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# Sustainable Electric Vehicle Batteries for a Sustainable World: Perspectives on Battery Cathodes, Environment, Supply Chain, Manufacturing, Life Cycle, and Policy

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**Li-ion batteries (LIBs) can reduce carbon emissions by powering electric vehicles (EVs) and promoting renewable energy development with grid-scale energy storage. However, LIB production and electricity generation still heavily rely on fossil fuels at present, resulting in major environmental concerns. Are LIBs as environmentally friendly and sustainable as expected at the current stage? In the past 5 years, a skyrocketing growth of the EV market has been witnessed. LIBs have garnered huge attention from academia, industry, government, non-governmental organizations, investors, and the general public. Tremendous volumes of LIBs are already implemented in EVs today, with a continuing, exponential growth expected for the years to come. When LIBs reach their end-of-life in the next decades, what technologies can be in place to enable second-life or recycling of batteries? Herein, life cycle assessment studies are examined to evaluate the environmental impact of LIBs, and EVs are compared with internal combustion engine vehicles regarding environmental sustainability. To provide a holistic view of the LIB development, this Perspective provides insights into materials development, manufacturing, recycling, legislation and policy, and beyond. Last but not least, the future development of LIBs and charging infrastructures in light of emerging technologies are envisioned.**

## 1. Introduction

Li-ion batteries (LIBs) have achieved remarkable success in electric vehicles (EVs), consumer electronics, grid energy storage, and other applications thanks to a wide range of electrode materials that meet the performance requirements of different application scenarios. In recent years, EVs have become the major market of LIBs. Multiple countries have announced that they would progressively replace internal combustion engine vehicles (ICEVs) with EVs over the next few decades to satisfy the net-zero carbon emission criteria.[1] Examining the lifetime carbon emissions of EVs and ICEVs is imperative to demonstrate the validity of switching from ICEVs to EVs. Vehicle lifetime emissions include emissions during battery raw materials processing and battery manufacturing for EVs, vehicle manufacturing, and the well-to-wheel (WtW) process. For ICEVs, the WtW process relates to fuel production and vehicle use, whereas for EVs, it refers to electricity generation and distribution. The International Energy Agency (IEA) established the Stated Policies Scenario (STEPS) and the Sustainable Development Scenario (SDS) to evaluate the emissions under different circumstances.[2] The STEPS reflects the legislated or announced policies and targets for emissions by governments around the world.[2] It performs evaluations based on accomplished emission reduction goals. The STEPS is relatively conservative and does not presume all announced goals to be reached. In contrast, the SDS has set stricter emission standards and assumes all net-zero emission pledges are achieved. We chose the STEPS in the following comparison to avoid overstating the benefits of EVs.

Based on the STEPS, the global WtW greenhouse gas (GHG) emissions for EVs are all lower than their replaced ICEV counterparts for light-duty vehicles (LDVs), buses, trucks, and two/three wheelers (**Figure 1**a). By replacing ICEVs with EVs, the global net GHG emissions avoided during the WtW process are predicted to be 51.9 million tonnes of carbon dioxide equivalent (Mt $CO_2$-eq), 55.1 Mt $CO_2$-eq, 58.5 Mt $CO_2$-eq, 69.3 Mt $CO_2$-eq, 91.5 Mt $CO_2$-eq, and 120.9 Mt $CO_2$-eq, respectively, at every two year interval from 2020 to 2030 (Figure 1a).[3] The benefits of emission reduction offered by EVs will be more prominent in the SDS.[3] Taking into account vehicle manufacturing, mineral processing, and battery production, mid-size EVs still have lower GHG emissions than ICEVs throughout

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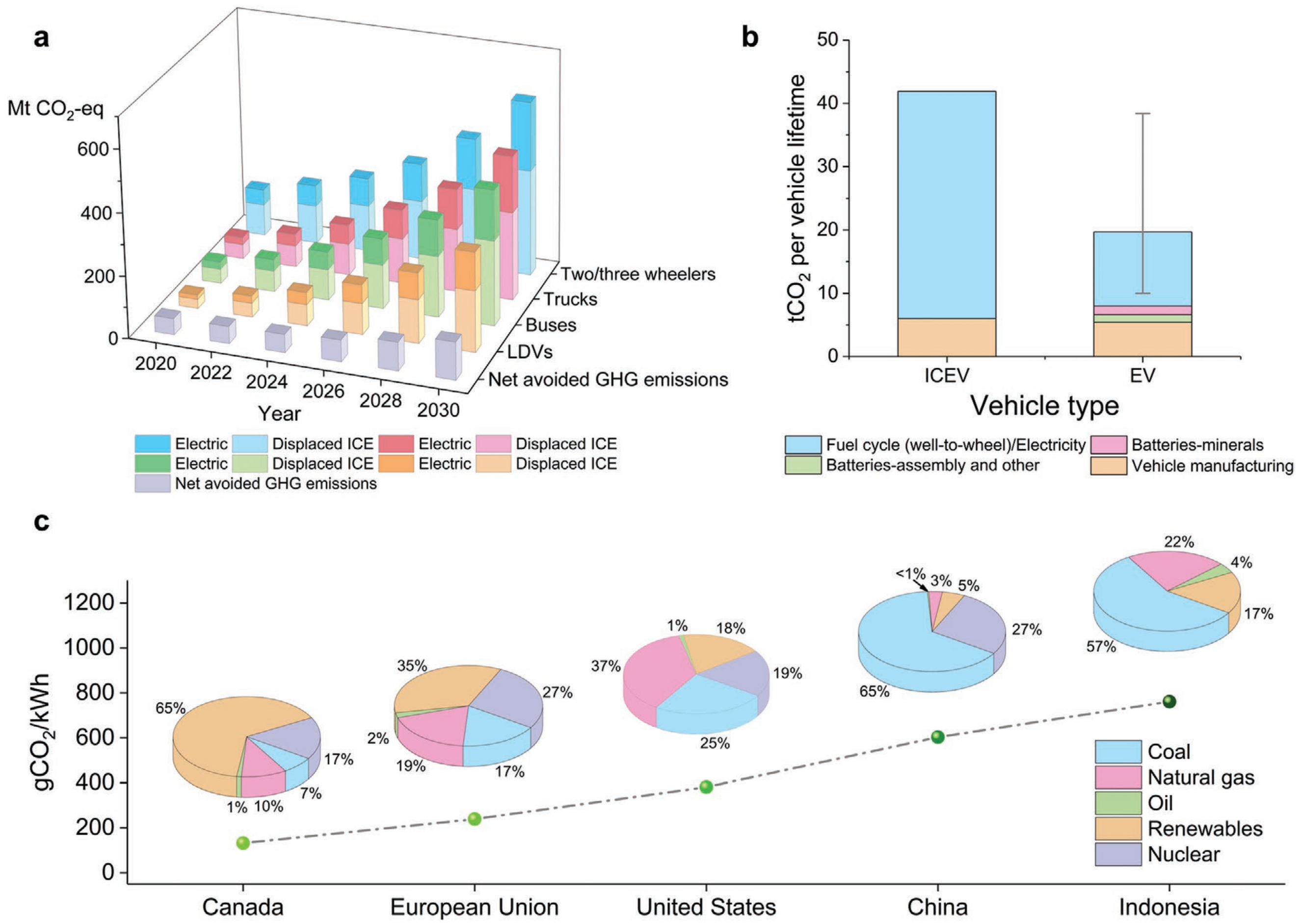


**Figure 1.** Environmental impact of EVs and ICEVs. a) GHG emissions of EVs and displaced ICEVs and net avoided GHG emissions from 2020 to 2030 in the Stated Policies Scenario.[3] b) Comparison of life-cycle GHG emissions of a mid-size EV and ICEV.[4] c) Electricity generation mix and emissions from electricity generation in selected regions in 2019.[4] Overall, EVs have less GHG emissions than displaced ICEVs, after considering the processes of mineral mining, battery assembling, vehicle manufacturing, and vehicle operation. However, the emissions of EVs largely depend on the source of electricity generation. The countries that heavily rely on fossil fuels have more emissions. To reduce the emissions of EVs, optimizing the electricity generation mix and developing renewable energy are of crucial importance. The figure is made based on the data from refs. [3] and [4].

the life cycle (Figure 1b).[4] However, electricity generation leads to large variations of the total lifetime emissions (Figure 1b).[4] Specifically, the electricity generation source can greatly affect total emissions (Figure 1c).[4] As of 2019, renewable energy sources account for 65% of power generation in Canada, and nuclear energy accounts for 17%, resulting in comparatively low $CO_2$ emissions of 132 $gCO_2$ $kWh^{-1}$ (Figure 1c). In Indonesia, however, fossil fuels account for 83% of electrical generation. The emission during electricity generation in Indonesia was 761 $gCO_2$ $kWh^{-1}$, which was nearly six times higher than that of Canada (Figure 1c).[4] The total emissions during electricity generation increase as the countries or regions rely more on fossil fuels (Figure 1c). To summarize, EVs generate lower lifetime emissions than their ICEV counterparts do. This is in good agreement with the literature.[5] In addition, increasing the share of renewable energy sources in the electricity generation mix can further enhance the environmental benefits of vehicle electrification. LIBs, being one of the most critical components of EVs, play a significant role in determining the long-term sustainability of the EV industry.

The development of LIBs needs to be driven in a more sustainable direction to satisfy the rising energy demand and simultaneously meet the criteria for net-zero carbon emissions. In this regard, it is crucial to assess the environmental impact and energy consumption of LIBs throughout the life cycle. Cathodes are a critical component of batteries and contribute considerably to the production cost of LIBs.[6] At present, cathodes still rely on scarce metals substantially, such as Ni and Co. These metals are less favorable in the cathode market due to their limited reserves and high price. Advancement of LIBs at the cathode materials level is required to balance sustainability, cost, and performance. More practical factors in industrial manufacturing need to be considered upon the commercialization of research-level materials and designs. Reliable LIB manufacturing requires the support from a robust supply chain. Evaluating the global LIB supply chain and manufacturing is critical to comprehend LIB development. In addition, massive LIBs will reach their end-of-life in the foreseeable future, given the substantial and increasing number of EVs around the world. Processing, repurposing, and recycling of these used batteries will be a pressing topic. Developing recycling technologies that are both economically and environmentally favorable can largely enhance the sustainability of LIBs. Recycling can in turn reduce the energy consumption and emissions during the virgin battery production. Furthermore, government policies and legislation can have a significant impact on the supply chain, manufacturing, and recycling of LIBs. Here, we systematically evaluate the environmental impact of LIBs, cathode chemistry, battery manufacturing and supply chain, battery recycling, and government policies regarding their roles in the sustainable development of LIBs. Last but not least, we conceive a visionary scheme for future LIB development and charging infrastructure construction.

