

Abstract: Battery electric vehicles (BEVs) have started to play a significant role in the transport sector and automotive industries. The broader market penetration of BEVs has still not been achieved due to significant barriers associated with initial costs and short driving ranges. The purchase price and a limited driving range are barriers that are inevitably associated with battery technology. Therefore, the growing demand for BEVs has expedited new innovative approaches to improve battery capacity and performance and to reduce battery costs. Considerable advancements have been employed to meet the challenges. However, there are still many challenges to make BEVs affordable and convenient for users. In this review, the main aims are to identify and address challenges by considering the prospects of BEVs in the future market and to explore the technological and financial difficulties of low energy density of battery materials, fast charging rate, battery lifetime, and cost-effectiveness, associated with effectively implementing and adopting BEVs. Moreover, potential suggestions are proposed for researchers, manufacturers, users, and government policy planners. Finally, a concrete conclusion is drawn by disseminating a vision about the future adoption of BEVs. This review of technologies, challenges, prospects, and potential solutions associated with BEVs could provide a base for effective strategic policy and could help policymakers to frame strategies for adapting and achieving targets. This review could help to achieve sustainable BEV transport and to adopt next-generation green vehicles.

Keywords: battery electric vehicles; driving range; energy density; fast charging; cost-effectiveness; sustainable transportation

1. Introduction

Internal combustion engine (ICE) vehicles account for a significant amount of carbon dioxide (CO₂) emission, which has severe consequences for global warming [1,2]. The transportation sector generates around 24% of the global CO₂ emissions from fuel combustion [3,4]. Driven by pressures from the energy crisis and environmental pollution, automotive manufacturers have led significant advancements in battery electric vehicle (BEV) technology [5,6]. Electric vehicles (EVs) are ahead of ICE vehicles in the sense of carbon emission and fuel consumption [5]. EVs present a solution to the problems created by ICE vehicle exhaust emissions, for example, ecological imbalance, climate change, and current living conditions [6]. However, in addition to these advantages, a few major drawbacks hold back the development of EVs, including high purchase cost, limited...
driving range, poorly designed charging infrastructure, etc. [7–9]. The EV market is still immature due to the scarcity of vehicle charging infrastructure, and, in terms of EV market penetration, consumers find this to be a major disadvantage of EV over ICE vehicles [10,11].

Most of the challenges of EVs are associated with the EV batteries. The critical component of a BEV is the rechargeable battery, since, for a long driving period, it needs a higher capacity [12]. Energy storage systems (ESSs) such as nickel-based, lead, and lithium-based systems are the most common; for instance, nickel cadmium (Ni-Cd), nickel metal hydride (Ni-MH), lead-acid (Pb-acid), and lithium-ion (Li-ion) batteries (LIBs) have been utilized in BEVs so far [13]. Table 1 summarizes the technical specifications of some periodically developed batteries. To find a safe, durable, and user-friendly storage technology with high energy density and fast charging, battery technologies have evolved from lead-acid to nickel-based to lithium-based types [14]. Recently, Pb-acid batteries have been used to power the components of combustion engines, specifically the starter, rather than BEVs. Though it has the advantage of being easily recyclable and producible, the primary barrier to using a Pb-acid battery is its bulk weight. It was used as the main energy storage system in BEVs, but after the 1980s, other technologies were introduced and have been used until now due to higher efficiency [15].

Table 1. Basic technical characteristics of different battery types used for EVs up to the use of Li-ion batteries (information collected from [13]).

<table>
<thead>
<tr>
<th>Battery Types</th>
<th>Specific Energy (Wh/Kg)</th>
<th>Energy/Volume (Wh/L)</th>
<th>Power/Weight (W/Kg)</th>
<th>Self-Discharge Coefficient (% per 24 h)</th>
<th>Recharging Cycles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb-acid</td>
<td>40</td>
<td>70</td>
<td>180</td>
<td>1</td>
<td>500</td>
</tr>
<tr>
<td>Ni-Cd</td>
<td>60</td>
<td>100</td>
<td>150</td>
<td>5</td>
<td>1350</td>
</tr>
<tr>
<td>Ni-MH</td>
<td>70</td>
<td>250</td>
<td>1000</td>
<td>2</td>
<td>1350</td>
</tr>
<tr>
<td>Li-ion</td>
<td>200</td>
<td>270</td>
<td>1800</td>
<td>1</td>
<td>1000</td>
</tr>
</tbody>
</table>

Nickel-based batteries were introduced to replace Pb-acid batteries. In addition to other advantages, Ni-Cd accumulators have the significant characteristics of higher storage density and a lifespan from around 500 to 1000 charging cycles [16]. In the past decade, nickel-based batteries have been used in most commercial EVs, especially Ni-MH batteries for propulsion [17]. However, Ni-based batteries have serious drawbacks, such as poor charge and discharge rate and decreasing performance in cold weather. Moreover, Ni-Cd batteries have been banned because of their toxic components [16].

Because of their long lifespan and relatively high energy density, LIBs are widely accepted [18]. Lithium is released from a solid lattice (intercalation) in lithium-ion cells. Intercalation significantly helps in increasing the number of charging and discharging cycles. By the time Li-ion battery systems started to reveal their potential, other ESSs had almost reached the pinnacle of their development [19]. A high-volume market entry of EVs is possible only because of the efficiency of Li-ion battery systems in terms of specific energy [20]. However, to meet the minimum criterion of driving range, LIBs still need huge advancements in terms of technological and financial points of view [21] in order to compete in tomorrow’s markets. Increasing energy density, reducing charging times, and increasing end of life (EoL) are the major technological challenges, whereas the initial battery cost which the price of a BEV largely depends on, is the prime concern from the financial outlook.

Extensive research has been devoted to increasing energy densities, including the search for new electrode materials and developing hybrid battery systems [22]. At a practical level, hybrid battery systems can perform much better as compared with LIBs. Hybrid battery systems include an organic polymer, for example, polyaniline vanadium oxide (PAni/V₂O₅). These hybrid systems have better intercalation capacity, cycle life, electrochemical performances, and conductivity of batteries as compared with traditional LIBs. Liming Jin et al. (2018) reported on a hybrid battery system that contained a lithium iron phosphate/activated carbon-based cathode and a Li-doped hard-carbon anode with
organic electrolyte for energy density improvement [23]. Moreover, the correct ratio of porosities for enhancing the energy density of Li-ion batteries [24] and how different types of templating can increase the electrochemical performances have been described in some studies [25]. In addition, increasing the cell output voltage can significantly increase the energy density of LIBs [26]. Additionally, some automotive companies have adopted various materials and technologies for increasing energy density and reliability, for example, the adaptation of positive electrodes developed with Ni-Co-Mn and coated-system cells by the Research and Development sector of the renowned automobile manufacturer Nissan for high energy density and reliability. In recent years, many researchers have reviewed and analyzed the progress in understanding different aspects of fast charging in their publications. Liu et al. (2019) reported some major strategies for increasing electrolyte conductivity and capability of electrodes, which were discussed briefly in this work [27]. The recent developments in battery materials to improve mass transport in electrolytes and transfer of charge in electrolytes were discussed by Liu et al. (2019) [27]. Meanwhile, when BEV batteries reach their EoL, the battery can either be used for alternative applications or raw materials can be restored by recycling. Global BEV makers, especially Bayerische Motoren Werke AG (BMW), Groupe Renault, Nissan Motor Co., Volkswagen, BYD Auto Co., and others, have been exploring different applications of second-life EV batteries [28]. However, renowned EV manufacturer Tesla claimed that batteries used in their EVs are not repurposed for a second life or alternate use but will be recycled directly. In literature, recycling has been recommended as the prime strategy of EoL for Li-ion batteries [29], but there are many drawbacks making the recycling process less justifiable. Ramoni et al. (2013) reported on remanufacturing as an alternative to recycling [30].

The commercial viability of BEVs depends significantly on the cost of batteries. Relatively lower operating costs as compared with those of ICE vehicles is one of the key benefits of BEVs. Nevertheless, the initial purchase cost is still the major barrier to widely adopting EVs. As the price of BEVs directly depends on the battery cost, reducing the battery cost makes BEVs more affordable.

In this review, the prospects of BEVs in the global market are anatomized. Moreover, technological and financial limitations of the current battery technologies associated with the global implementations of BEVs are addressed. Recent progress to meet the challenges are critically analyzed, highlighting the possible directions for future research. This study solely focuses on the challenges, current progress, and future recommendations of EV battery technology.

Following the Introduction, in Section 2, the prospects of EV batteries are discussed along with a comparison between different EV models and the development in EV batteries over time. Challenges, including both technological and financial, are addressed in Section 3. In Section 4, we describe the current progress with respect to the challenges mentioned in the previous section. The final section concludes the overall study including future suggestions and a discussion on the possible scope of research to overcome the challenges.

2. Prospects of Electric Vehicle Battery

BEVs are anticipated to surpass the extensive use of ICE vehicles within the next decade and to play a vital role in reducing carbon pollution in the road transport sector. Many developed countries, including UK, France, Denmark, The Netherlands, and Sweden, have banned ICE vehicle sales in upcoming years [31]. Meanwhile, targets for adopting EVs have been set by several governments around the world. However, to compete with ICE vehicles in the global market, BEV manufacturers must deal with some major technical and financial difficulties. Low driving range, long charging time, and high initial battery cost of BEVs are the most concerning factors that hinder the mass acceptance of BEVs as compared with ICE vehicles [32]. A battery is the sole propulsion source for BEVs. Different types of batteries have been proposed for BEVs. Energy density, charging and discharging time, and per kWh cost are the critical parameters for a comparison of batteries [33]. The United States Advanced Battery Consortium (USABC) set a short-term objective of 150 kW/kg
power goal and a long-term objective of 200 kW/kg to enable long-term commercialization of BEVs. However, the technology is still a long way from reaching this aim in 2022 [34]. Battery capacity and the speed of the charger are significant factors influencing the amount of time to charge a car. A simple formula for the calculation of the charging time is:

\[
\text{Charging time} = \frac{\text{Battery capacity (kWh)}}{\text{Charging power (kW)}}
\]

(1)

This calculation results in the hours required to charge a car battery from totally discharged to fully charged. Here, charging power, or even charging speed, is the quantity that measures the amount of effective energy per unit of time transferred from the charging station to the battery of the car. Ideally, it could be the same as the charging station power, but it is almost always limited by a series of factors, including charging station power, maximum charging power of the machine, maximum current of the charging cable, and grid energy availability. The time it takes to charge a Tesla Model S with a standard wall charger is 8 h, while it takes 1 h to charge with a supercharger. However, at least 2 h is needed to fully charge a discharged battery, even with the adequate charger [35]. Table 2 compares the charging times of three different models of EVs.

