop suitable recycling processes that are low-cost, flexible, and more efficient. Despite these feasibility challenges, recycling presents a promising solution to reduce costs and environmental impact by creating a closed loop in the Li battery value chain.

6. The development cycle of the automotive industry is long, suggesting that it is unlikely for any "nextgeneration" battery technology to replace current LIB technology in the short term, such as within the next decade. However, once matured, these technologies are expected to exhibit the following characteristics: (1) improved performance, with significant advancements in charging time and overall lifespan; (2) increased energy density, capable of storing two to three times more electricity compared to previous technology, with a potential battery lifespan three times longer, all without a substantial increase in size or weight; (3) advanced battery management, utilizing various technologies for enhanced battery management, such as fiber-optic online monitoring for real-time charging status updates; and (4) modular design, battery pack designs featuring modular characteristics to facilitate easier installation and improve recycling efficiency. Looking forward, it is foreseeable that the forthcoming generation of commercial EV batteries will represent an evolved iteration of current LIBs.

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#### CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest. Dr. Shuhui Sun is an Associate Editor of SusMat and a coauthor of this article. To minimize bias, he was excluded from all editorial decision making related to the acceptance of this article for publication.

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