## 2. Status of LIB Development

### 2.1. Sustainability Assessment

The capability of LIBs to power EVs and store electricity generated from renewable energy sources has led to the erroneous public perception that LIBs are "zero-emission" technologies. In reality, LIBs, just like other batteries, are essential tools to store and release electrical energy. The fact that LIB production is energy- and resource-intensive, and that current electricity generation still heavily relies on fossil fuels, can potentially cause environmental concerns. Moreover, disposal of spent LIBs without recycling could be detrimental to the environment. Life cycle assessment (LCA) is a systematic analysis of the potential environmental impacts of products, processes, or services throughout their entire life cycle.[7] It is generally accepted as a standard methodology to quantify the environmental influences of the production, usage, and recycling of LIBs.

Many LCA studies have been conducted to assess the environmental impacts of the production of different LIB chemistries including $LiFePO_4$ (LFP), $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC), $LiMn_2O_4$ (LMO), and $LiNi_xCo_yAl_{1-x-y}O_2$ (NCA), but their results are far from agreement.[8] The reported cradle-to-gate GHG emissions for battery production (including raw materials extraction, materials production, cell and component manufacturing, and battery assembling as shown in **Figure 2**) range from 39 to 196 kg $CO_2$-eq per kWh of battery capacity with an average value of 110 kg $CO_2$-eq per kWh of battery capacity.[8b–8j] The discrepancies in GHG emissions in prior studies can be attributed to a variety of reasons such as different battery chemistries, regions of manufacturers, assumptions in LCA models, and modeling approaches to estimating energy demand in battery manufacturing.[8g,j] LFP, NMC, and LMO are the most studied battery chemistries in LCA mainly due to their popularity in the current EV market and availability of the manufacturing data from the battery industry. Peters et al. critically analyzed a wide array of LCA studies of battery production and found that LMO has the lowest GHG emissions among the three battery chemistries, followed by NMC and LFP based on the averages of results from published studies.[8g] Hao et al. examined GHG emissions from LIB production in China and reported a similar conclusion that the production of LMO automotive LIBs leads to the lowest GHG emissions and the production of LFP leads to the highest GHG emissions.[8f] GHG emissions of LIB production could also vary with the locations of manufacturers due to different quality of electricity used and electricity generation source.[8f,j] For example, the production of LFP, NMC, and LMO

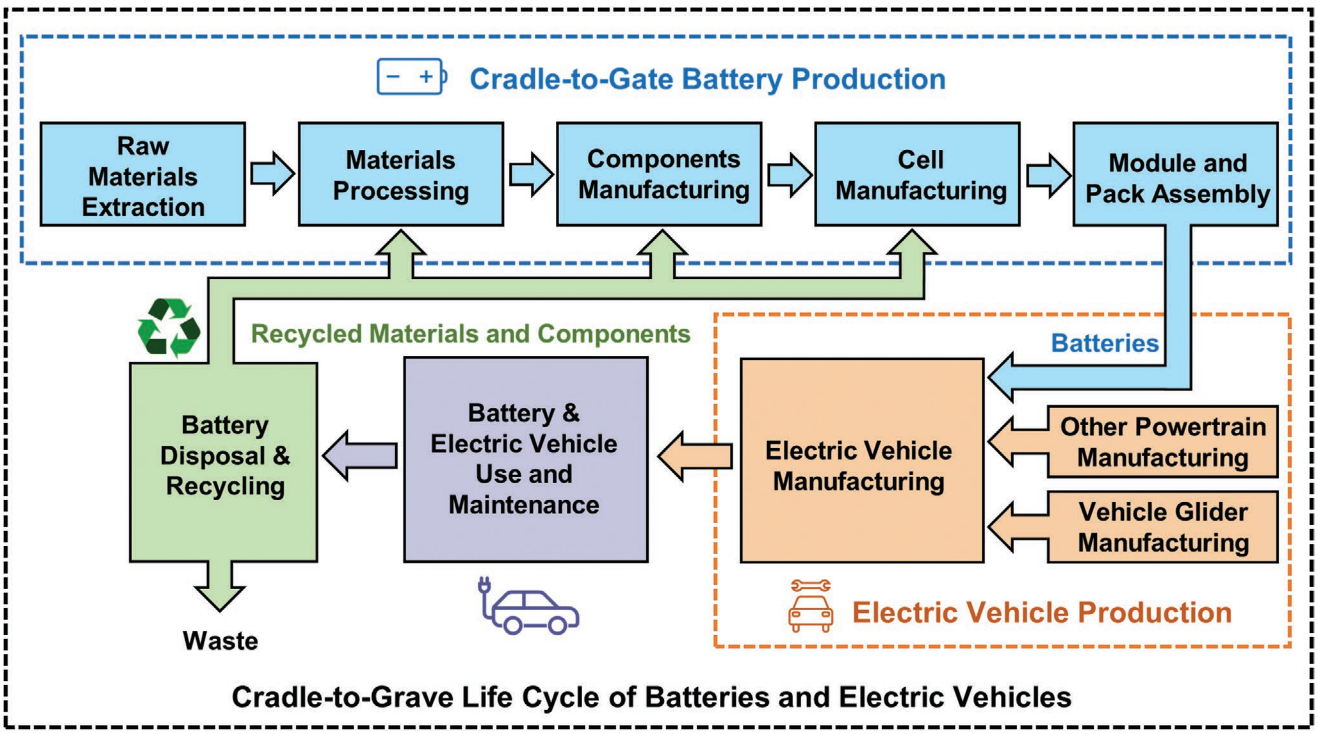


**Figure 2.** Major cradle-to-grave life cycle stages for batteries and EVs, including cradle-to-gate battery production, EV production, the use and maintenance of batteries and EVs, and battery disposal and recycling. The color-coded dashed rectangles show different system boundaries.

batteries in China has nearly three times higher emissions than that in the US because electricity generation in China relies more on coal (Figure 1c).[8f] Besides battery chemistries and regions of manufacturers, the approach for modeling the battery manufacturing process to estimate energy demand also contributes to the wide discrepancies of the LCA results.[8g,9] Prior studies used two modeling approaches to estimate the total energy demand in battery manufacturing: 1) the bottom-up approach which uses data from theoretical simulations or lab-scale experiments of the critical processes in the manufacturing line, and 2) the top-down approach which uses data from a real manufacturing plant. It was found that the latter approach usually results in a much higher estimated energy demand in battery manufacturing compared to the bottom-up approach. For example, using the top-down approach, Kim et al. assessed the cradle-to-gate GHG emissions from mass-produced LIB used in the Ford Focus EVs based on the primary energy data from the battery cell and pack industries.[8c] In Kim et al.'s study, the estimated GHG emission from the cell and pack manufacturing process is 65 kg $CO_2$-eq per kWh of battery capacity, which is over one order of magnitude higher than the range of 1.5–1.9 kg $CO_2$-eq estimated by other studies using the bottom-up approach.[8c] Furthermore, the real industrial manufacturing data are critical in LCA to obtain legitimate and practically informative results.

Prior LCA studies agreed that battery electric vehicle (BEV) production generates more GHG emissions than ICEV production does. Hawkins et al. found that the cradle-to-gate GHG emissions associated with EV production are almost twice that associated with ICEV production, and battery production contributes 35% to 41% of the GHG emissions from EV production.[8b] This finding is supported by Kim et al., who reported a 39% increase in GHG emissions switching from ICEV to EV in vehicle production.[8c] Hao et al. also indicated that there is around 30% increase in GHG emissions for EV production compared to that of traditional vehicles.[8f] The higher GHG emissions associated with EV production in comparison to ICEV production is mainly because of the high GHG emissions from the production of battery, generator, and motor, i.e., the electric powertrain system. Although EV production has higher environmental impacts, extending the system boundary to include the use phase of EV (as shown in Figure 2) leads to a clear advantage of EV compared to ICEV because EVs offer higher powertrain efficiency and zero tailpipe emissions. Hawkins et al. found that light-duty EVs powered by the present European electricity mix offer a 10% to 24% decrease in GHG emissions relative to ICEVs assuming 150 000 km lifetime of EVs, and extending the lifetime of EVs to 200 000 km boosts the environmental benefits of 27% to 29% decrease in GHG emissions.[8b] In another study of the comparison between EVs and ICEVs, it was found that transport services with an EV result in a 35.6% (in percentage of the EV, 37 700 kg $CO_2$-eq) decrease in GHG emissions compared to an ICEV,[8a] which is in general agreement with Hawkins et al.'s study. In addition, electricity generation source plays a vital role in GHG emissions. EVs powered by coal electricity can lead to an increase in GHG emissions compared to ICEVs.[8h] Therefore, it is vital to promote clean electricity sources to power EVs to maximize their environmental benefits. Although global warming potential (GWP), as expressed in GHG emissions, is the most frequently assessed environmental impact category (EIC) in the majority of LCA studies, other critical EIC, such as abiotic depletion (ADP), acidification, eutrophication (EP), human toxicity (HTP), and ozone depletion (ODP), are equally, sometimes even more, important to assess the environmental impacts of EV technology. Studies have raised concerns that production and use of EVs could potentially lead to increases in HTP, EP, and ADP categories, mainly emanating from the EV supply chain.[8b,h,10]

Recycling spent LIBs reduces the demand for virgin raw materials and the toxic waste entering the environment, which can potentially decrease the environmental impacts of the battery life cycle.[8j] However, the environmental benefit of LIB recycling depends on the recycling route and battery chemistry. In a comprehensive study conducted by Ciez and Whitacre, the authors examined the GHG emissions associated with recycling NMC, LFP, and NCA battery cells using three recycling routes: pyrometallurgical, hydrometallurgical, and direct cathode recycling.[8i] It was found that for NMC and NCA cells, there is a median reduction (roughly 0.2–1 kg $CO_2$-eq per kg of battery) in GHG emissions from hydrometallurgical and direct cathode recycling. However, pyrometallurgical recycling of NMC and NCA cells results in net increases in GHG emissions compared to no recycling, mainly because of the high energy consumption of the high-temperature processing and the loss of lithium in slag in pyrometallurgical recycling. For LFP cells, all three cycling routes result in net increases in GHG emissions due to the relatively small gain from recovering iron materials in LFP cells (compared with Ni and Co materials in NMC and NCA), indicating that recycling LFP cells may not be sustainable from GWP perspectives. Among the three cycling routes, direct cathode recycling offers the highest environmental benefits because it avoids the energy- and chemical-intensive thermochemical unit operations and maintains the cathode's crystal structure and internal energy. However, there are concerns regarding the quality of cathode materials recovered from direct cathode cycling and this route has not been proved at a commercial scale. Ciez and Whiteacre's findings were supported by a recent study that assessed the environmental impacts of advanced hydrometallurgical recycling of LIBs based on primary data from a battery recycling company.[11] In this study, high environmental benefits (12–25% reduction of GHG emissions in comparison to no recycling) are obtained via advanced hydrometallurgical recycling of NMC and NCA cells mainly because of the recovery of precious cobalt and nickel, but recycling of LFP cells is proved not to be environmentally sustainable. The substantial difference in the environmental impacts of recycling different battery chemistries highlights the necessity of developing a battery chemistry-specific approach. Yang et al. also showed that the preprocessing steps, such as collecting, sorting, dismantling of spent LIBs, and transport between recycling facilities could contribute substantially to the emissions.[8j] In addition to being recycled, spent battery packs recovered from end-of-life EV could be reused in stationary applications as part of "smart-grid," and this scenario has been demonstrated to be environmentally beneficial.[12] However, its reliability and economic feasibility need to be further examined as the reuse of spent LIBs in "smart-grid" has not yet been developed commercially.