Table 2. Charging characteristics of three different models of EVs (information collected from [36]).

<table>
<thead>
<tr>
<th>Battery Capacity (kWh)</th>
<th>Range in One Full Charge (km)</th>
<th>Empty to Full Charging Time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Slow</td>
<td>Fast</td>
<td>Rapid</td>
</tr>
<tr>
<td>3.7</td>
<td>7</td>
<td>43–50</td>
</tr>
<tr>
<td>13.8</td>
<td>24</td>
<td>250</td>
</tr>
<tr>
<td>Mitsubishi Outlander PHEV</td>
<td>13.8</td>
<td>Not usable</td>
</tr>
<tr>
<td>Nissan Leaf</td>
<td>24</td>
<td>4</td>
</tr>
<tr>
<td>Tesla model S</td>
<td>43–50</td>
<td>4</td>
</tr>
<tr>
<td>40</td>
<td>143</td>
<td>6</td>
</tr>
<tr>
<td>75</td>
<td>238</td>
<td>5</td>
</tr>
</tbody>
</table>

The Tesla model S supports the 250 kWh charging port, however, it needs around 1 h to charge [37]. One report has stated that retired Li-ion batteries from BEVs will be growing at an exponential rate. By 2030, the total amount of EV batteries reaching EoL is expected to reach 12 million tons per year [38]. Recycling can be an option, but in sense of circular economy, it is the least sustainable measure and should be the final step if the batteries can not be used for other applications. Therefore, repurposing and remanufacturing for second life should be considered for retired EV batteries before recycling. However, battery chemistry significantly affects the decision of whether to recycle or repurpose a second life for a retired EV battery [39].

As previously mentioned, the total cost of a BEV highly depends on the price of the battery; reducing the battery cost is a major concern. The maximum price evaluated by the USAB with an important market share of BEVs was USD 150/kWh (with a long-term goal of USD 100/kWh). In the past decade, the prices of BEVs have fallen from USD 1200/kWh to USD 137/kWh [40]. However, the price below USD 100/kWh has to be achieved. Lastly, the cycle life is a crucial criterion for battery selection. An inferior lifetime of the battery as compared with the life of the vehicle will result in changing a battery after a few years, and the overall cost of ownership of the vehicle will increase immensely [41]. Battery lifecycle depends significantly on how the battery is used (e.g., charging and discharging rate and temperature). The battery management system is important for improving lifespan and safety. With a better battery management system, battery lifespan and cost can be improved in any technology [42].

At present, Li-ion batteries are the most used batteries in EVs. The demand for Li-ion batteries will increase with an increase in BEV sales. A primary barrier for using BEVs is the driving range which is directly affected by the relatively low energy density of the batteries currently used in such vehicles [17]. In addition, the slow rate of charging has put BEVs at a disadvantage, since the driving range from one charging cycle is still lower...
than the expectations of manufacturers, and BEVs need the adaptation of fast charging processes [43]. EoL of batteries is a major concern, as most BEV batteries need replacement after 8 years or 160,000 km, which is not feasible as compared with ICE vehicles [44]. In the last decade, there have been significant improvements in EV battery technology, which has gone through a few development phases, notably in energy density, fast charging, EoL, cost-effectiveness, and safety. Figure 1 shows the timeline of EV battery development.

![Timeline of EV Battery Development](image)

**Figure 1.** Tentative timeline of EV battery development.

Previous EV battery technology seemed to rely on Pb- and Ni-based batteries; the present EV industry has focused on Li-ion batteries. The future of battery industries is going through revolutionary research and development; however, Li-ion batteries are still a better option.

### 3. Challenges of the Electric Vehicle Battery

#### 3.1. Technological Challenges

Limited driving range is one of the major concerns of customers regarding BEVs, which is solely associated with some technological limitations [4]. Major technological challenges that current LIBs are facing include increasing energy density with compact battery packs and a slow charging rate that requires a long time to charge.

A key concern for increasing the limited driving range of BEVs is to improve the energy density of LIBs. Gasoline provides a nominal energy density of 13,000 watt-hours per kilogram (Whkg\(^{-1}\)), and EV battery cells reach 140 to 170 Whkg\(^{-1}\) [17]. Typically, the resulting battery pack provides 30–40% lower specific energy. To double the energy density in the next ten years, battery packs would need to store only about 200 watt-hours per kilogram weight of the battery. If it is assumed that 20–25% of the total weight of today’s typical small cars is because of the battery weight, a driving range of 300 km (about 190 miles) can be managed by doubling the energy density. A battery’s amount of power supplied per kilogram of mass, also called specific power, is addressed well by current battery technologies. Specific power has a specific importance for hybrid vehicles that discharge slowly. Specific power has relatively lower importance than specific energy for EVs. The manufacturers have established design parameters for EV batteries to optimize the trade-off between specific power and energy. Currently, the performance of batteries concerning specific power meets that of ICES. Therefore, increasing specific energy for certain power levels has attracted the attention of researchers.

The issue of long charging times is one of the key limiting factors for electric vehicles. Most BEVs require 2 to 6 h to fully charge (depending on the charger type) the battery, which results in owner anxiety regarding range issues [45]. One way to solve consumers’ range anxieties could be fast charging, which would also increase the acceptance of BEVs [46]. The California Air Resources Board (ARB) has defined fast charging as a 10 min charge that gives a vehicle the ability to reach 100 miles of driving range [47]. According to the
U.S. Advanced Battery Consortium (USABC), fast charging is the ability to gain 40% of the initial charge of a battery in less than 15 min [48]. This means that battery electric vehicle (BEV) battery charging times need to be reduced to the 5–10 min range to compete with petroleum-based vehicles. Three types of charging systems for an EV battery have been identified, as follows: Type 1 is standard charging, which has a charge power less than 5 kW; Type 2 is fast-charging, which has a charge power that ranges between 5 kW and 50 kW; Type 3 is super-fast charging, which has a charge power greater than 50 kW [49]. Type 3 charging consists of an exterior charger. Because of size constraints, carrying onboard power electronics required for Type 3 charging is impractical. As fast charging includes the simultaneous development of battery cells, including electrode materials, power systems, charging piles, electric grids, etc, it is a complicated and systematized obstacle to overcome for BEVs [50].

With the adoption of BEVs, in order for the auto industry to become environmentally friendly, it is essential to have a well-defined EoL plan for the discarded battery cells of EVs [1]. The options to properly handle the spent Li-ion batteries include remanufacturing, recycling, or repurposing. The life of the batteries is extended by remanufacturing or repurposing, but recycling closes the loop by returning raw materials to the value chain. There are three types of recycling processes available: pyrometallurgy, hydrometallurgy, and direct recycling. However, all the current recycling technologies have multiple drawbacks [51] (Table 3).

Table 3. Recycling technologies and drawbacks.

<table>
<thead>
<tr>
<th>Recycling Technology</th>
<th>Major Drawbacks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pyrometallurgy [52]</td>
<td>• Operates in high temperature</td>
</tr>
<tr>
<td></td>
<td>• Involves direct smelting</td>
</tr>
<tr>
<td>Hydrometallurgy [51]</td>
<td>• Produces soluble sulphur salt which can be difficult to handle higher cost ratio</td>
</tr>
<tr>
<td></td>
<td>• Time consuming</td>
</tr>
<tr>
<td>Direct Recycling [53]</td>
<td>• Direct contact with lead content which is carcinogenic</td>
</tr>
</tbody>
</table>

The large-scale development of second life battery repurposing could be hindered by higher battery refurbishment costs such as transaction costs and collection costs and the variable behaviors of battery quality, welfare, and lifetime issues [54].

3.2. Financial Challenges

BEVs are considered to be an environmentally friendly system for transportation by reducing carbon emissions and improving air quality. However, the transition from a petroleum-based transport system to an electric mobility system faces a variety of obstacles. One of the key problems for this transition is the unavailability of EVs as compared with typical IC engine vehicles. The battery of an EV accounts for almost 50% of the total cost of the EV and is the most expensive component; thus, the affordability of this type of vehicle linearly depends on the affordability of a battery. Therefore, reducing battery costs is the main concern for BEV manufacturers.

4. Progress in Technological and Financial Challenges

4.1. Progress in Energy Density Enhancement

Rechargeable Li-ion batteries with higher energy density have become desired because of the increasing demand for BEVs in markets. However, increasing the energy density of Li-ion batteries is not an easy task. The total LIB research community is working on it. Current Li-ion technology is based on intercalation cathode (positive electrodes of LIBs) chemistry, which leaves little room for further enhancement in the energy density because the specific capacities of these cathodes approach the theoretical levels. In electrode
research, a lot of the focus has been on cathode materials as they significantly contribute to the battery performance from an electrochemical perspective. Recently, efforts have been made to improve the energy density of Li-ion batteries. Most of the efforts have involved developing electrode materials with higher energy densities by holding more charge in a fixed volume. In recent years, the scientific community has been prioritizing the design of porous electrode materials so that oxygen and electrolytes can be rapidly transported and the cathode can accommodate solid reaction products, which can be an efficient approach to increase energy density in Li-ion batteries. Madhav Singh et al. (2016) reported that the electrode structure could be stabilized, and the electrode kinetics could be improved by introducing optimal porosity. They also stated that the porosity of the cathode could minimize the aging losses during cell cycling, and energy density could be improved [24]. Moreover, modification of the cathodes by templating has been found to be effective [25]. Vu et al. (2012) discussed the synthesis of different types of porous electrodes and the different types of templating (e.g., soft, hard, colloidal crystal, and bio-templating) and their effects on the electrochemical performances of electrodes. They also described the effects of non-templated porous materials and electrode materials with hierarchical porosity [25]. Previously, many researchers have reported on the effects of pore size on electrochemical performances of batteries. On the one hand, according to Tungjin Lee, to perform high usage of Li/MnO$_2$, porosity and distribution of pore size were the main factors that determined the discharge rate. Therefore, cathodes with higher porosity exhibited higher electrode utilization at high discharge rates [55]. On the other hand, in the case of Sn-Co alloy film electrode that was electroplated within a colloidal crystal template supported on a Ni-coated Cu sheet, better cycle performance was observed for materials with smaller macropores (180 nm vs. 500 nm) [56].