Besides the environmental considerations, economic analysis of LIB recycling is also of great concern because it determines the industrial selection of different recycling technologies for profitability. However, detailed economic evaluations of the entire recycling process and comprehensive comparison of different recycling routes and battery chemistries are relatively scarce probably because there are few recycling processes operating at the commercial scale. The profitability of battery recycling depends on two main factors, namely, the costs of collecting and processing spent batteries and the revenues of selling recovered materials. Yang et al. reviewed the economic benefits of different battery materials.[8j] The authors showed LCO recycling exhibits the best economy, followed by NMC. However, recycling Fe- or Mn-based cathodes that are free of valuable metals like Ni and Co leads to negative net economic value.[8j] Lin et al. found that the recycling of LFP cathode material is marginally profitable, about \$196 for recycling 1 ton of spent LFP batteries, which is mainly due to the low price of the recycled chemicals and high leaching reagent (acetic acid) consumption.[13] On the other hand, recycling LCO cathode materials could be highly profitable due to the high prices of Co and Li metals. According to Lin et al.'s economic analysis, recycling 1 ton of spent LCO powders can result in a total profit of \$31032 using the conventional sulfation roasting technique.[14] Regarding the popular NMC battery, Xiong et al. analyzed the entire remanufacturing cycle which includes chemicals recycling, cathode remanufacturing, and cell remanufacturing processes.[15] The authors found that the potential cost-saving from hydrometallurgical remanufacturing of NMC battery cells is about \$1870 per ton compared with the production of batteries from virgin materials. Selection of recycling route also plays a major role in determining the economics of the spent battery recycling and it becomes evident that direct recycling is the most economic route compared with the energy- and chemical-intensive pyrometallurgical and hydrometallurgical routes.[8j] However, the quality of the recycled cathode materials needs to be assessed to meet the standard of reuse. Overall, the currently limited studies indicate that the economic feasibility of battery recycling highly depends on the battery chemistry and recycling route. In addition, the cost of recycling at different locations varies significantly but the revenue is similar due to global trading.[8j] Therefore, the locations of recycling should be taken into consideration in practice.

### 2.2. Cathode Materials

The LCA studies showed cathode materials are a substantial contributor to GHG emissions and energy consumption for manufacturing LIBs.[8h,i,16] LIBs were first commercialized by Sony Corporation in 1991, adopting $LiCoO_2$ (LCO) as the cathode and graphite as the anode.[17] As graphite remains the primary anode in most commercial LIBs at present, the bottleneck regarding energy density is still cathodes. From 1991 to the 2010s, the price of LIBs has dropped by nearly 97%.[6] The cathode is the largest cost contributor among all battery components according to the cost model Ziegler et al. developed.[6] Efforts to boost cell charge density and lower cathode prices accounted for 38% and 14% of the LIB cost reduction, respectively.[6] Therefore, manipulating cathode design at the materials level is essential to the sustainability of the LIB industry. Several factors need to be considered when evaluating a cathode material, including but not limited to electrochemical performance (e.g., energy density, cycle life), raw material abundance, cost, and carbon emissions during production. These factors are largely dependent on the transition metals in cathodes. Here, we categorize the state-of-the-art cathodes by their chemical compositions, especially their transition metals (**Table 1**). LCO is widely applied in consumer electronics due to its high energy density, good conductivity, and high discharge voltage. However, LCO is unsuitable for large-scale applications due to the toxicity, scarcity, and high cost of Co. Shortly after the successful commercialization of LCO in the 1990s, the Ni-based cathode $LiNiO_2$ (LNO) received much attention.[18] LNO is an isostructural compound to LCO and has a similar theoretical capacity (275 mAh $g^{-1}$) but avoids the problematic Co. However, severe Li/Ni cation mixing and phase transformation issues in LNO lead to its low stability.[19] The inherent low stability brings about durability and safety concerns, which hinder its commercialization even today. The battery community has made great efforts in enhancing the performance of LNO and other Ni-rich variants.[20]

Introducing several other metal cations (e.g., Mn, Al, Co) to partially substitute Ni is one of the most successful practices to optimize LNO. This is how NMC and NCA came into play.[21] NMC and NCA are families of cathodes that have various ratios of Ni to other metal cations in the chemical formula. The cycling and thermal stability are enhanced, but the energy density is limited with lower Ni content. NMC and NCA have promoted the development of the automotive battery industry in the past decade. Moving toward higher Ni contents has become the trend of the Ni-based layered cathode development as the demand for high energy density increases. However, this is not simply traveling back to LNO. Various approaches have been applied to balance the energy density and stability, such as doping and coating.[22] In the meantime, decreasing or eliminating Co in NMC and NCA has become more pressing. Co contributes a significant portion to the entire cost and carbon emissions of cathode production.[8h] Besides, the child labor and human rights issues during the Co mining have drawn criticism widely. Therefore, Ni-rich Co-free cathodes have gained extensive attention recently. A variety of metal cations were selected to replace Co to improve the stability of Ni-rich cathodes.[23] Ni-based cathodes will persist due to the advantage of high energy density before more appealing cathodes appear. However, Ni will eventually become limited and expensive. The IEA forecasted that the demand for Ni and Co in clean energy applications will increase 40-fold from 2020 to 2050.[16] The LIB supply may evolve dynamically as the mineral extracting technologies develop and more mineral resources are discovered. However, such a rapid increase in demand for Ni and Co can barely be digested without partially shifting away from the Ni-rich chemistry. Additionally, the demand increase will further induce price volatility of raw materials.

The exploration of Ni-free and Co-free cathodes never stops. LFP is one of the most successfully commercialized cathodes, possessing long cycle life, high stability, and safety.[24] The only transition metal adopted in LFP is Fe, which is abundant,

**Table 1.** Selected LIB cathodes categorized by elements.

| Category | | Advantages | Disadvantages | Status | References |
|---|---|---|---|---|---|
| Co dominating | $LiCoO_2$ (LCO) | High energy density; high conductivity; high discharge voltage | Expensive and toxic Co; low accessible capacity compared to the theoretical value | Consumer electronics | [45] |
| Ni and Co containing | $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC)/ $LiNi_xCo_yAl_{1-x-y}O_2$ (NCA) | Increasing energy density with higher Ni content; lower cost and less toxicity than LCO | Low thermal and cycling stability and safety concern with higher Ni content | EVs, stationary energy storage | [21] |
| Ni-rich and Co-free | $LiNi_xM_{1-x}O_2$ (M = Mg, Al, Ti, etc.) | High energy density; low cost; environmentally friendly | Low thermal and cycling stability | Research | [23] |
| Low-Ni/Ni-free and Co-free | $LiFePO_4$ (LFP) | Inexpensive; high stability and safety; environmentally friendly; long cycle life | Low operation voltage; low energy density; low conductivity; additional processing needed | Power tools, EVs, and stationary energy storage | [24] |
| | $LiMnPO_4$ (LMP) | Inexpensive; high stability and safety; higher energy density than LFP | Poor electronic and ionic conductivities; large volume change; Jahn–Teller distortion of $Mn^{3+}$ | Research | [24a,27] |
| | $LiMn_2O_4$ (LMO) | Inexpensive; safety; high power density; environmentally friendly; good rate capability | Low energy density; fast capacity fade especially at high temperatures (>50 °C) | Power tools, EVs | [28a,b] |
| | $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) | High energy density; high power density; low cost; safety | Electrolyte decomposition at high voltage; inferior electronic conductivity; low cycling stability | Research | [32] |
| | $xLi_2MnO_3(1-x)LiMnO_2$ | High discharge capacity; low cost; safety | Severe voltage decay; poor rate capability; large first cycle irreversible capacity; low volumetric energy density | Research | [36] |
| | $Li_{1+x}M_{1-x}O_2$ (M = Mn, V, Ti, etc.) (DRX) | More choice of metals and more flexible cathode design; high energy density; low cost | Low stability at high voltage; immature synthesis route and large-scale production | Research | [40] |

inexpensive, and environmentally friendly. However, the energy density of LFP is relatively low compared to Ni-based cathodes, and the conductivity is intrinsically low.[25] Therefore, LFP requires further processing, such as coating and particle size engineering.[26] Mn is another inexpensive, abundant, and low-toxicity metal. $LiMnPO_4$ (LMP) is an isostructural compound to LFP with similar theoretical capacity but higher operating potential, thereby giving higher energy density than LFP.[24a,27] The polyanion framework leads to high oxygen stability and safety. However, the slow Li kinetics and low electronic conductivity result in inferior rate performance. LMO is another promising Mn-based alternative to Ni- and Co-containing cathodes.[28] LMO has a spinel structure and a 3D Li ion transport framework compared to the 2D layered structure of Ni- and Co-based cathodes. Therefore, LMO possesses excellent rate capability and high power density, which is often applied in power tools. However, LMO suffers from short cycle life especially at elevated temperatures (>50 °C) due to the structural degradation and Mn dissolution issues, and the energy density is relatively low.[29]A successful demonstration of LMO is its application in EVs by blending LMO with NMC to exploit the advantages of both materials. In this case, LMO improves acceleration with high power density and NMC supports long-distance driving with high energy density.[30] It was also reported that NIO Inc. has announced to adopt NMC and LFP hybrid battery packs in EVs.[31] This approach can be a temporary solution to reduce the reliance on Ni and Co during the transition to Ni- and Co-free cathodes. Meanwhile, it is crucial to develop high-energy cathodes that are not substantially reliant on Ni or Co. $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) is a low-Ni Mn-based spinel cathode material. LNMO has a high working potential (4.7–4.9 V vs $Li/Li^+$) and high energy density (around 650 Wh $kg^{-1}$) at the materials level.[32] The high power density of LNMO also makes it a good candidate for power tools. However, the decomposition of conventional liquid electrolytes at high voltage, inferior electronic conductivity, and Mn dissolution issues raise concerns on the cycling stability and safety, which impedes the commercialization of LNMO.[33]

There was a clear shift toward Ni-based cathodes and higher Ni content for EV batteries as higher energy density was in large demand, but Ni- and Co-free cathodes regained attention in recent years. The share of NMC and NCA materials in electric LDVs has experienced a steady growth from 2014 to 2019.[34] In 2019, NMC and NCA accounted for more than 80% of total cathode materials in new electric LDVs around the world.[34] LFP is primarily adopted in China, and the LFP share of all cathodes decreased from 60% in 2014 to around 10% in 2019.[34] LMO has also experienced a decreasing market share year by year possibly due to the growing energy density need. However, the worldwide increasing demand for Ni and Co drove the cost of these raw materials higher as the battery chemistry leaned to Ni-rich NMC and NCA materials. Furthermore, the COVID-19 pandemic has caused extensive damage to the global economy, driving up the cost of EVs. The supply and price of Ni and Co can be greatly impacted by geopolitical instability and wars.[35] The LIBs with modest energy density can satisfy the daily commuting needs of most EV consumers. High energy density is less significant as fast charging develops and more charging infrastructures are constructed. Therefore, LFP undergoes a resurgence due to its safety, low cost, and fast charging capability. The LFP market share across all EV cathodes grew from about 10% in 2019 to 19% in 2020 and 24% in 2021.[31]

Introducing excess Li to the cathodes is another strategy to further enhance the battery energy density. Li-rich layered oxides (LLOs) are a series of high-energy Mn-based cathodes. In the 1990s, Thackeray et al. demonstrated the $Li_2MnO_3$-stabilized layered materials and reported $x Li_2MnO_3(1-x)LiMnO_2$.[36] Shortly after that, more metal ions were incorporated and the LLOs family was expanded to $x Li_2MnO_3(1-x)LiMO_2$ (M = Ni, Mn, Co).[37] We want to highlight that the nature and history of these materials, either solid solution or composite, are still under debate. LLOs have high discharge capacity and benefit from the use of low cost, nontoxic Mn. However, the voltage decay, low rate capability, and low initial Coulombic efficiency issues bring difficulties to their commercialization.[38] The high voltage operation condition of LLOs induces more oxygen loss and lowers the cycling stability. Furthermore, compatible electrolytes at high voltages are rare, which further hampers the practical applications of LLOs. Many researchers have been attempting to understand the degradation mechanism of LLOs and enhance their electrochemical performance, but LLOs are still primarily at the research level at present.[39] Disordered rocksalt (DRX) cathodes, another class of Li excess materials, have recently obtained extensive research attention. DRX cathodes have various choices of 3d and 4d metals in contrast to the conventional layered oxides that use particular scarce metals.[40] The design of chemical compositions for DRX cathodes is more flexible. These earth-abundant and inexpensive metals, such as Mn, Fe, and Cr, can be utilized more efficiently. Therefore, DRX cathodes are a promising alternative to existing commercial cathodes due to their wide range of raw materials, cathode compositions, and high energy density. However, the development of DRX cathodes is still limited to the lab research. The synthetic routes and scale-up production are immature at the current stage. Li et al. comprehensively reviewed the progress and potential hurdles for the commercialization of DRX cathodes.[41] To enable superior capacity, the DRX cathode particles are normally pulverized into nanoparticles for shorter Li diffusion distances. The nanosized particles lead to large surface area and more parasitic reactions at the surface, resulting in faster performance decay. The active material to carbon and binder ratio is low for DRX cathodes to achieve better conductivity and Li kinetics. To the best of our knowledge, the normal active material mass loading is around 70% and the highest is 80% in the literature,[41] which is significantly lower than the loading of commercial cathodes. However, the Li diffusivity of these Li-excess DRX cathodes is still remarkably lower than that of conventional layered oxides by orders of magnitude, which raises Li kinetics problems.[41] Such low active material loading and small particle size bring challenges to enhancing the volumetric energy density in practical uses. For these Li-excess cathodes, oxygen redox reaction can readily take place due to the population of high energy Li-O-Li state, which could induce oxygen loss at the particle surface and irreversible transition metal migration.[42] These undesired structural transformations can cause capacity and voltage fading and eventually performance degradation. All these hurdles currently impede the commercialization of Li-excess cathodes. In addition, the availability and price of Li should be addressed when developing Li-excess materials as Li is the most essential metal in all types of LIBs. Additionally, conversion-type cathodes normally require significantly cheaper and more abundant elements from the Earth's crust, such as chalcogens. For example, Li-S batteries have relatively high energy density and low cost. However, the large volume expansion and the shuttle effect of soluble species lead to performance degradation hindering their further applications in EVs.[43] Overall, no single electrode material can suit all application scenarios. Distinct applications require different advantages of specific cathodes, such as the high energy density of Ni-rich NMC, and the stability and low cost of LFP. Each of the cathodes discussed is expected to undergo advancement and they will co-exist for different applications, but the general trend will be toward a more sustainable approach.

Shifting from Ni and Co-based cathodes to sustainable materials will become the general trend as the minerals become more limited, similar to the transition from fossil fuels to green energy resources. However, Ni and Co-based cathodes still possess an appealing advantage of high energy density that can practically convert to long driving mileage in EVs. Comprehensive reviews on LIB electrode materials suggested that various chemistries and configurations for LIBs will still be present for specialized applications.[30,44] There are several approaches to make the Ni- and Co-free cathodes more competitive not only at the cathode level, but also at the anode and battery pack levels: 1) advancement of anode materials, such as Li metal and Si anodes, can greatly improve the cell-level energy density; 2) optimizing the form factor of the individual cell and battery pack can further enhance the performance of batteries. Furthermore, the development of associated supporting facilities can reduce the mileage anxiety and the demand for high-energy electrodes: 1) developing fast charging and more efficient charging methods (e.g., wireless charging during driving); 2) constructing more distributed charging stations; 3) combining battery charging and battery pack swapping methods for different types of EVs.

### 2.3. Supply Chain and Manufacturing

More practical factors beyond materials design need to be considered in industrial manufacturing, such as supply chain and battery pack manufacturing. The LIB supply chain can be tracked back to the extraction and processing of raw minerals. A volatile supply chain or inefficient manufacturing may offset the performance benefit promoted by electrode materials. The mining and processing of LIB raw materials are more scattered across the world, compared with the procedures of fossil fuels (**Figure 3**). The uneven distribution of essential raw minerals may potentially give rise to geopolitical challenges and impact the global LIB industry. For example, Ni is a predominant metal in commercial cathodes for LIBs. Russia is one of the major countries that extract and refine Ni mines (Figure 3a,b). Due to the Russia-Ukraine war, the price of Ni almost doubled within one week from late February to early March in 2022.[35a] The IEA reported that the total battery cost could increase by 6% if

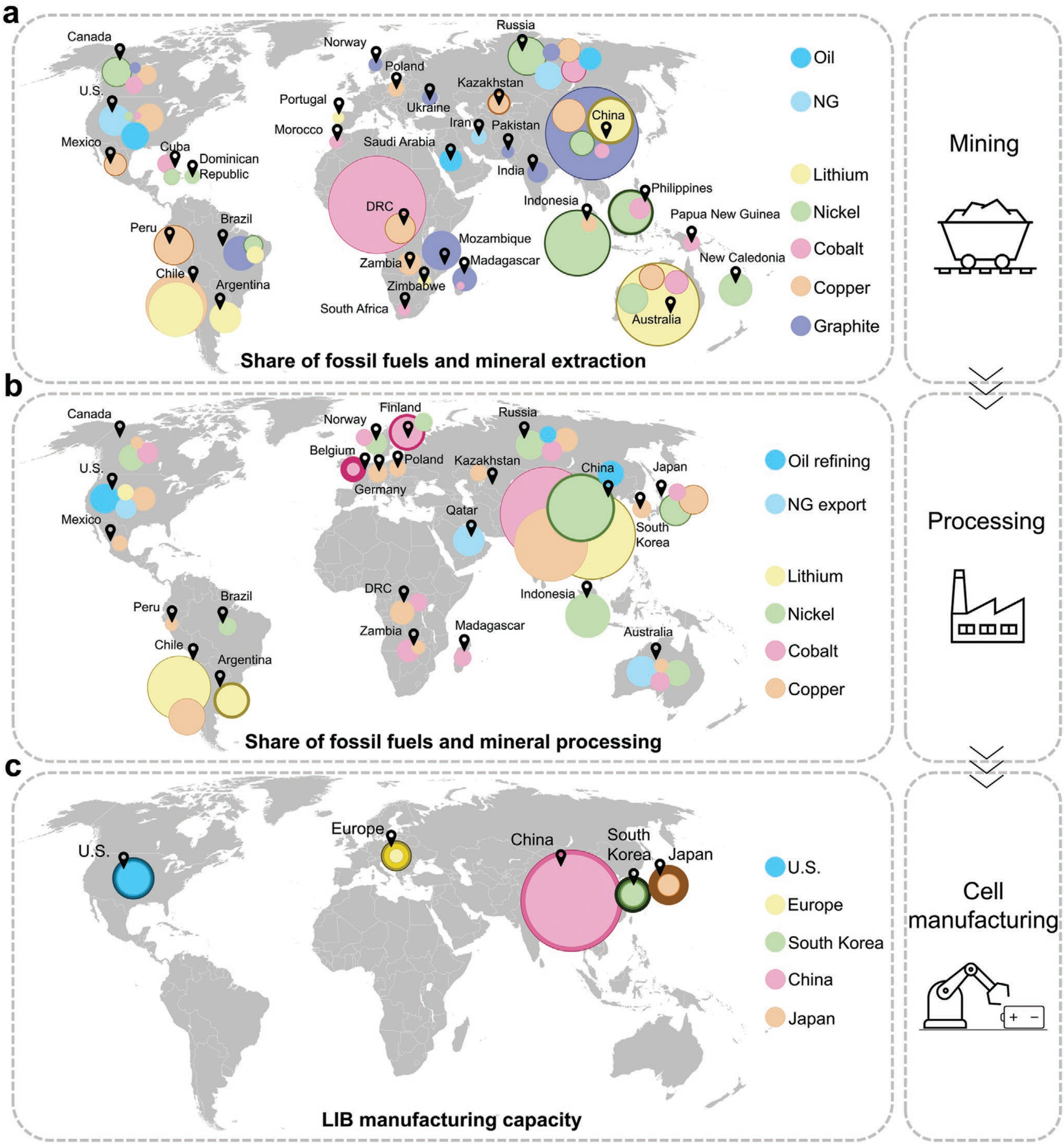


**Figure 3.** The worldwide distribution of LIB cradle-to-gate processes. a) Share of fossil fuels and mineral extraction in 2019 and 2020.[4,48,75] b) Share of fossil fuels and mineral processing in 2018, 2019, and 2020.[4,48,62] c) LIB manufacturing capacity by country in 2016, 2019, and 2020.[57,63,65] The area of the color-coded circles is proportional to the share (percentage) or capacity (GWh). NG: Natural gas. DRC: The Democratic Republic of the Congo. The shaded rings around specific circles are "error rings" calculated based on collecting data from different references. This figure only displays the top three countries for fossil fuel extraction and processing and the major contributing countries/regions in LIB cradle-to-gate processes for visualization. The quantitative results for more countries and regions can be referred to Tables S1–S3 in the Supporting Information. The distribution of LIB raw mineral materials is sporadic across the world, but raw material processing and LIB manufacturing are mainly in Asia.

the prices of Ni or Li were doubled.[4] Contemporary Amperex Technology Co. Ltd. (CATL), the largest LIB manufacturer in the world, has announced to raise prices for some battery products due to raw materials cost increase.[46] Accordingly, some EV companies, such as Tesla, Rivian, and BYD, have raised the prices of their EVs due to the increasing supply chain cost.[47] Therefore, evaluating the global supply chain and manufacturing capability is vital to gaining a comprehensive understanding and reasonable forecast of LIB development.