The volume energy density of batteries is an important parameter that should be considered to design a porous cathode [57]. It refers to the amount of energy that can be contained within a given volume. Higher volumetric energy density allows EVs to contain comparatively more energy (increased driving range) without increasing the size of the battery pack. The compaction of porous materials and volume changes during charging/discharging should be considered to design a battery pack. Increasing the cell output voltage could be one possible solution to increase the energy density of batteries with compact size. Ongoing research is being conducted to explore >5.0 V cells, but the traditional electrolytes have narrow electrochemical stability which is the reason behind limiting advances. Recently, Chen et al. (2019) reported on a 5.5 V electrolyte (1 M LiPF$_6$ in FEC/FDEC/HFE with LiDFOB additive) that enabled LiCoMnO$_4$ cathodes with 5.3 V to supply 720 Wh/kg energy density for a thousand cycles and graphite LiCoMnO4 cells with 5.2 V to supply around 480 Wh/kg energy density for a hundred cycles [26]. Additionally, using hybrid electrode materials could be another possible way to increase energy density in Li-ion batteries. Previously, a special battery system with both a hybrid cathode and a hybrid anode (LiCoO$_2$ + LiV$_3$O$_8$ as cathode and graphite + Li-metal as the anode) was developed by Bae et al. (2019) [58], and the system was analyzed and compared with the systems that were currently in use. Adding LiV$_3$O$_8$ increased the discharge capacity of LiCoO$_2$ + LiV$_3$O$_8$ cathode from 142.03 mAh/g to around 182.88 mAh/g which indicated around a 30% improvement. Wang et al. (2020) designed a hybrid cathode electrode with LiFePO$_4$ (LFP) and graphite which operated with a Li$^+$ and PF$_6^-$ intercalation/deintercalation mechanism [59]. The conductivity of hybrid electrodes could be accelerated by introducing graphite, which made it possible to extract/insert Li$^+$ ion quickly from/into the LFP phase within the range of 2.5 V to 4 V. Due to the high capacity and broadened range in the sense of voltage, the energy density of hybrid electrodes was enhanced.

However, increasing energy density demands extra space. Thus, the important point is to make small-sized batteries, while keeping the storage capacity the same as before. Therefore, BEV manufacturers have been trying to develop new electrode materials that can store a high amount of charge in a fixed volume. The Li-ion battery technology by Nissan employs such materials which allow storage of a higher density of lithium ions,
resulting in an increment in travel distance. Nissan’s Li-ion battery has adopted Ni-Co-Mn positive electrode material and laminated structure cells, making it possible to have high energy density and reliability. The advantages of laminated structure battery cells are high cooling performance and compact battery size. The battery capacity warranty guarantees 160,000 km or 8 years because of its high durability and reliability. In the first Nissan LEAF (equipped with a 24 kWh battery pack), each battery module was constructed with a four-cell configuration, with a total of 24 modules onboard the vehicle. In the second-generation LEAF, each battery module was constructed with an eight-cell configuration, and the vehicle was equipped with a battery pack having 40 kWh energy as standard, increasing the filling efficiency [60]. Storage capacity and reliability were retained in this new battery pack. For the first time, a new module structure was introduced in the 2019 LEAF e+ which allowed customization of the cell number. The total module length was shortened by adopting laser welding technology to join cells, allowing for optimal battery module shape and the best height for the vehicle platform and customer needs. Therefore, the adaptation of the above-mentioned techniques can help to overcome the limiting energy density barrier, which will eventually enhance the driving range of BEVs.

4.2. Progress in Optimization of Fast Charging

The number of manufacturers of battery-supported cars is increasing, but they all still have to deal with the highly consuming recharging time of batteries. Fast charging is still less favorable, as it reduces energy efficiency and power fade due to the high currents needed to accelerate the charging process. Fast charging has several problems; therefore, insights from the atomic to system level are required to understand and to improve fast-to-charge performance, as illustrated in Figure 2 [61].

![Figure 2. Li-ion battery fast charging considerations (adapted with permission from [61] for noncommercial use under a creative commons license, which permits use, sharing, adaptation, distribution, and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, 2022, Elsevier).](image)

The criterion for fast charging is to return 60% of a battery’s state of charge (SOC) within 6 min, which is difficult to achieve with less than $10^{-5}$ Scm$^{-1}$ ionic or electrical conductivity. Consequently, the considered critical value of ionic and electronic conductivity is $5 \times 10^{-5}$ Scm$^{-1}$ for fast charging electrodes. An electrode is not recommended in a fast-charging battery for lower ionic or electrical conductivity than the mentioned value. Higher intrinsic conductivity is necessary for an electrode with a high active area or larger
particle size [62]. High ion and electron transport kinetics need to be developed to obtain a composite electrode with high rate capability to serve the purpose of fast charging; high ions and electrons transport kinetics [63].

Scientists have continuously explored efficient anode materials and suggested some developed materials for Li-ion battery anodes. Because of its good reversibility and safety at low cost, graphite is considered to be the most used material in Li-ion battery anodes [64]. However, practical applications of anodes consisting of graphite for fast charging are hindered by poor intercalating capability [65]. Guo et al. (2011) stated that the poor rate performance of graphite was because of the slow lithium diffusion rate in the bulk carbonaceous material [66]. Therefore, a porous carbon anode was applied to reduce the lithium-ion diffusion pathway in bulk counterparts, which also increased transport channels for conducting ions [67]. Yu et al. (2015) reported on hierarchically porous carbon architectures embedded with hollow nano capsules, which delivered a very high capacity of 805 mAh g\(^{-1}\) at a current density of 0.1 A g\(^{-1}\), (38 s to fully charge) [68]. Surface modifications could be another way to improve the rate performance in anodes made of carbonaceous material [69]. The presence of heteroatoms in their surface can enhance the kinetics and electrical conductivity of carbon-based anodes.

Sahoo et al. (2015) revealed a simple technique for large-scale and environmentally friendly synthesis of boron-doped graphene as an anode material with high performance for use in Li-ion batteries [70]. A 548 mAh g\(^{-1}\) of discharge capacity at 100 mAg\(^{-1}\) was found in graphene doped with boron in anode material after the 30th cycle. Doping of boron increased specific capacity almost 1.7 times higher than the current density value as compared with pristine graphene. The nitrogen-doped graphene and boron-doped graphene showed high capacity, precisely 199 and 235 mAh g\(^{-1}\) at 25 Ag\(^{-1}\) (about 30 s to full charge), respectively. Huang et al. (2020) suggested that the performance in fast-charging of Li-ion batteries could be increased significantly by combining heterogeneous doping and porous structure [71]. A reversible capacity of 226 mAh g\(^{-1}\) at a current density of 20 A g\(^{-1}\) for about 40 s to full charge was found from prepared nitrogen-doped porous carbon nanofiber webs.

Considering the other side, research efforts have been made to identify new materials that could be used in anodes to achieve high-rate capacity. An intelligent approach to solve the slow charging barrier is to develop anode materials that consist of high ions with high electron transport rate. Li\(_4\)Ti\(_5\)O\(_{12}\) (LTO) is the most promising material to use in the anode of lithium batteries for fast charging. Studies have been conducted because of the outstanding structural stability in the intercalation/deintercalation process of Li-ion. However, unsatisfactory rate capability of pure LTO has been obtained due to the poor electrical conductivity as well as sluggish diffusion behavior of Li-ion [72]. To address these severe limitations, efforts have focused on increasing electrical conductivity by doping or limiting ion diffusion routes through the construction of nanostructured particles [73].

Xia and co-workers first described that carbon-coated nanoporous microsphere LTO exhibited a reversible capacity of 160 mAh g\(^{-1}\) at 0.2 C, and showed remarkable rate capability by maintaining 79% of the capacity at 20 C (vs. 0.2 C), as well as excellent cycling stability with a capacity retention of 95% after 1000 cycles at 1 C rate (vs. 0.2 C) [74], whereas rutile-TiO\(_2\) terminated LTO nanosheets have been reported to deliver a capacity of 110 mAh g\(^{-1}\) with an area loading of 10 mgcm\(^{-2}\) at 60 C [75]. LTO/graphene foam composite anode exhibits a capacity of 130 mAh g\(^{-1}\) at 100 C [76]. This kind of rate performance has been found because of the hybrid electrode’s porous structure and their higher electrical conductivity. Furthermore, other anodes with high performance have been explored to test fast charging. Research performed by Cho and co-workers demonstrated the preparation of disordered graphene-like and a 0.69 nm enlarged interlayered distance of MoS\(_2\) nanoplates, which delivered a reversible capacity of around 700 mAh g\(^{-1}\) at 50 C [77]. Luo et al. (2013) described that a capacity of 190 mAh g\(^{-1}\) at 60 C had been exhibited from 3D graphene foams loaded with mesoporous Fe\(_3\)O\(_4\) [78]. Li et al. (2008) found an anode constructed with mesoporous Co\(_3\)O\(_4\) with a higher capacity of 350 mAh g\(^{-1}\) at 50 C [79].
An anode made with silicon nanotube constructed with Si-SiOx double wall was proposed by Wu and co-workers, which showed a very high capacity of 540 mAhg\(^{-1}\) at 20 C because of the effective prevention of the reaction in the inner silicon wall and electrolyte by the outer wall of SiO\(_x\) \[80\].