The fossil fuel supply chain has been established and remained relatively stable, whereas the LIB supply chain is under development and evolving rapidly. The primary oil producers are the United States (US), Russia, and Saudi Arabia, and the top three oil refining countries are the US, China, and Russia (Figure 3a,b).[4] Regarding the natural gas production and distribution, the major producing countries are the US, Russia, and Iran, and the top three exporting countries are Qatar, Australia, and the US (Figure 3a,b).[4] In contrast, the global competition for the LIB supply chain has just begun. The distribution of the essential minerals for producing LIBs is scattered globally. In 2019 and 2020, three major Ni mine extraction countries are Indonesia, the Philippines, and Russia, which accounted for the global share of 29.8–33.0%, 12.0–15.7%, 10.1–11.3%, respectively (Figure 3a and Table S1, Supporting Information).[4,48] For the global share of Co mine extraction, the Democratic Republic of the Congo (DRC) dominates with 69.0–70.4%, and Australia and Russia contribute 4.0–4.2%, 4.0–4.7%, respectively (Figure 3a and Table S1, Supporting Information).[4,48a] The top three contributing countries to the global share of Li mining are Australia (48.7–52.0%), Chile (21.9–22.0%), and China (13.0–17.0%).[4,48a] Given the dispersed distribution of raw materials, the LIB supply chain requires worldwide collaborations. Moreover, the leading countries for the mining of certain materials may change as new minerals are discovered and mining technology improves. For example, a recent assessment of worldwide metal reserves indicated that the top-reserve countries for $Li_2CO_3$, Ni, and Co mines are Chile, Indonesia, and DRC, which possess 52 670 1000 metric tons, 28 750 1000 metric tons, and 2970 1000 metric tons, respectively.[49] However, unexplored regions that have considerable potential for mineral resources can potentially be a game-changer.

Assessing the future demand-supply balance of LIB raw materials is challenging because of the uncertain factors, including but not limited to the pace of automotive electrification and spent EV battery recycling, reliance on Ni- and Co-based cathodes, supply from new mining resources (e.g., seabed), production efficiency enhancement by technology improvement, and corresponding policies. It is still controversial in the literature if Co supply will meet the demand in the coming decades.[50] Tisserant and Pauliuk estimated that the Co reserve in the ground will be sufficient to supply to at least 2050.[51] Sverdrup et al. have created a model to assess the long-term Co supply.[52] The authors predicted that the supply of Co will stay sufficient until 2130 and reach a peak level in the period of 2040–2050. The price of Co could experience a sharp increase after 2050. From 2080, Co recycling rate will increase and Co supply from recycling will exceed primary extraction in response to the increased price. One potential concern of these earlier studies is the underestimation of the automotive electrification ambitions and corresponding Co demand. On the other hand, some studies are relatively conservative and less optimistic. Zeng et al. suggested that although battery technology improvement and recycling can bring long-term Co supply sustainability, there will be an inevitable Co shortage during 2028–2033.[53] Valero et al. also predicted that Co demand will exceed supply as early as 2030.[54] Some studies also projected a possible Ni shortage in the upcoming years. Valero et al. showed a bottleneck period for Ni, when the demand exceeds the supply, is expected to be 2027–2029.[54] This prediction agrees with another study that forecasted there will be Ni deficit by 2028.[55] It is commonly believed that Li supply will not be a constraint in this century, but recycling is vital to avoid an early supply deficit.[56]

Regional supply and geopolitical factors can add discrepancies to the estimation of global mineral demand–supply risk. The uncertainties of Co supply mostly arise from the heavy reliance on the production from the DRC and the unstable political environment in the DRC (Figure 3a).[57] If the scope, however, is focused on regional supply, 96% of the imported Co in the European Union (EU) was from Russia.[58] Therefore, the global estimation might not apply to the forecast of regional supply. In addition, competition for raw materials exists between different regions, adding more uncertainties to the global LIB supply chain. Sun et al. evaluated the competition intensities of 15 LIB-related commodities (lithium mineral, cobalt ore, nickel ore, etc.) of 238 countries and regions in 2019.[59] For example, the competition for lithium hydroxide and lithium carbonate between Japan and South Korea is the most intense among all competitions analyzed.[59] Both countries heavily relied on the import of lithium raw materials for battery manufacturing. The competition between China and Finland for cobalt ore is also rather intense because they are the top two cobalt refining countries while the domestic cobalt reserves are insufficient (Figure 3a,b).[59] Further details can be referred to the original reference.[59]

Mineral processing capability also plays a crucial role in the LIB supply chain besides mineral reserves. The mineral reserves may not necessarily reflect the exact demand in the LIB industry. The reason is that LIB production normally needs high purity precursors, which require further processing after mining and specific mineral sources.[57] For example, Class 1 Ni sulfate (Ni impurity of 99.8% or greater) that is eligible for LIBs is mostly produced from Ni sulfide ores, which only accounts for around 40% of available Ni reserves.[57,60] Such strict requirements on raw materials also bring challenges to recycling. The fact that many essential metals for LIBs are mainly produced as byproducts of other metal mining further complicates the LIB supply chain.[57] For instance, over 80% of Co is produced as byproducts of Ni and Cu.[61] The worldwide distribution of mineral processing is relatively concentrated. China has the largest processing volumes for multiple essential metals. The global share of China for Li, Ni, Co, and Cu processing is 55.0–58.0%, 29.9–35.0%, 63.6–65.0%, and 39.8–40.0%, respectively (Figure 3b and Table S2, Supporting Information).[4,48a,62] Other major processing countries are distributed in the rest of Asia, Europe, and South America. Separators are another critical component of LIBs, which serve as a physical barrier to prevent cell short-circuiting and electrolyte reservoir for Li transport. The major LIB separator manufacturers are located in Asia. China, Korea, Japan, and the US accounted for 43%, 28%, 21%, and 6% of the global separator manufacturing, respectively.[63] The separator market of Asia-pacific region is forecasted to undergo a rapid growth and will still be dominant in the next 5 years.[64]

LIB manufacturing is the next key step. Regarding the share of the global LIB production in the past few years, China, the US, Europe, South Korea, and Japan accounted for 62.3–76.9%, 7.9–13.1%, 1.4–7.0%, 4.0–9.8%, and 3.2–11.8%, respectively (Figure 3c and Table S3, Supporting Information).[57,63,65] The statistics show that LIB manufacturing is dominated by Asia, especially China. More facilities will be established in Europe, North America, Australia, and Asia considering those under construction or planned.[66] China, Japan, and South Korea are rated first, second, and third in the 2020 LIB supply chain ranking, which is evaluated based on raw materials, manufacturing, battery demand, etc.[67] Europe and North America are progressing and their gap with Asia is narrowing. In 2021, the US and Germany moved up to second and third place, respectively, which is predicted to be maintained in 2026.[68] Additionally, Asian countries, including China, South Korea, and Japan, have vertically integrated the supply chain from raw materials processing to battery manufacturing.[57] In the 1990s, Japan determined to develop its LIB industry. The Japanese government supported the research and development (R&D) of private sectors and helped them establish low-cost manufacturing plants.[57] China and South Korea have copied the success of Japan through developing partnerships between the government and LIB industry and providing subsidies since the late 2000s.[57] The mature LIB supply chain and accumulated technology and production experience gave the Asian countries advantages in the EV era. The US and Europe did not focus on domestic LIB manufacturing until the late 2010s. Therefore, the supply chain is not as robust, and manufacturers are relatively inexperienced.[57] Overall, Asia has the edge on raw materials processing and LIB manufacturing at present, but Europe and North America are accelerating the construction of their supply chains.

The quality and efficiency are significant in the battery manufacturing. The manufacturing technology determines the manufacturing efficiency and battery performance, thereby impacting the manufacturing capacity. It was reported that the Ford Motor Company announced the deployment of the fifth-generation (5G) technology in manufacturing to enhance connectivity and achieve higher manufacturing efficiency.[69] Manufacturing technology and battery design largely stem from battery R&D. Public and private R&D is the major driving force for the LIB cost reduction in the past three decades.[6] Specifically, the R&D of chemistry and materials science has played a major role in the cost reduction.[6] Similar attempts may further reduce the cost and enhance the performance of LIBs in the future. In this regard, the US has a solid foundation for battery research and technology. The government has maintained a good strategic and financial support for the fundamental research. Recently, the US Department of Energy (DOE) announced to use $209 million for the vehicle battery research.[70]

A battery management system is essential to keep the batteries in proper working condition. In EVs, semiconductors play an important role in the electronic control of automotive power systems, such as fast charging and reducing energy loss.[34] Semiconductor chips are also indispensable components in the power control systems of EVs. The interruption in any part of the supply chain may impact the whole production. For example, the semiconductor industry has been hit hard since the outbreak of the COVID-19 pandemic. The production of vehicles is largely limited due to the shortage of semiconductor chips.[71] In 2019, the global share of the automotive semiconductor industry for Europe, the US, Japan, and China is 37%, 33%, 26%, and 2%, respectively.[34] Developing the semiconductor industry is equally important to build a robust supply chain for LIBs and EVs. In all, a resilient supply chain requires not only a stable raw material supply, but also joint effort from the related fields.

The EV industry is the largest market of LIBs. It can reflect the global competition of the LIB supply chain and manufacturing. EV sales are expected to have a steady growth of tens of millions each year in the next few decades.[72] In 2020, the top three countries possessing BEVs and plug-in hybrid electric vehicles are China, the US, and Germany.[73] In addition, EV penetration rate in the vehicle market indicates the intention of vehicle electrification. The top ten countries for EV share of new car sales in 2020 are all located in Europe thanks to their incentive policies.[72] More countries have sped up their pace in transitioning to EVs as the time approaches the pledge on carbon neutrality. LIBs will play a critical role in the skyrocketing growth of EVs. The LIB industry is less vulnerable to raw material disruption than fossil fuels to some extent because of the wide range of available electrode materials. Furthermore, the consumers will not be impacted immediately by the LIB supply chain interruption due to the relatively long life cycle of LIBs. For instance, the price of crude oil has recently hit a new record since 2008.[74] Every gasoline refill has a direct impact on ICEV owners. Although the manufacturing and sale of new EVs are affected by the LIB supply chain fluctuation, the operation of existing EVs is barely altered. In addition, fossil fuels require continuous inputs once combusted while minerals in LIBs can be reused and recycled. Therefore, a worldwide energy crisis for the LIB industry is unlikely to occur. However, the diversified mineral types and their concentrated geographical distributions bring more uncertainties to the supply chain. A stable supply chain is critical for the sustainable development of LIBs. Major LIB producing countries have been attempting to establish their domestic supply chains and gain an edge on the LIB manufacturing. A sustainable supply of raw materials is needed as a prerequisite. In this regard, developing local mining and processing abilities are essential for boosting domestic LIB manufacturing capacity. The competition between leading countries can accelerate the advancement of LIBs and the establishment of a global LIB industry ecosystem. On the other hand, a closer collaboration (e.g., multiyear agreement on supply) across the world and associated legislation are needed to achieve a sustainable supply chain.

### 2.4. Recycling

The proper processing of used LIBs has become a pressing and inevitable task as more first-generation EVs approach end-of-life and raw materials become resource-limited. At present, the global recovery rate of used LIBs is rather low.[76] A substantial amount of used LIBs is handled inefficiently and dangerously, such as by landfilling and illegal disposal.[76] The inappropriate processing has caused extensive damage to the environment, human health, and massive fire and explosion incidents.[76] Instead, giving used LIBs a "second life" through reusing, remanufacturing, and repurposing appears to be a promising strategy to harness the remaining energy.[76] For example, used EV LIBs can be repurposed for grid energy storage. However, there are few regulations for second-life LIBs at present. Corresponding legislation should be established in advance to avoid the potential chaos in the market as more used LIBs emerge.

Recycling is another approach to satisfying the carbon neutrality criteria and easing anxiety over finite resources. Recycling can potentially lower the overall energy consumption and emissions of virgin battery production as the LIB recycling industry grows larger and becomes more mature.[77] For instance, the production of transition metals is largely from sulfide ores. This process produces $SO_x$ emissions that cause damage to the environment, such as acid rain and soil contamination. Effective recycling of transition metals can substantially reduce the emissions during raw mineral processing. Dunn et al. reported that LIB recycling can effectively reduce GHG and $SO_x$ emissions especially for Ni and Co-containing cathodes based on the LCA.[8h] Therefore, recycling can significantly foster the establishment of a sustainable LIB industry. However, the LIB recycling industry still faces numerous practical hurdles at present, such as the technical limitations of different recycling routes.

Generally, there are three major LIB recycling routes, which are schematically shown in **Figure 4**. The direct recycling route (denoted by the blue arrows) involves the least processing among the three routes. Initially, the electrolyte is extracted from the spent LIB using supercritical carbon dioxide. The retrieved electrolyte can be recycled after further processing. The remaining components will be separated based on their properties through a series of physical processes. The recovered cathode material can be reintroduced to the battery assembly lines. Re-lithiation or additional processing of the recovered cathode is normally needed to compensate for the performance loss.[78] Direct recycling can recover nearly all battery materials and requires less treatment than other routes. However, the performance of recovered materials may be compromised.[57] Hydrometallurgical recycling involves leaching, which recovers the metal species from aqueous media (denoted by the orange arrows). Spent LIBs are pretreated with several physical processes, such as shredding and screening, to obtain black mass and Cu and Al foils. The black mass is leached to obtain the solution containing metal cations. After solvent extraction, the dissolved salts can be separated and recovered, and then reintroduced into the supply chain for cathode synthesis and battery manufacturing.[77] Hydrometallurgical recycling is efficient to isolate the component of interest in the aqueous environment, and the obtained product is pure. The procedures are relatively energy-efficient and environmentally friendly because they do not involve high-temperature processing. However, the treatment of tremendous effluents elevates the cost.

The third route requires additional energy-intensive smelting steps. Pyrometallurgical recycling recovers different metals through oxidation or reduction reactions at high temperatures. As indicated by the green arrows, the spent LIB can be dismantled and shredded first to generate the black mass, or it can be fed directly to the furnace to obtain the mixed metal alloy (e.g., Ni, Co). The latter approach is generally preferred because separating the black mass from Al and Cu foils adds additional cost, and the Al can be utilized as a reductant in the furnace to save the smelting energy.[77] The mixture of Al and Li oxides remains in the slag if adopting the latter approach, which is normally not recovered due to economical inefficiency. Next, the obtained metal alloy undergoes leaching and solvent

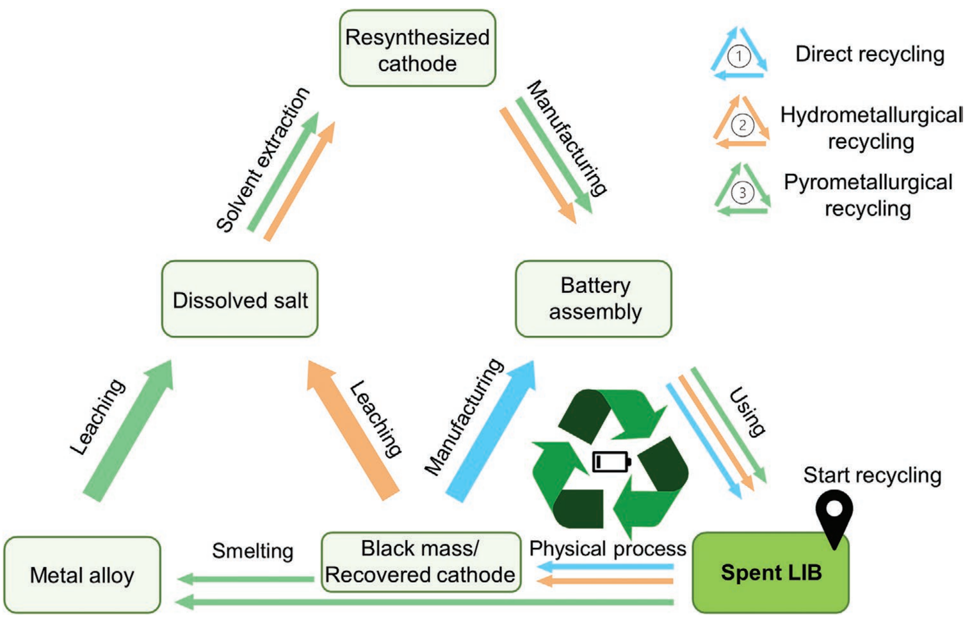


**Figure 4.** Three LIB recycling routes as indicated by the blue, orange, and green arrows. Physical processing (mechanical separation and dissolution) is usually adopted as a pretreatment for all recycling routes. Depending on the materials that need to be recovered, one can choose specific recycling routes to achieve higher recycling efficiency at a lower cost. For example, hydrometallurgical and pyrometallurgical recycling are economical for high Ni and Co cathodes but not appropriate for cathodes containing fewer precious metals such as LFP and LMO.[77,78]

extraction processes (hydrometallurgical recycling) for cathode resynthesis and battery assembly. Pyrometallurgical recycling needs little mechanical pretreatment and can efficiently recover metals. However, it results in substantial energy consumption, emissions, and transportation expense. The choice of recycling routes may vary with different types of spent LIBs and the material of interest to recycle. For example, LIB recycling mainly focused on recovering Co initially due to its high value. Subsequently, recycling at the cathode level may offer higher revenue than recycling particular metal constitutes as the Co content becomes lower and other cell components are in higher demand. Direct recycling may offer additional benefits in this regard, although maintaining the performance of recycled materials to the level of virgin materials remains challenging.[77] Overall, the technical obstacles need to be addressed to make recycling more economically attractive.

For these aforementioned traditional recycling approaches, preprocessing steps such as discharging and pulverization are normally required. The spent LIBs entering the recycling line are mostly at partially charged states. The lithiated graphite anode is highly reactive in air, which can readily lead to aggressive thermal release and safety concerns. Therefore, discharging the spent LIBs first is critical yet tedious. The discharging process is time-consuming and requires large spaces to prevent cell contact. The salt solution used for degassing during discharging raises additional costs. After discharged, the spent LIBs can be safely pulverized into mixed powder. As discussed, hydrometallurgical and pyrometallurgical recycling require extensive heat and chemical treatments to separate and recover the desired metals. However, impurities, such as Cu, Al, Fe, and organic compounds, are still inevitably present in final recycled products even for direct recycling that needs the least processing.[79] Zhang et al. and Peng et al. investigated the effect of Cu impurity in recycled NMC and LCO batteries.[79a,b] Metallic Cu, especially from physical separation with direct recycling approach, can easily cause short-circuiting.[79a] Low content of Cu ion impurity can benefit the recovered cathodes regarding the capacity and capacity retention, while excess Cu ion introduces impurity phase to the cathode and deteriorates the performance.[79a,79b] Similar beneficial effects at low content and detrimental effects at high content for Al and Fe have also been reported.[79c,d] However, the actual impurity content in industrial recycling processes can be more random. It is critical to monitor and control the impurity level for the quality of recycled materials. Related regulations on the standard of impurities in recycled electrodes are recommended.

In addition to the three traditional recycling routes, more novel approaches have been demonstrated at the research level to address the energy-intensive, high cost, and heavy waste issues. Zhao et al. reported a precise separation method that could simply separate jellyroll cell components in water.[80] The pretreatment of discharging is avoided without compromising safety because water can isolate oxygen and immediately extinguish the potential fire during disassembly. $LiC_x$ in the graphite anode can react with water, accompanied by heat and bubble generation, which can facilitate the dissolution of binder and peeling-off of anode materials from the current collector. This method achieved higher recycling efficiency, simpler processing, and higher revenue at the lab research scale than traditional recycling approaches. Wang et al. added ammonium sulfate during recycling to reduce the decomposition temperature of LCO to below 400 °C, which lowered the energy consumption and enhanced the recycling efficiency.[81] These novel recycling methods showed appealing results at the research scale, but more technical challenges and economic efficiency should be addressed as applied to the industrial-scale recycling.

Recycling can relieve the pressure on the primary production of essential metals (e.g., Co and Ni) in the long term, but there could be a concern of short-term shortage affecting Ni-based cathodes, as discussed in the last section. A consensus nevertheless is that recycling will play an indispensable role at least in the long term. IEA estimated that the spent EV batteries will surge after 2030.[4] Therefore, boosting the primary production is still the most feasible way to address the short-term Co or Ni supply risk. The World Bank reported that even the end of life recycling rate of Co could reach 100%, there is still a large demand for primary Co production.[82] An additional concern is the long lead time for mining projects, which is estimated to be around 16 years on average from discovery to production.[4] Such long duration may not satisfy the ramping demand in the short term. Therefore, we suggest that more incentive policies should be established to simplify and accelerate the mining process and encourage the mining companies and investors to develop new mining projects. Meanwhile, governments should provide stronger support to the LIB recycling industry, because the profitability is relatively low at the early stage when recycling is at a small scale and primary production can still digest the demand. For the countries not possessing a robust domestic supply chain, recycling can supplement the supply of primary battery components.[57]

Supportive policies are also critical to tackle practical hurdles faced by the LIB recycling industry. For instance, LIB manufacturing was not systematically regulated initially for the convenience of recycling. Therefore, identifying and classifying various cell components and electrode materials raises recycling expenses. In the US, spent LIBs are classified as hazardous waste, and the cost of transporting spent LIBs accounts for more than half of the total recycling expenses.[63] Corresponding policy support can give rise to more monetary incentives for manufacturers to resolve these challenges. For example, standardizing the cell design and labeling the materials can reduce the pretreatment cost during recycling. Labeling battery chemistries in a standard way and classifying different batteries during recycling would also allow the highest environmental benefits of battery recycling based on the LCA. Ma et al. have discussed several challenges faced by the LIB recycling community.[83] In addition to the technical obstacles of the three aforementioned recycling approaches, the evolving battery design (e.g., Tesla's 4680 cylindrical cells and "tabless" design, BYD's blade battery pack, and CATL's cell-to-pack technology) brings more difficulties to disassembly and pretreatment.[83] It is expected that there will be nearly 2 million metric tons of global spent LIBs per year by 2030, pushing recycling to a large scale.[83] However, the profit of large-scale recycling is limited because of the lack of regulation support, nonstandardization issues, high cost of transportation, and storage of spent batteries at the large scale.[83] The trend of Co-free materials further diminishes the economic benefit of recycling. Convincing battery manufacturers to adopt recycled materials is also challenging because the performance of recycled materials needs to match or exceed the virgin ones.[83] Therefore, more collaborations between the industry, universities, and laboratories are needed to meet the practical industrial requirements.[83] Government incentives and policies can attract more researchers, manufacturers, and investors, enabling recycling technologies to progress and total costs to be reduced.