LiMnO\(_2\), LiFePO\(_4\), Li(Ni,Mn,Co)O\(_2\), and LiCoO\(_2\) are the most used cathodes for Li-ion batteries \[81\]. These cathodes have low electrical conductivity and poor performance. The current studies have focused on making a conductive pathway for electrons and constructing diffusion channels for Li-ions. For example, Kang and Ceder achieved a capacity above 100 mAhg\(^{-1}\) at 60 C and 60 mAhg\(^{-1}\) at 400 C for a LiFePO\(_4\) cathode made by introducing a lithium phosphate coating on the nanoparticle surface of these materials where fast ion conduction occurs \[82\]. Another study found that a LiNi\(_{0.5}\)Mn\(_{1.5}\)O\(_4\) electrode coated with 1.0 wt.% zirconia (ZrO\(_2\)) could recover around 86% of the initial capacity after almost 1000 cycles at 40 C \[83\]. Nanoporous LiNi\(_{1/3}\)Co\(_{1/3}\)Mn\(_{1/3}\)O\(_2\) filled on vapor-grown carbon fibers has been reported to deliver 72 mAhg\(^{-1}\) at 180 C \[84\]. A capacity of 75 mAhg\(^{-1}\) has been found in the nanocrystalline LiCoO\(_2\) with a particle size of 17 nm at 100 C \[85\]. Carbon-coated LiMn\(_2\)O\(_4\) nanoparticle clusters have exhibited a capacity of about 80 mAhg\(^{-1}\) at 100 C \[86\]. Yoshida and co-workers stated that polymethacrylate-bond pyrene-4,5,9,10-tetraone (PYT) organic cathode delivered about 220 mAhg\(^{-1}\) at 30 C because of the high redox energy change of the core structure of PYT \[87\]. Furthermore, Billaud et al. (2016) reported that a reduction in the tortuosity of lithium-ion pathways among the active particles and a possible increase in insertion routes of lithium ions could be achieved by the approach of magnetic alignment on the orientation regulation of active particles based on the difference in particle sizes into specific directions. Consequently, a three times higher capacity was achieved in their contribution than that of non-structured electrodes \[88\].

In summary, constructing fast Li-ion and electron channels while maintaining the structural stability of materials is the best way to build effective electrodes for fast charging in Li-ion batteries. In terms of high-rate performance, electrodes having nanoporous structures with high conductive substrate are more favorable. Consequently, cathode and anode optimization strategies are analogous for Li-ion batteries built with liquid-type electrolytes. Moreover, Liu et al. (2019) reported on the recent growth in battery construction materials to overcome the barriers to transferring charges in electrodes and to mass transport in electrolytes and also described the important characterization approaches for material study \[27\].

4.3. End of Life Issue

The use of Li-ion batteries in electric vehicles will continue to expand because of the efficient energy storage mechanism of Li-ion batteries. However, the batteries that have fallen below regulatory standards and EoL values need to be improved for further use in on-road vehicles. The economic value of post vehicle batteries can be reclaimed by any of the following approaches: (1) Remanufacturing for the purpose of reuse in vehicles; (2) reengineering and repurposing for an off-road, stationary storage application; and (3) recycling, by disassembling the battery and safely separating chemicals and other by-products followed by the extraction of precious metals. Positive progress has been made in each of these post-vehicle application areas with profitable margins. To upgrade the EoL value of post-vehicle batteries, several techniques have been followed. According to the auto industry, around 80% capacity remains after a degraded battery is removed from an EV, indicating that the bulk materials in the battery are still active, even though this much capacity is insufficient to power the vehicle. Recycling of such batteries would reduce active bulks in the batteries to material constituents and would cause a total loss of the remaining available capacity of the batteries. Regarding whether the recycling process would be profitable or not, some constrains were reported by Jungst \[89\]. These included the market's capacity to occupy the vast amount of material found from recycling that could occur in the long run if it was not used directly in new batteries. Every material
 recovered from a recycled EV battery would have a different market size. However, if the market for recovered lithium became overloaded with recycled material, then, there could be a chance to collapse the price range. In addition, it could result in the inability to sell recycled material.

In an extensive study by Kesler et al. (2012), the authors compiled data on 103 deposits comprising lithium with a prominence on 32 deposits which had almost 100,000 metric tons or higher lithium resources in each deposit [90]. The collected data had various parameters, including dimensions, lithium content, geological type, deposition location, and recent production status. It was concluded that, during the twentieth century, electrification of the automobile will not be constrained by lithium availability. Furthermore, according to the exclusion principle regarding chemical process economics, materials with high prices tend to have limited markets, while materials with higher market capacity have lower unit prices. Thus, it would be profitable to retrieve lithium from EV battery recycling; Li-ion batteries contain only around 1 wt.% of lithium, indicating each kWh storage capacity contains almost 0.08 kg Li, which is around 1.5 kg per EV battery.

Given the above issues associated with recycling EV batteries, it is important to search for other EoL strategies as potential appropriate ways. One efficient solution could be remanufacturing the batteries for reuse in EVs. According to the auto industry, if the capacity of a battery decreases by 20% of its original capacity it is considered to have reached its EoL [91]. With such a high capacity remaining, it would be advantageous if remanufacturing of the battery after the first life cycle was possible. The process of restoring EV batteries to their initial condition can be done by remanufacturing with minimum investment [92]; complete disassembly of the EV battery will be needed in the process.

4.4. Progress for Financial Challenges

The current cost of battery packs must be reduced for effective and broader commercialization of competitive battery electric vehicles (BEVs). For BEVs to compete with internal combustion engine (ICE) vehicles in terms of cost, the battery cost needs to be reduced below US 150 per kWh. By varying the physical and electrical properties of Li-ion batteries, cost reductions could be made. The steps in Li-ion manufacturing vary for individual manufacturers. For the prototype, the desired pair of electrodes and other chemicals are chosen which are used further to build the cell. The performance and potential of the cell will differ depending on the selection of the mentioned elements and their arrangement. In manufacturing, electrodes coated with chemicals are the most common steps. Drying steps are used for solvent removal. To form a complete cell assembly, cathodes and anodes, electrolytes, separating materials, binders, etc. are mandatory. After assembling, testing, and grading the cell, the full battery pack is packaged containing single or multiple cells.

If we consider the process from the same point of view of manufacturers, the goal is to reduce the inconsistency of each step to the minimum level to obtain the final product within a range of targeted values. Applying several techniques in the manufacturing process, as described in the following paragraphs, could help achieve this goal. Battery manufacturing processes need to change from statistical process control (SPC) to advanced process control (APC) to reduce process variability. In addition to the semiconductor industry, the benefits of APC have been well documented in other industries, for example, biopharmaceuticals [93]. Table 4 summarizes the progress and limitations in technological and financial challenges.
Table 4. Progress in overcoming different issues for EV batteries.

<table>
<thead>
<tr>
<th>Technological/Financial Challenges</th>
<th>Actions or Progress to Overcome the Challenge</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy density</td>
<td>Developing electrode materials holding more charge in a fixed volume enhance energy density. Using effective porous electrode material to accommodate solid reaction products.</td>
<td>Increasing energy density needs additional space. Other concerns with energy density are durability and reliability.</td>
</tr>
<tr>
<td>Fast charging</td>
<td>Improving ionic and electrical conductivity, introducing porous carbon anode to reduce li-ion diffusion pathway along with boron doping in the anode.</td>
<td>Reduces energy efficiency and power fade because of the high current used to accelerate the charging process.</td>
</tr>
<tr>
<td>End of life</td>
<td>Remanufacturing for reuse, reengineering for stationary energy storage, recycling by separating part by part, and recovering precious metals.</td>
<td>Each different material from the recycling process has a different market size, which will collide with each other, and can cause a sudden collapse in the price range.</td>
</tr>
<tr>
<td>Financial challenges</td>
<td>Variation in physical and chemical properties of battery component. Changing the manufacturing process from SPC to APC to reduce process variability.</td>
<td>Changing physical and chemical properties can affect other battery properties.</td>
</tr>
</tbody>
</table>

5. Potential Suggestions and Ongoing Research to Overcome the Challenges

5.1. Research on Technological Challenges

To increase the limited range of BEVs, improving the energy densities of LIBs is one of the major obstacles. For vehicles with comparable cost, the driving ranges of EVs are generally only around one-fifth of those of vehicles powered by internal combustion engines [94]. However, technologically developed countries such as China, USA, Germany, etc. reported record breaking sales of BEVs in 2021. In terms of EV sales volume and market share, China continues to be the largest EV sales country, with an annual growth rate of 137.5% and a market share of 58.2%, placing USA and Germany in the second and third positions, respectively [95]. EV producers continue to try to increase the driving range of EVs; the driving ranges of the latest EVs are 804 km for the Cybertruck (Tesla), 930 km for the NIO ES7 (Chinese SUV), and 600 km for the BYD Han (Chinese car), which is far beyond that of traditional fuel vehicles. However, for a sustainable and long-term market penetration of EVs, ongoing research on different aspects of EVs has to be accelerated. The engineering behind simultaneous performance enhancement and cost reduction is discussed in the following subsections.

5.1.1. Energy Density Improvement

Improving energy density and lowering the relative weight of the inactive components (for example, binder, a conductive additive, current collector, and separator) can be done simultaneously with electrode engineering. Another approach is the utilization of active materials with improved energy contents. However, a simple approach for increasing a cell’s energy density is to optimize cell engineering by improving the active materials’ volume ratio in a battery pack. Numerical modeling has also been used in cell engineering to study electrochemical systems. The operation of an LIB is in agreement with the porous electrode theory and electrochemical reaction thermodynamics, and the scientist Newman advanced the governing equations [96].

Efforts to build electrodes with variable porosity have only led to negligible improvements as compared with electrodes with constant porosity in terms of energy density. This indicates that decreasing tortuosity is more important [97]. A numerical simulation method has been applied to investigate the relationship between energy and power density in \( \text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2 \) (NCA)/graphite cells with thick electrodes [98]. As the carbon rate
increased, the limiting constraints became more prominent in thick electrodes, resulting in saturated energy density vs. thickness graphs. The surface layer saturated with Li-ion in the solid phase and the depletion of Li-ion in the electrolyte phase resulted in underutilization of active materials. To understand the improvement of energy density, many different porosity gradients have been modeled.