### 2.5. Policies and Legislation

Government policies and legislation often regulate and guide the LIB materials development, supply chain, manufacturing, and recycling. Investigating the regulations in various countries is crucial to comprehend the global LIB industry. Several countries and regions have declared their targets for addressing environmental issues and the global climate change. Specifically, the US, Canada, and EU plan to achieve net-zero emissions by 2050, and China aims for carbon neutrality by 2060 (**Table 2**).

**Table 2.** Policies of selected countries/regions for EVs development.

| Country/Region | China | European Union | United States | Canada |
|---|---|---|---|---|
| Emission | Carbon neutrality by 2060.[90] | 1) Carbon neutrality by 2050 and at least 55% emissions reduction by 2030.[91]<br>2) $CO_2$ emissions standards (in terms of g $CO_2$ $km^{-1}$) for cars tighten by 37.5% and for vans by 31% between 2021 and 2030.[92] | Achieving carbon-pollution-free electricity by 2035, and achieving net-zero emissions, economy-wide, by 2050.[93] | Net-zero emissions by 2050.[94] |
| EVs market | 1) NEVs sales reaching 20% of vehicle sales by 2025, NEVs become the mainstream of new vehicles sales by 2035;[1a]<br>2) Target for manufacturers: annual NEV credit as a percentage of their annual vehicle sales (14% in 2021, 16% in 2022, 18% in 2023).[95] | At least 30 million passenger ZEV stock by 2030 and nearly all passenger LDV and heavy commercial vehicle being zero-emission by 2050.[1b] | 1) 30% ZEV sales for all new MDVs and HDVs by 2030 and 100% by 2050 in 16 regions/states.[1c]<br>2) 22% ZEV sales in passenger LDVs in ten states by 2025.[96]<br>3) Executive order requires that all new cars and passenger trucks sold in California to be ZEVs by 2035.[97] | ZEV targets 10% of LDV sales by 2025, 30% by 2030, and 100% by 2040.[1d] |
| Charging infrastructure | 1) Building more than 120 000 charging stations and more than 4.8 million charging outlets by 2020.[98]<br>2) The 13th 5 year plan included RMB 90 million in funding for installation of charging infrastructure.[99]<br>3) Over 30 cities offer subsidies for home or public EV charging.[99] | Target of 1 million publicly accessible chargers installed by 2025.[100] | 1) Building a national network of 500 000 EV chargers by 2030.[101]<br>2) Invest $7.5 billion to build a national network of EV chargers.[102] | Invest an additional $150 million over 3 years in charging and refueling stations across Canada, as announced in 2020.[103] |
| Manufacture | 1) Restricting subsidies to only larger battery production facilities (at least 8 GWh production capacity);<br>2) Tax exemptions for battery producers;<br>3) Restricting electric vehicle incentives to vehicles with batteries manufactured in China to attract foreign investment.[85] | The European Investment Bank supported the construction of an LG Chem Li-ion battery cell-to-pack manufacturing Gigafactory in Poland in early 2020 (EUR 480 million).[86a] | Near-term objective (2025):<br>1) Develop federal policies to support the establishment of resilient domestic and global sources and supplies of key raw materials;<br>2) Decrease cost to enable a $60 kWh$^{-1}$ cell cost.<br>Long term objective (2030):<br>1) Eliminate Co and Ni in LIB;<br>2) Reduce the cost of EV pack manufacturing by 50%.[63] | 1) Investing CAD 590 million to the Ford Motor Company Canada to support EVs production.[86b]<br>2) The federal and Québec governments are providing CAD 100 million to Lion Electric to support a battery pack assembly plant project.[104] |
| Recycling | Encourage the standardization of battery design, production, and verification, as well as repairing and repackaging for second life utilization.[87] | 1) From 1 July 2024, only rechargeable industrial and EV batteries established a carbon footprint declaration, can be placed on the market;<br>2) Increasing transparency of the battery market and the traceability of large batteries throughout their life cycle by using new IT technologies, such as Battery Passport.[88] | Near-term objective (2025):<br>1) Foster the design of battery packs for ease of second use and recycling;<br>2) Increase recovery rates of key materials such as cobalt, lithium, nickel, and graphite.<br>Long term objective (2030):<br>Create incentives for achieving 90% recycling of consumer electronics, EV, and grid-storage batteries.[63] | Lithion Recycling Inc. received $3.8 million, and Li-Cycle Corp. received $2.7 million for LIB recycling.[89] |

Automotive electrification will play a key role in such decarbonization process. Many countries and regions have announced policies for EV development, infrastructure construction, and LIB recycling in response to the growing competition in the EV market. Herein, we select several active players in the worldwide race of EVs and LIBs. We demonstrate how their policies can impact the EV and LIB development on a national or global scale.

Present global EV sales are primarily contributed by Europe, China, and the US.[3,72] These major contributors have established targets for EV sales to further stimulate their EV developments (Table 2). The EU announced the goal that nearly all cars, vans, buses, and new heavy-duty vehicles will achieve zero-emission by 2050.[1b] China targets to have 20% of new vehicle sales from new energy vehicles (NEVs) by 2025.[1a] Furthermore, BEVs are expected to become the mainstream of new vehicles sold in China by 2035.[1a] The policies for EV sales in the US are primarily at the state level. 16 states and regions expected zero-emission vehicle (ZEV) sales to constitute 30% of all new mid-duty vehicles (MDVs) and heavy-duty vehicle (HDVs) sales by 2030 and 100% by 2050.[1c] In Canada, ZEV sales are expected to account for 10%, 30%, and 100% of LDV sales by 2025, 2030, and 2040, respectively.[1d] More countries in Asia, Europe, and North America have been actively deploying the EV industry, and the specific policies can be referred to ref. [84]. Charging infrastructures need to be developed accordingly to accommodate the rapid growth of EVs. Wide implementation of charging infrastructures can enhance the confidence of EV consumers and stimulate EV manufacturers. Therefore, charging infrastructures determine the potential market of EVs. The countries leading EV sales also invest heavily in charging infrastructures (Table 2). Furthermore, geologic and climate factors need to be addressed upon EV deployment because the performance of automotive batteries is susceptible to the environment, such as temperature. In summary, several top economies in the world have shown their determination in the development of EVs and established aggressive targets. Such positive signals can potentially spread confidence to the LIB community.

LIB is one of the most critical components in EVs. The global LIB production capacity and distribution have evolved over time, largely due to the policies of different countries and regions. At present, LIB manufacturers mainly distribute in Asia, North America, and Europe (Figure 3). Many countries have worked to establish a resilient domestic supply chain and manufacturing to gain some advantages in the global competition of EVs and LIBs. The Chinese government announced tax exemptions for battery producers to promote domestic manufacturing.[85] The US, Canada, and many European countries have also provided strong finical support to domestic battery manufacturers.[63,86] At the materials level, there is a general trend toward Ni- and Co-free cathodes. The US DOE announced the goal to reduce the dependence on Ni and Co and expected to eliminate Ni and Co in LIBs by 2030.[63] More automotive companies have been exploring Ni/Co-free cathodes.[31] The two types of cathodes will coexist for various purposes. However, Ni and Co battery chemistries could potentially be supplemented or substituted by more sustainable alternatives. Importantly, it is becoming more challenging for emerging materials to be cost-competitive considering the extensive investments that have already been committed.[30] Therefore, legislation support might be required to prevent the "lock-in" of incumbent materials when developing new materials. In general, many countries are attempting to develop domestic manufacturing in a more sustainable manner.

Recovering the scarce metals through recycling is another path to building a sustainable LIB industry. Unstandardized battery labeling, as discussed, results in tremendous additional efforts during recycling. Regulations at the country level can significantly enhance the efficiency of battery recycling. The Chinese government issued the Interim Measures requiring battery manufacturers to keep their products standardized.[87] The Measures recommend cooperation between battery manufacturers and new energy vehicle manufacturers for easy tracking of battery life cycles.[87] The European Commission proposed to increase the transparency and traceability of batteries throughout the entire cycle life by using new IT technologies, such as Battery Passport.[88] The relatively immature technology, and limited investment and profit are several other challenges of the LIB recycling. Financial and strategic support from governments can help attract more investments, which will stimulate the scaling up of the LIB recycling industry and technology improvement. The European Commission proposed mandatory requirements for batteries on the EU market, such as including a minimum amount of recycled components in new batteries.[88] Canada supports the development of recycling by funding several LIB recycling companies.[89] The US intends to establish federal policies to promote LIB recycling, and targets to recycle 90% of batteries in consumer electronics, EVs, and grid storage by 2030.[63] To summarize, governments across the world are enacting more supportive policies for the LIB recycling industry.

## 3. Perspectives

In summary, we have reviewed the status of EV batteries from the perspectives of environmental impact, electrode materials, supply chain, manufacturing, recycling, and governmental policies. Generally, the GHG emissions of EVs are lower than that of ICEVs due to high powertrain efficiency and zero tailpipe emissions, although producing an EV could generate more emissions than producing an ICEV because of the manufacturing of batteries and electric powertrain system. Electricity generation sources also largely determine the final emissions of EVs. Promoting the use of renewable energy to generate electricity is vital to maximizing the environmental benefits of EVs.

At the materials level, enhancing Ni content is the general trend of Ni-based cathodes, but Ni-free and Co-free cathodes have regained attention in recent years, especially LFP. Ni and Co are more vulnerable to supply chain disruption than Fe because they are more valuable and unevenly distributed. From the perspective of most EV consumers, high energy density is not critically necessary anymore if it can satisfy their daily commuting needs. Besides, high energy density is in less demand due to the fast charging capability of LFP and the expansion of the charging station network. Therefore, the advantages of low cost and safety of LFP stand out. However, Ni-based NMC or NCA materials still undergo rapid development and constitute a large portion of the EV battery market. To narrow the energy density gap between the Ni- and Co-free cathodes and Ni-based cathodes, we have provided several directions: 1) enhance the cell-level energy density by developing high-energy anode materials, such as Li metal and Si anodes; 2) optimize the form factor of the individual cell and battery pack design; 3) construct fast charging facilities and develop novel charging methods (e.g., wireless charging in **Figure 5**); 4) develop battery pack swapping methods for suitable EV types, such as public transportation. In addition, Li excess cathodes, such as LLOs and DRX, show high capacity at the research level without extensively relying on Ni or Co. However, the fast capacity decay, limited Li kinetics, and oxygen loss issues bring practical concerns on performance and safety, hindering their commercialization. We believe that the demand for both Ni-based cathodes and Ni- and Co-free cathodes will exist for specific applications. Sustainable cathode chemistry will be imperative as supply chain issues emerge.