Figure 3A indicates four linear (i–iv) and six second-polynomial (v–x) variations of the porosity gradient. Figure 3B shows the energy density relation with the porosity gradient. Four gradients have higher energy density than the baseline, but the improvement is insignificant. However, a poorly designed porosity gradient can be responsible for decreasing the energy density, as shown by the porosity gradient (v) due to worse electrolyte depletion [98]. It should be noted that the Bruggeman relation estimates the Li-ion diffusion length. The Bruggeman relation assumes that the insulating phase has a low volume percentage represented by random and isotropic spheres. This relation may be invalid for the real electrode structure which has more complexity, and the real cell performance might differ from the results found with simulation [99].

![Figure 3. (A) Four linear (i–iv) and six second-polynomial (v–x) variations of the porosity gradient; (B) 150 μm thick cathode's energy density under 1.5 C discharge. Adapted with permission from ref. [94] (Jianlin Li et al., 2017).](image)

Slower diffusion of Li-ion is the rate-limiting process in power performance for thin electrodes. However, as electrode thickness increases, Li-ion transport restrictions in electrolytes become progressively critical [100] since the diffusion time in the liquid phase is no longer negligible because of the large increase in diffusion length in the thick porous electrodes. Consequently, it is important to design thick electrode architectures to take advantage of the increased energy density available from the higher active material volume ratio as well as to minimize the tortuosity and transport limitations that can affect power performance. Different designs have been suggested including graded porosity and hierarchical architectures [101–103], and many simulations have shown that by decreasing the Li-ion diffusion, active material with smaller particle size can assist to reduce capacity loss and polarization, particularly at a higher rate of discharge [104]. However, the packing structure of an electrode is affected by particle size variations. It can contribute to variable pore size and distribution, along with differences in contact resistance between the current collector and the electrode. Improvements in transport properties and battery performance can be achieved by leveraging these variations. Multi-layer structural designs have been introduced to balance different particle size potential advantages. Using parameters such as particle size to tune the electrode architecture could enable flexible cell optimization, meeting different application requirements. Separate or simultaneous coatings can create a multi-layer architecture that allows different electrode components or each layer formation for certain purpose. For example, to ensure good adhesion between the current collector and the electrode, the bottom layer might have a larger binder concentration [105]. An electrode with a homogenous structure could be obtained by utilizing particles with
spherical shape, which could minimize tortuosity [106]. A heterogeneous structure has been found with anisotropic tortuosity for non-spherical particles (example, graphite) [107]. Diffusion of Li-ion from the bottom layers to the top layers could be beneficial in aligning the direction perpendicular to current collector with the lowest tortuosity. Introducing a well-controlled pore former that can be eliminated while sintering has been shown to be an effective method to create straight channels in electrodes [101,108]. Linear channels can also be made by laser structuring [109], co-extruded electrodes with low-density areas beside higher density areas, or 3D printing interdigitated electrodes [110]. However, these special structures need a better understanding of the current distribution and its effect on lithium plating. They could also increase manufacturing costs.

In addition to electrode engineering, an effective way to increase LIB energy density is by utilizing electrode materials with higher energy density. A value >360 mAhg\(^{-1}\) has been achieved for reversible capacity from commercial graphite anodes used in LIBs, which is around 372 mAh/g, the theoretical value for LiC\(_6\) [111]. Even the best commercial cathodes are limited to gravimetric capacities around 200 mAh/g [111,112]. The choice of cathode also limits cell voltage. Therefore, recently, there has been increased interest in building high-voltage and high-capacity cathodes to increase the specific energy content of LIBs [113].

Cathode materials must also satisfy stability, including thermal and electrochemical, sensible cost, sufficient conductivity, and other design criteria. Sony introduced the first commercial LIB in 1991, in which LiCoO\(_2\) was used as the active material of the cathode. LiCoO\(_2\) is limited to an amount of 140 mAh/g of reversible capacity [114,115], but an improved capacity of layered oxides could be achieved by partial substitution of Co using different metal ions (Figure 4) [116,117]. In particular, NMC cathodes that have high nickel contents (LiNi\(_x\)Mn\(_y\)Co\(_{1−x−y}\)O\(_2\) where x > 0.6) with reversible capacities >200 mAh/g have shown affirmative results for increasing the energy density in Li-ion cells and are replacing lower capacity chemistries for the next generation EVs [118]. However, several critical challenges with Ni-rich NMCs need to be addressed. To achieve a cell level energy density >250 Whkg\(^{-1}\), high voltage cycling (>4.4 V) is required, in spite of the NMC cathode being coupled with an advanced anode made of silicon-graphite composites [119]. Impedance rises and capacity loss have been reported to occur due to high voltage cycling [120]. An increase in impedance has been partly imposed on a rock-salt surface reconstruction layer due to the loss of oxygen and reduced oxidation states of transition metals [121]. High voltage cycling has also been shown to be responsible for decomposing current generation electrolytes and forming a surface reaction layer [118,119]. In particular, Ni \(4^{+}\) ion in contact with electrolyte has been shown to be unstable [118,122].

![Figure 4](image-url)  
Figure 4. (Left) Layered oxide cathodes crystal structure and, (Right) Average capacity and voltage of different layered oxide cathodes. Adapted with permission from ref. [94] (Jianlin Li et al., 2017).

Interest in studying the cathode electrolyte interface (CEI) has recently grown because of the complex interplay among phase change, electrolyte degradation, and transition metal dissolution [123]. Decomposition of electrolyte products and deposition on the surface of the cathode occurs in the form of LiOH, Li\(_2\)CO\(_3\), LiXPOFy, LiF, polycarbonates, and species, which are very specific to cathode and electrolyte material [124]. Doping [125],...
coating on the surface [126], and using electrolyte additives [127] are the common techniques used to improve surface properties. These methods generally result in a trade-off between maximum specific capacity and the coulombic efficiency of the first cycle for reduced impedance rise, thermal stability, and reduced CEI build-up to increase cell cycle life [128,129]. The development of compositionally graded cathodes with a lower amount of Ni at the surface is a better promising approach for reducing surface reactivity and prolonging the cycle life of Ni rich NMCs [130,131]. In general, a Mn-rich surface and a Ni-rich core are found in these particles. Mn$^{4+}$ shows electrochemically inactive properties. Thus, a stabilized electrode/electrolyte interface is found because of the Mn-rich surface, while the Ni-rich core enables high capacity and energy density. Cathodes with a Mn-rich surface show better thermal stability than their homogenous counterparts [132]. Ultimately, to produce NMCs that contain high nickel content with satisfying performance, a combined approach is required. For example, Al-doping [133,134] and surface coating [135] are techniques applied to boost capacity further and improve rate performance in NMCs with concentration gradients. A significant amount of attention has been given to NMCs with high lithium content (xLi$_2$MnO$_3$·(1–x) LiMO$_2$ where M = Co, Mn, Ni) due to a higher capacity (>250 mAh/g) at a comparatively high voltage (2.5–4.7 V) [136]. Despite the expectations for these cathodes, suffering from irreversible loss of capacity in the first cycle, increased impedance during the high-voltage cycling, and most importantly, a sharp drop in voltage profile (voltage fade) with cycling have been observed [137]. Voltage fade has been identified in structural and chemical changes including growth of oxygen, migration of Co and Ni to the bulk from the surface, reduction in Mn ion, and spinel-like phase formation [138,139]. A CEI formation mechanism with more complexity has also resulted from the phase change [140,141]. Coatings [142,143], synthesis routes [145], and electrolyte additives [146] are the common routes for stabilizing the surface of NMCs containing high lithium content. Most methods have been unsuccessful in preventing the underlying mechanisms that are responsible for voltage fade. However, compositions which combine small amounts of spinel domains into the layered structure are promising [147]. Stabilization in the structure cannot be achieved, but in applications with acceptance of some voltage fade or as part of the cathode blends of Li-rich NMCs with 10–30% Li$_2$MnO$_3$ could be used. Another strategy to increase capacity moves beyond layered transition metal oxides, i.e., cathode materials capable of multi-electron redox reactions [148]. Sulfur cathodes [149], materials that undergo conversion reactions such as FeF$_3$ [150], organic electrodes [151], and several polyanionic chemistries [152] are the leading candidates. While these rising cathodes hold promise, they face many barriers to commercial success. There has been no apparent winner in the race to replace layered transition metal oxides such as NMCs for Li-ion batteries. Regardless of the cathode chemistry, electrolytes with new formula or additives with improved effectivity are required to improve the cell voltage to 4.4 V or higher.

Cathodes with higher energy density need to be matched with anodes having equal energy. Manufacturers are trying to use alternative anode materials which are capable of increasing the energy density of the battery, while retaining other performances. Though conventional graphite-based anodes give two-fold specific capacity as compared with the best cathode available on the market, materials with higher capacity rather than graphite would still allow for lighter and thinner anodes, resulting in higher density batteries. The electrochemical reaction between graphite and Li occurs through intercalation and forms LiC$_6$ [153]. Alternatively, other elements (such as Sn and Si) can alloy with Li and form Li$_x$Si$_4$ and Li$_y$Sn$_5$ [154]. Though the capacities of these alloying materials seem very attractive, they are responsible for an identical set of problems during large-scale production implementation. Graphite can only expand 10% due to Li intercalation. Alloys formed with lithium such as Si can increase up to 300% in size and comprise the additional lithium [155]. A significant change in volume between charged state and discharged state causes mechanical failure, which results in the anode’s active material loss [156]. Therefore, to alleviate pulverization of the particle nano, Si is used. In addition, reducing the particle
size of Si to the nano domain results in distributing particle expansion and alleviating stress on the electrode throughout the whole electrode framework [157]. Alloying nano particles tend to conglomerate and ripen during cycling, making them insufficient when utilized alone [158]. The most straightforward approach to prevent agglomeration is by mixing Si nanoparticles in small quantities with graphite electrodes. A theoretical specific capacity of 1330 mAhg\(^{-1}\) results from 30 wt.% of Si/graphite composite which can operate up to 90 cycles while the capacity is limited to 500 mAh/g. [159]. By utilizing carbon nanotubes, graphene, and other oxide or carbon supports, nano Si architecture with more complexity have been developed and excellent capacity retention has been reported, but these results have been reported at a laboratory scale and larger scale work would be costly [160–163]. Improving the cycle life of Si anodes depends on forming a stable SEI that must be sustained throughout cycling. Although the additive fluorinated ethylene carbonate (FEC) has been used to form a more stable SEI [164], until now, the jury has been out regarding its effectiveness, since it is more likely to be consumed during cycling, initiating capacity fade for a long time [165]. Although there has been significant progress in this field, many barriers such as cycle life remain unsolved. In most of the literature, the results have been obtained from half cells and considered to be mostly Li. However, applications of Si in LIBs have been limited to those with lower Si content as well as short life cycle electronic devices. To fulfill 1000 deep charge/discharge cycles, a key breakthrough is required in passivating the Si surface. Other factors in addition to the aforementioned problems could also affect the use of Si in EVs, including nano Si is highly expensive, the complex electrode manufacturing process with Si as additive, obstacles in the pack and cell design to achieve effective volume expansion, etc.

Commercially scalable technologies for producing nanostructured materials with well-defined shape and size, with characteristic size bellow 150 nm, have also hindered the development of silicon anodes for Li-ion batteries. Ball milling is one of the traditional top-down methods which is inexpensive and produces highly agglomerated Si materials, with relatively large particle sizes and poor morphology and surface. Another explored method of synthesis action of silicon nanomaterials is chemical etching assisted by metals. Commercialization of this method is limited by various factors such as control over the morphology, issues regarding mass transfer, necessity of highly toxic reagents, and etching direction. Meanwhile, bottom-up approaches such as chemical vapor deposition (CVD) have received a lot of attention [166]. CVD-based synthesis produces well-defined particle sizes as well as a wide range of nanostructured structures such as nanowires (NWs), nanoparticles (NPs), and thin films [167]. Key barriers to using CVD methods are high capital cost, low production, and pyrophoric and risky gaseous Si feedstocks. A silicon carbon hybrid anode built with nanostructures with more Li-ion storage capacity might be the solution; however, there is the possibility of damage when the silicon expands. Cyclohexasilane (CHS) can be used to manufacture these nanostructures commercially due to the ability of readily functionalized, more auspicious reaction conditions and more ideal handling conditions. All of these attributes could end in roll-to-roll manufacturing and single-step processing. As a replacement to the existing manufacturing processes this would allow a reduction in costs and avoid using CVD methods which solve the high capital costs issue [168].

5.1.2. Fast Charging

Coating, introducing porous structure, and reduced particle size are methods to improve the fast charging of batteries, to reduce the packing density, and to obtain lower energy density [169–171]. Generally, a trade-off must be considered between fast charging and energy density when designing electrodes based on the application. The imagination of electrodes with high theoretical capacity and high lithium plating driving force is the solution to make a trade-off between power density and energy density. For example, graphite with a lithium plating driving force of only 120 mV readily deposits Li-ions on its surface, resulting in poor charging ability. To compare, Li\(_4\)Ti\(_5\)O\(_{12}\) provides a high
lithium plating driving force of 1.5 V (vs. Li/Li\(^+\)) and relatively low theoretical capacity (175 mAh g\(^{-1}\)), which makes the Li\(_4\)Ti\(_5\)O\(_12\) electrode available with a better rate performance and sluggish energy density. A higher rate performance and higher energy density can be achieved by using anodes constructed with red phosphorus composite because of its higher lithium plating driving force of 0.8 V (vs. Li/Li\(^+\)) and high theoretical capacity around 1400 mAh g\(^{-1}\) [62]. In addition, other effective ways to achieve high energy density as well as a power density energy storage system include introducing more ion and electron transport channels [62].

A fast charger configuration is one of the key criteria for implementing fast charging. Two possible configurations are available for fast chargers: one configuration is based on on-board AC/DC conversion (AC chargers); the second configuration is based on the power supply in a DC vehicle. In this latter option, an advantage could be weight reduction of the vehicle, but complexity increases for the requirements of the vehicle-to-grid communication; charging is dependent on the off-board charging device, which requires communication with the onboard battery management system (BMS) [172].

According to The International Standard IEC 61851-1, the first option is “Mode 3”: An AC supply network is used and the EV is connected to it directly using electric vehicle supply equipment (EVSE); to perform safety control, a pilot control circuit is introduced to verify that the connector has been correctly inserted into the vehicle inlet, to test the continuity of the earth circuit, and to provide a power-off switch without stopping the control circuit (in the case of pilot circuit failure). The second option according to IEC 61851-1 is “Mode 4”: The EV is connected to the power source through an off-board AC/DC converter and battery charger; “pilot” functionalities are enabled for Mode 4; for the BMS to control the off-board charger, a serial data communication line is required. Interoperability of EVs is a significant concern associated with EVs, charging station manufacturers, and global standardization authorities. Different DC fast charger topologies are in use today to harmonize connections and communication standards [174].

The establishment and distribution of ultrapowered charging stations is the ultimate goal for the future of EVs. We are not far from achieving this goal, as XPeng Motors (Chinese EV manufacturer) declared, in 2021, that its new S4 fast charging column (in operation) offers a maximum charging power of 480 kW. The Xpeng G9 was able to recharge a CLTC (Chinese equivalence to the Worldwide Harmonised Light Vehicle Test Procedure (WLTP)) range of 210 kilometres in five minutes with this ultrafast S4 charger.

5.1.3. End of Life

Currently, LIB recycling processes have mainly been aimed at precious metal recovery, and recovery of organic compounds has not been considered [175]. The leading element in the recycling process of Li-ion is cobalt; cobalt is rare as well as expensive, which makes the recovery process economically healthy. Cost-efficient recycling could be achieved by replacing the cobalt cathode in a Li-ion battery with another material [176]. Even though cobalt-based cathode LiCoO\(_2\) has revolutionized electronics such as cell phones and laptops, the high-cost issue, toxicity, and chemical instability at deep charge combined with LiCoO\(_2\) hinder its EV application. Thus, recycling processes of EV batteries without considering the cobalt cathode increase doubts regarding the cost efficiency of EV battery recycling [177]. During the recycling process of EV batteries containing LiFePO\(_4\) and LiMnO\(_4\) cathodes, the reduction in the temperature of these components may surpass the operating temperature during the recycling process [178]. As a result, metal oxides enter slag for smelting before converting into metallic form. The recovery of components from slag generally makes a higher impact than that of the primary production of the component, resulting in reducing the sustainability of the battery recycling process. In addition, the cost of recovering the elements by recycling batteries from EV might be relatively higher than that of the normal market price of the primarily produced elements. Electrochemical performance directly depends on particle size; larger particle sizes of LiFePO\(_4\) in a cathode require longer ion diffusion time. Therefore, high available capacity is obtained from particles
with relatively small sizes but high specific surface areas. Manithram et al. (2011) stated that LiFePO₄ with smaller and uniformly distributed particle sizes could obtain a high performance cathode [179,180]. Several studies have verified this fact [181,182]. Famous EV battery manufacturers such as A123 Systems and Advanced Battery Technologies (ABAT) have put efforts into decreasing the particle size of battery materials. Recent trends in EV battery manufacturing technology include utilization of nanomaterials in electrodes that are a few nanometers in size and decrease volume growth during the charging and discharging process and finally improve the cycling stability of EV batteries. However, many difficulties occur in recovering materials from the recycling of EV batteries due to particle size reduction which also hinders the recycling process efficiency because of the irregular melting behavior of nanoparticles. The melting point for nanoparticles differs from the melting point of bulk materials. Since particle size reduction results in a large surface-to-volume ratio, the melting point decreases with reduced particle size [183]. Therefore, the nanomaterials incorporated inside Li-ion EV batteries are predicted to be slightly melted during the reduction process. The redox (oxidation-reduction) process starts after reaching the reduction temperature and is disrupted by pre-melting, thereby, compromising any recycling process. Understanding the battery degradation process at which point capacity is insufficient for use in EVs is important to develop remanufacturing processes. Several theoretical and experimental studies based on modeling have shown that forming and developing solid electrolyte interface (SEI) layers are the major reasons behind the impedance increase at both electrodes. Studies have confirmed through characterization techniques such as Raman spectroscopy, atomic force microscopy (AFM), and X-ray diffraction (XRD), that passive SEI forms significantly and is responsible for the reduction in the capacity of Li-ion battery [92,184]. The studies have stated that electrolyte degradation on the cathode forms a blocking layer on the active material, which restricts Li diffusion through the porous electrode. Manithram explained that an important challenge with a high-voltage Li-ion battery is the reaction between cathode surface and electrolytes [180]. As the battery is cycled, a reaction occurs between the cathode and electrolyte, generating an SEI on the cathode. This impacts ionic mobility via pore plugging and adds resistive electrical routes to cathode structural components [185]. SEI, a highly resistive thick layer, reduces Li⁺ ion movements at the interface of the electrode/electrolyte and weakens the cells’ working capacity, causing a fade in capacity. To obtain ineffective SEI removal using a chemical solution, the solubility of SEI compositions need to be studied and determined, inculcating the requirements to perform experiments on SEI solubility in different solvents to determine the optimal solvent for removing SEI and a solvent that is less vulnerable to the surfaces of battery electrodes. In addition, removal of SEI might have been accomplished using a physical process. Thus, exploring physical processes for SEI removal from battery electrodes is also needed. To achieve SEI removal and porous morphology through a physical process, the process must have the following features: (i) The ability to remove both organic and inorganic chemical compounds, but be very selective and not damage the bulk materials of the cathode. (ii) The process needs to be capable of removing solid electrolyte interface with a high degree of control. (iii) The process must not induce rapid aging because of undesirable concerns such as heat load and shockwave propagation.