A volatile supply chain could offset the performance benefit promoted by electrode materials. The distribution of the essential minerals for producing LIBs is scattered globally. No single country possesses all the essential raw materials for battery manufacturing. However, the global distribution of each specific metal is relatively concentrated. For example, DRC possesses 69.0–70.4% of global Co mining, and Australia shares 48.7–52.0% of global Li mining. Such uneven distribution of essential raw minerals may easily induce LIB supply chain disruption or price volatility due to potential geopolitical conflict or global challenges. Regarding the current mineral processing and battery manufacturing capacity, Asia countries, especially China, have been playing the dominant role globally. Developing technology and enhancing the efficiency of manufacturing are also significant besides production capacity. Public and private R&D has been the major driving force for the LIB cost reduction in the past. The US has a solid foundation for battery research and technology. The government continues to provide strategic and financial support for fundamental research. We believe the support for diversified fundamental battery research and lab-to-market development is of great

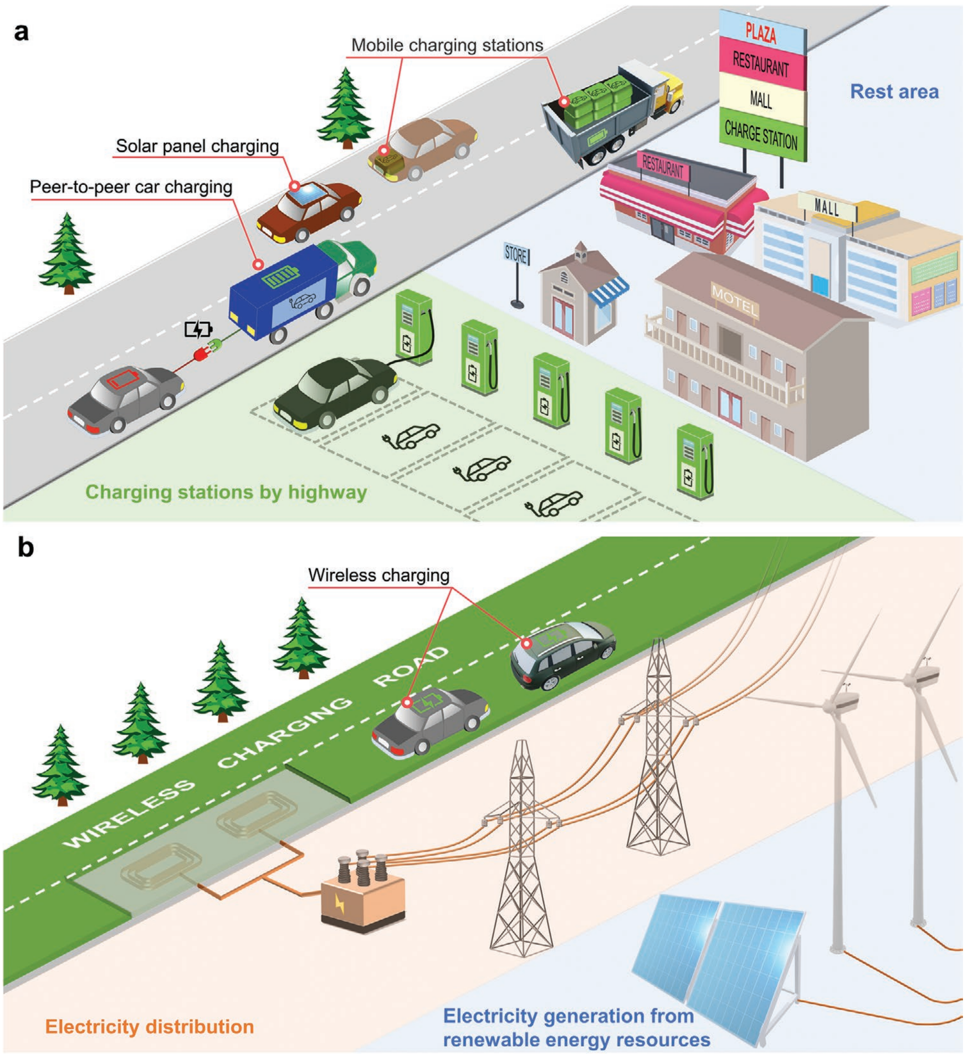


**Figure 5.** Scheme of visionary charging infrastructures in the future. a) Short-term scenario that requires less technology advancement. b) Long-term scenario that requires more technology advancement.

importance to the evolution of next-generation EV batteries. Consumers and existing battery products are less impacted by the LIB supply chain disruption than by fossil fuel shortages, but the stability of the supply chain is necessary for the long-term sustainable development of LIBs. A closer collaboration across the world and associated legislation are recommended to achieve a sustainable supply chain.

Strengthening the supply chain ensures stable primary battery production. Meanwhile, battery recycling becomes indispensable as mineral resources become limited and massive spent LIBs are generated. A common consensus is that recycling can relieve the shortage of raw materials in the long term. However, it is also forecasted that there could be a short-term deficit of several essential metals, such as Co and Ni, in the decade to come as discussed earlier. Therefore, we suggest that more incentive policies on both primary mineral production and recycling should be implemented. Large-scale recycling facilities and high throughput can effectively enhance the energy utilization efficiency and potentially lower the overall energy consumption of primary battery manufacturing. However, the environmental impact may vary with different recycling routes and battery chemistry. For example, recycling LFP even results in a net increase of GHG emissions compared to not recycling, no matter with which of the three major recycling routes.[8i] In addition, the traditional recycling methods could generate impurities and affect the performance of recycled products. Therefore, it is critical to monitor the impurity level and establish standards. More investigations on developing novel recycling approaches are expected to address energy-intensive and environmental issues of current methods. Besides, the unstandardized spent batteries from different battery manufacturers lead to extensive inefficient labor in pretreatment, such as sorting. Therefore, we suggest that regulations should be established to standardize the primary battery production.

Government policies and regulations are imperative not only for the recycling industry, but also for guiding the direction of LIB development. Several top economies in the world have shown their determination to secure domestic supply chains and develop local battery manufacturing. They have established aggressive targets of partial or full automotive electrification in the next couple of decades to meet carbon neutrality. At the cathode chemistry level, the US DOE set targets to eliminate Ni and Co in LIBs by 2030. We also want to highlight the significance of government regulations in preventing the "lock-in" of incumbent materials during the development of new materials.

LIBs have reshaped the way we transport and connect with each other since the first commercialization. In the past decade, LIBs have played a significant role in the revolution of smartphones. In recent years, LIBs have demonstrated remarkable success in EVs for a greener automotive industry. In the next few decades, LIBs will not only experience expeditious growth but also promote the construction of associated infrastructures. LIBs will shape the world into a more interconnected and smarter place with the rise of the 5G networks. Herein, we envision the potential development of energy storage technologies and EV charging infrastructures in the future (Figure 5). In the short-term scenario that requires relatively less technology advancement, EVs running out of electricity can be charged via peer-to-peer car charging (P2C2) by other vehicles (Figure 5a). The P2C2 requires low-battery EVs to be plugged into full-battery vehicles.[105] Low-battery EVs can also be charged during driving by the mobile charging stations, which are portable and temporarily installed in the EVs.[105] However, the weight of the portable charging stations may potentially lead to a low efficiency in vehicle mobility. Service vehicles will be on duty to provide P2C2 services or mobile charging stations. These charging methods are potentially not appropriate for routine charging but can offer rescue to EVs in an emergency. In the regions that receive sufficient sunshine, solar panel charging can be integrated on EVs to provide auxiliary support to the battery system. Sono Motors has announced their solar cell integrated EV Sion, which utilizes solar power.[106] Regarding the infrastructure, more charging stations can be constructed along the highways. More entertainment facilities will accompany the charging stations, such as restaurants and shopping malls, which can serve the people waiting for charging and boost the local economy. To summarize, we proposed several approaches to modifying the current EV charging methods based on relatively mature technologies. In addition, we conceived the long-term scenario that involves relatively significant adjustments to the current charging methods and infrastructures.

In the long-term scenario, both EVs and infrastructures will experience more technical breakthroughs, and there will be more interactions between them. Smart roads can be constructed to combine wireless charging with various functions, such as ice melting in winter and traffic condition monitoring (Figure 5b). EVs can be charged on the roads during driving, which will alleviate the anxiety of mileage. In addition, energy harvesting technologies, such as piezoelectric systems, can be integrated into smart roads to generate electricity based on the stress and motion of vehicles, which can enhance the energy utilization efficiency.[107] Several projects of smart roads integrated with wireless charging have been demonstrated in Europe and the US by Electreon Wireless Ltd and Integrated Roadways.[108] This approach might open the door to a broader range of automotive batteries with lower energy density but superior economic and environmental benefits. During the transition period, the roads can be partially reconstructed to accommodate one or two wireless charging lanes based on established infrastructures. Vehicle-to-everything (V2X) communication will be strengthened significantly thanks to the high-speed 5G network that is undergoing rapid development.[69] V2X can create an information network by exchanging data between vehicles and traffic systems. As a result, a smarter, safer, and more efficient traffic ecosystem will become a reality, including more mature autonomous driving, collision avoidance, and traffic jam forecast and reduction. The breakthrough on electricity generation is also expected for the greener long-term scenario. Electricity generation using fossil fuels is still a significant source of carbon emissions today. More renewable energy sources, such as wind and solar energy, will be applied in suitable regions for future electricity generation. Electricity distribution stations can be constructed accordingly along with the charging infrastructures to achieve more flexible and convenient charging. In all, we anticipate that EVs will be more interconnected to other facilities in the long-term scenario. The innovation of charging methods will also provide more room for battery materials development.

It is the worst of times when resources on the earth are becoming more limited, and the human society is facing energy crises and environmental degradation. It is the best of times when people are actively seeking strategies to address energy and environmental challenges with advanced and sustainable energy storage technologies. LIBs have changed and will continue to change the lifestyle of people and the ecosystem in the twenty-first century. In the upcoming decades, LIBs at the materials level will gradually shift to more sustainable chemistries. With extensive financial and strategic support from governments around the world, the LIB manufacturing and associated infrastructure construction will undergo a rapid growth. Climate issues can be alleviated by substituting ICEVs with EVs. Recycling will occupy a larger portion of the market and in turn enhance the economic and environmental efficiency of the battery production. The healthy growth of the LIB industry can never be solely accomplished by one specific field. We believe that building the synergies between different fields surrounding the LIB industry will greatly promote the development of LIBs for a more sustainable society. This Perspective aims to inspire more interdisciplinary discussion and investigations on the advancement of LIBs.

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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## Conflict of Interest

The authors declare no conflict of interest.

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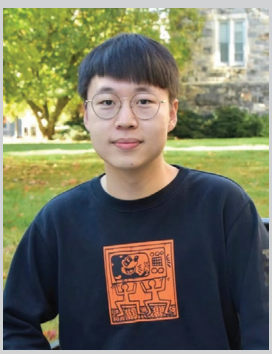

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