5.2. Suggestions for Financial Challenges

The overall value chain for a battery pack is complicated, including battery component fabrication, cell production, module production, and pack assembly. Continuous engineering development can reduce the associated costs with component fabrication and cell production. However, a substantial portion of the cost is dependent on innovation of better alternative raw materials with high energy density to reduce the number of materials needed [186]. Figure 5 represents some methods of cost reduction based on stakeholder interviews and modeling exercises [187].
Figure 5. Methods of cost reduction for BEVs.

5.2.1. Quality Control

To reduce the scrap rate, having one manufacturing technique with improved quality control (QC) would eventually result in reducing battery costs. The scrap rate in LIB production has been reported to be ~2% [96]. A charge-coupled device (CCD) and beta transmission camera inspection [186] is an example of a technology currently used in industries for detecting coating defects such as agglomerates, pinholes, and non-uniform coating. During off-the-line material testing, techniques such as Raman spectroscopy are also used to identify varieties in coating composition [188]. These approaches are sensitive enough to detect defects such as non-uniform coating and pinholes, but they may lack the resolution to detect faults such as agglomerates and metal contamination in the electrode manufacturing process, which lead to malfunctioning cells and increase production costs.

An alternative technique is laser calipers which have been utilized for measuring wet coating thickness, replacing the beta transmission method that is expensive and poses safety concerns because of radiation [189]. In-line measurement provides a feedback loop for adjusting the coating protocol to timely correct errors. This is obtained by using two sensors which emit laser light, and then capturing the laser profile with a camera. The sensor shows a corresponding value of voltage as output which is linearly related with thickness. Figure 6A shows the laser caliper system used to measure coating thickness. Achieved precision of <2% was obtained for in-line laser measurement. For effectively detecting defects in-line of a coating system consisting of a high speed electrode, infrared (IR) thermography has been used [190]. IR energy emitted from an object was detected in IR cameras as a function of temperature. This system, as shown in Figure 6B, comprised of an IR camera, focused at the electrode emerging from the hot drying oven through a series of mirrors. The camera measured the coating thermal radiation and it was analyzed. As the electrode coatings were objects made from solid materials, the heat transfer from the coating to the surface was by convection and internal conduction through pores. Reduced scrap rate and consequently lower production costs may be addressed further by studying the correlations between different types of flaws and long-term cell performance. Although the correlations appear to be minor for some defects, electrodes having these errors still can be utilized in batteries. To achieve this purpose, an extensive study has to be carried...
out regarding intentionally introduced electrode flaws of different types with controlled sizes [190].

![Image of electrode coating control]

**Figure 6.** Electrode coating control: (A) Laser caliper setup for measuring coating thickness; (B) IR thermography for measuring coating defect in line. Adapted with permission from ref. [94] (Jianlin Li et al., 2017).

### 5.2.2. Electrode Processing

While adaptation of aqueous processing has been achieved in graphite anode manufacturing, an organic solvent-based process has been used to manufacture most of the composite cathodes using polyvinylidene fluoride (PVDF) and N-methyl-2-pyrrolidone (NMP) as the binder and solvent, respectively [191]. Switching cathode fabrication from NMP-based processing to aqueous processing has received significant attention because of various advantages, for example, low raw material cost [192], low capital cost [193], less effect on the environment [194], and easier to run a battery manufacturing plant.

However, several processing problems occur while replacing NMP with water, such as bulking of electrode components in aqueous suspensions due to the heavy interaction between colloidal particles, poor wettability of aqueous suspension on aluminium foils responsible for high surface tension of water [195], leaching of metal in water suspension, cracking in electrode caused by high residual stress because of water surface tension during the drying of electrode [196], and removing leftover moisture content [197]. Fortunately, most of these issues are solvable and significant progress has been made. For instance, adding dispersants would control the agglomeration issue [198, 199], and/or optimize the mixing time, sequences, and strategies [200, 201]. Poor wettability of slurry on aluminium foils can be mitigated by improving the current collectors’ surface energy such as treating the aluminium foils with carbon layer coating or corona plasma [195].

Volatile organic compounds (VOCs) are not taken into consideration during aqueous processing; suitable amounts of water are required for dissolving binders and for keeping the slurry at the desired viscosity for electrode coating, which could be energy intensive during electrode drying. Energy curing is a solventless coating technique in which low molecular weight (MW) polymers/oligomers and/or monomers are instantaneously cured into cross-linked, high molecular polymers under electron beam (EB) or ultraviolet radiation (UV) [202]. An energy curing process has three benefits as compared with thermal oven drying: (i) space reduction, (ii) low energy consumption, and (iii) high input rate attributed for the fast-curing speed of UV/EB technology. EB curing does not need photo initiators such as in UV curing and has higher penetration depth which is also controllable from the electron beam’s accelerating voltage. Recently, it has been reported that oligomers having lower MW and used in the preparation of slurry and EB curing at high line speeds provided crosslinked polymers [203].

### 5.2.3. Shortening the Formation Period

The slowest processing steps in assembling a cell are SEI formation and electrolyte wetting, which can take between one to three weeks. This time depends on the manufacturer and cell chemistry. The processes cost about USD 23/kWh, which is the second most expensive cost following the electrode processing cost (USD 36/kWh) [192]. To maintain
production rates, large equipment space and high energy is required since all cells must be placed in environmental chambers (typically between 30 °C and 60 °C) and need to be connected to the cyclers. Thus, it is advantageous to decrease the number of steps and formation time without including the cell’s performance to estimate cost reduction. The anode side SEI formed while the electrolyte is reduced at lower potential, is generally below 0.9 V vs. Li/Li$^+$ for ethylene carbonate (EC) and 0.75 V vs. Li/Li$^+$ for propylene carbonate (PC) [204,205]. At higher voltages, additives used in the electrolytes can decompose as well as form precipitate on the anode side. Anode surface chemistry and structure affects the potential of increased reduction at a higher temperature. It is important to have a stable SEI layer on an electrode, as most common commercial electrolytes with carbonate component are not stable and have a high irreversible decompose rate (e.g., reduction reactions below 0.9 V vs. Li/Li$^+$ at anode and oxidation reactions above 4.2 V vs. Li/Li$^+$ at cathode). The SEI layer prevents irreversible reduction reactions (decompositions) by blocking (ideally blocking) electron transfers between the electrolyte and electrode while allowing Li-ion diffusion through the SEI layer. The formation of SEI, resulting in irreversible capacity loss, is dramatically reduced after the first cycles because of the preformed SEI layers which passivate electrode surfaces. After the SEI is properly formed, it reserves the electrode electronically and prevents electrolyte consumption and loss of lithium inventory. Insufficient electrical conduction, high Li-ion selectivity and permeability, stability/flexibility in volume changes of active materials, thermal stability, and stability in the electrolyte are the properties of a proper SEI formation. To this end, proper SEI formation on electrodes is a slow process, slowing the production speed and ultimately adding additional cost to LIBs. It is difficult to reduce the time required to form SEI while retaining the performance of the cell. Studies have been attempted to reduce the time of SEI formation. One of the simplest strategy has been to increase the C rate, but it can result in non-uniform and discontinuous SEI [206,207]. An alternative could be to reduce the upper cut-off voltages during formation cycles as a trade-off with high capacity fade [208].

SEI formation is affected by both C rate and surface properties of active materials. A uniform distribution of electrolytes is needed before the first charge to obtain a uniform SEI layer on an electrolyte. One commonly used effective method is to apply a vacuum during cell sealing to provide uniform and accelerated wetting. Interactions between the electrodes and electrolytes are determined by electrode surface properties that impact electrolyte wetting and reduction potentials, resulting in SEI formation [209,210]. Several investigations have been conducted on changing graphite surfaces, such as heat and acid treatment, to regulate surface chemistry. (e.g., oxygen and nitrogen) [211–213]. Interactions between nitrogen or oxygen on a graphite surface are higher with Li$^+$ in electrolytes because of their high electron density. Reducing oxygen from the graphite surface can be achieved with heat treatment in an inert environment. The graphite with the low oxygen contents experiences exfoliations due to poor SEI formation [214]. Oxygen levels on graphite surfaces can be increased by chemical treatments such as HNO$_3$ and (NH$_4$)$_2$S$_2$O$_8$, which result in higher reversible capacity [215]. Ultraviolet light treatment has also been utilized to reduce the oxygen level on graphite anodes, resulting in higher capacity retention and lower impedances.

6. Conclusions

BEVs are the future of the automotive industry. Due to high CO$_2$ emissions and the contribution to global warming, electric vehicles are becoming more popular than ICE vehicles. However, improvements are needed in EVs to compete with ICE vehicles, especially in the battery technology. This study has taken comprehensive approaches to identify the challenges, prospects, and potential solutions of batteries in BEVs. The concrete findings are summarized as follows:

- One of the major challenges of EVs is the limited driving range. Increasing the energy density of the batteries can solve the problem; however, it has the drawback of increased battery weight and cost of the vehicle. Porous cathode materials, hybrid
electrode materials, increasing cell output voltage, and laminated structure battery cells can solve this problem.

- Slow charging capacity of batteries creates a range anxiety problem among BEV consumers that can be solved with fast charging. To support fast charging with sufficient charging capacity, further developments in battery cells, electrode materials, power system, charging piles, etc. are needed. Hierarchically porous carbon anode, boron-doped graphene as an anode material, and \( \text{Li}_4\text{Ti}_5\text{O}_{12} \) (LTO) can be the most promising material to be used in the anode of lithium batteries for fast charging. The establishment and distribution of ultrapowered charging stations is important to achieve maximum EV market penetration.

- EoL of EV batteries is another concern from the environmental perspective. A well-defined EoL plan for battery cells used in BEVs is required. Remanufacturing, repurposing, and recycling are some options, but each of these approaches has several drawbacks including environmental and health impacts.

- The battery used in a BEV accounts for almost half of the vehicle’s price. There is a need to reduce the battery cost to make EVs more affordable and to compete in the market with ICE vehicles. Methods of cost reduction based on stakeholder interviews and modeling exercises should be introduced.

**Author Contributions:** Conceptualization, H.R. and M.H.; methodology, H.R. and M.H.; resources, H.R. and B.N.R.; writing—original draft and final paper preparation, H.R. and B.N.R.; writing—review and editing, M.H., M.S.I., A.S.A.-K., M.S.H. and S.A.; supervision, M.H. and M.S.I.; project administration, M.H., M.S.I., A.S.A.-K., M.S.H. and S.A. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was financially supported by the funding of advanced research projects (ARP) grant ARP2020.R29.7.

**Data Availability Statement:** Not applicable.

**Acknowledgments:** The authors would like to acknowledge the financial support from the ITIDAs ITAC collaborative funded project under the category type of advanced research projects (ARP) and grant number ARP2020.R29.7.

**Conflicts of Interest:** The authors declare no conflict of interest.

**References**


18. Gao, Y.; Pan, Z.; Sun, J.; Liu, Z.; Wang, J. High-Energy Batteries: Beyond Lithium-Ion and Their Long Road to Commercialisation. *Nano-Micro Lett.* 2022, 14, 1–49. [CrossRef]


22. El Kharbachi, A.; Zavorotynska, O.; Latroche, M.; Cuevas, F.; Yartys, V.; Fichtner, M. Exploits, advances and challenges benefitting beyond Li-ion battery technologies. *J. Alloys Compd.* 2020, 817, 153261. [CrossRef]


30. Ramoni, M.O.; Zhang, H.-C. End-of-life (EOL) issues and options for electric vehicle batteries. *Clean Technol. Environ. Policy* 2013, 15, 881–891. [CrossRef]


34. Liu, W.; Placke, T.; Chau, K. Overview of batteries and battery management for electric vehicles. *Energy Rep.* 2022, 8, 4058–4084. [CrossRef]


77. Li, N.; Chen, Z.; Ren, W.; Li, F.; Cheng, H.-M. Flexible graphene-based lithium ion batteries with ultrafast charge and discharge rates. *Proc. Natl. Acad. Sci. USA* 2012, 109, 17360–17365. [CrossRef]


82. Bylaska, B.W.; Palapati, N.K.R.; Subramanian, A.; Pomerantseva, E. The role of electronic and ionic conductivities in the rate performance of tunnel structured manganese oxides in Li-ion batteries. *APL Mater.* 2016, 4, 046108. [CrossRef]

83. Nisar, U.; Amin, R.; Essehli, R.; Shakoor, R.; Kahraman, R.; Kim, D.K.; Khaleel, M.A.; Belharouak, I. Extreme fast charging characteristics of zirconia modified LiNi0.5Mn1.5O4 cathode for lithium ion batteries. *J. Power Source* 2018, 396, 774–781. [CrossRef]

84. Wang, F.; Xiao, S.; Chang, Z.; Yang, Y.; Wu, Y. Nanoporous LiNi0.5Co1.5Mn1.5O2 as a ultra-fast charge cathode material for aqueous rechargeable lithium batteries. *Chem. Commun.* 2013, 49, 9209–9211. [CrossRef]


88. Billault, J.; Bouville, F.; Magrini, T.; Villevieille, C.; Studart, A.R. Magnetically aligned graphite electrodes for high-rate performance Li-ion batteries. *Nat. Energy* 2016, 1, 16097. [CrossRef]


92. Ramoni, M.O.; Zhang, H.-C. An entropy-based metric for product remanufacturability. *J. Remanuf.* 2012, 2, 2. [CrossRef]


102. Lee, S.H.; Huang, C.; Grant, P.S. Layer-by-layer printing of multi-layered heterostructures using Li$_4$Ti$_5$O$_{12}$ and Si for high power Li-ion storage. *Nano Energy* 2019, 61, 96–103. [CrossRef]


106. Huang, C.; Dontigny, M.; Zaghb, K.; Grant, P.S. Low-tortuosity and graded lithium ion battery cathodes by ice templating. *J. Mater. Chem. A* 2019, 7, 21421–21431. [CrossRef]


116. Choi, Y.-M.; Pyun, S.-I.; Moon, S.-I. Effects of cation mixing on the electrochemical lithium intercalation reaction into porous Li$_{1-x}$Ni$_{1/3}$Co$_{1/3}$O$_2$ electrodes. *Solid State Ion.* 1996, 89, 43–52. [CrossRef]

117. Vogler, C.; Hemmer, R.; Arnold, G.; Trépo, A.; Wohlforth-Mehrens, M. Lithium nickel oxide Li(Ni$_{0.75}$Al$_{0.17}$Co$_{0.08}$)$_2$O$_2$ as cathode material for lithium ion batteries. *Ionics* 1999, 5, 421–425. [CrossRef]

118. Noh, H.-J.; Youn, S.; Yoon, C.S.; Sun, Y.-K. Comparison of the structural and electrochemical properties of layered Li$_x$Ni$_{1/3}$Co$_{1/3}$Mn$_{1/3}$O$_2$ ($x = 1/3$, 0.5, 0.6, 0.7, 0.8 and 0.85) cathode material for lithium-ion batteries. *J. Power Source* 2013, 233, 121–130. [CrossRef]


121. Lin, F.; Markus, I.M.; Nordlund, D.; Weng, T.-C.; Asta, M.D.; Xin, H.L.; Doeck, M.M. Surface reconstruction and chemical evolution of stoichiometric layered cathode materials for lithium-ion batteries. *Nat. Commun.* 2014, 5, 3529. [CrossRef]


124. Liang, L.; Jiang, F.; Cao, Y.; Hu, G.; Du, K.; Peng, Z. One strategy to enhance electrochemical properties of Ni-based cathode materials under high cut-off voltage for Li-ion batteries. J. Power Source 2016, 328, 422–432. [CrossRef]


126. Liao, J.-Y.; Manthiram, A. Surface-modification concentration-gradient Ni-rich layered oxide cathodes for high-energy lithium-ion batteries. J. Power Source 2015, 282, 429–436. [CrossRef]


130. Liao, J.-Y.; Oh, S.-M.; Manthiram, A. Core/Double-Shell Type Gradient Ni-Rich LiNi0.76Co0.10Mn0.14O2 with High Capacity and Long Cycle Life for Lithium-Ion Batteries. ACS Appl. Mater. Interfaces 2016, 8, 24543–24549. [CrossRef]

131. Sun, Y.-K.; Myung, S.-T.; Park, B.-C.; Prakash, J.; Belharouak, I.; Amine, K. High-energy cathode material for long-life and safe lithium batteries. Nat. Mater. 2009, 8, 320–324. [CrossRef]


134. Lim, B.-B.; Myung, S.-T.; Yoon, C.S.; Sun, Y.-K. Comparative Study of Ni-Rich Layered Cathodes for Rechargeable Lithium Batteries: Li[Ni0.85Co0.11Al0.01]O2 and Li[Ni0.84Co0.06Mn0.09Al0.01]O2 with Two-Step Full Concentration Gradients. ACS Energy Lett. 2016, 1, 283–289. [CrossRef]

135. Wang, J.; Du, C.; Xu, X.; He, X.; Yin, G.; Ma, Y.; Zuo, P.; Cheng, X.; Gao, Y. Lithium Phosphorus Oxynitride Coated Concentration Gradient Li[Ni0.73Co0.12Mn0.15]O2 Cathode Material with Enhanced Electrochemical Properties. Electrochim. Acta 2016, 192, 340–345. [CrossRef]


137. Manka, S.K.; Nanda, J.; Veith, G.M.; Dudney, N.J. Electrochemical and rate performance study of high-voltage lithium-rich composition: Li2Mn0.55Ni0.15Co1.2O2. J. Power Source 2012, 199, 220–226. [CrossRef]


142. Qiao, Q.Q.; Zhang, H.Z.; Li, G.R.; Ye, S.H.; Wang, C.W.; Gao, X.P. Surface modification of Li-rich layered Li(Li0.17Ni0.25Mn0.58)O2 oxide with Li–Mn–PO4 as the cathode for lithium-ion batteries. J. Mater. Chem. A 2013, 1, 5262–5268. [CrossRef]

143. Zhang, X.; Belharouak, I.; Li, L.; Lei, Y.; Elam, J.W.; Nie, A.; Chen, X.; Vassar, R.S.; Axelson, R.L. Structural and Electrochemical Study of Al2O3 and TiO2 Coated Li2Ni0.13Ni0.54Co0.13O2 Cathode Material Using ALD. Adv. Energy Mater. 2013, 3, 1299–1307. [CrossRef]

144. Mohanty, D.; Kalnaua, S.; Meisner, R.A.; Rhodes, K.J.; Li, J.; Payzant, E.A.; Wood, D.L.; Daniel, C. Structural transformation in a Li2Co1.2Ni0.55Mn0.15O2 lithium-ion battery cathode during high-voltage hold. RSC Adv. 2013, 3, 7479–7485. [CrossRef]

145. Ito, A.; Li, D.; Sato, Y.; Arai, M.; Watanabe, M.; Hatano, M.; Horie, H.; Ohswawa, Y. Cyclic deterioration and its improvement for Li-rich layered cathode material Li[Ni0.17Li0.2Co0.67Mn0.16]O2. J. Power Source 2010, 195, 567–573. [CrossRef]

146. Maruta, S.K.; Nanda, J.; Kim, Y.; Unocic, R.R.; Pannala, S.; Dudney, N.J. Solid electrolyte coated high voltage layered-layered lithium-rich composite cathode: Li2Mn0.55Ni0.17Co0.13O2. J. Mater. Chem. A 2013, 1, 5587–5595. [CrossRef]
196. Li, J.; Armstrong, B.L.; Daniel, C.; Kiggans, J.; Wood, D.L. Optimization of multicomponent aqueous suspensions of lithium iron phosphate (LiFePO4) nanoparticles and carbon black for lithium-ion battery cathodes. J. Colloid Interface Sci. 2013, 405, 118–124. [CrossRef] [PubMed]
200. Xu, K. Electrolytes and Interphases in Li-Ion Batteries and Beyond. Chem. Rev. 2014, 114, 11503–11618. [CrossRef]
201. Bhattacharya, S.; Alpas, A.T. Micromechanisms of solid electrolyte interphase formation on electrochemically cycled graphite electrodes in lithium-ion cells. Carbon 2012, 50, 5359–5371. [CrossRef]


