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Lithium-Ion Battery Technologies for Electric Mobility – State-of-the-Art Scenario

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ABSTRACT

Rechargeable batteries are an integral part of all types of electric vehicles (EVs). Batteries must contain higher energy-power densities and longer cycle life for an EV system. Lead-acid batteries, Nickel-metal hydride batteries, and Lithium-ion batteries (LIBs) have been employed as charge storage in EV systems to date. Lead-acid batteries and Nickel-metal hydride batteries were deployed in EVs by General Motors in 1996. However, the low specific energy in Lead-acid batteries (34 Whkg^{-1}) and high self-discharge (12.5% per day at r.t.) in Nickel-metal hydride batteries have marked these batteries obsolete in EV applications. LIB~~s~~ currently occupy most of the EV market because of their high specific power (~130-220 Whkg^{-1}) and a low self-discharge rate (~5% per month). The current technological maturity and mass production in LIBs have reduced the overall battery cost by ~98% in the last three decades, reaching an average value of \$140 kWh^{-1} in 2021. Although a game-changer in battery technologies, LIBs encounter various challenges: high cost, low safety, less reliability, and immature infrastructure despite environmental benignness. Overcharging and overheating of LIBs can cause thermal runway leading to fire hazards or explosion. Declining Li-resources also raise concerns regarding the reliability and shelf-life of LIB technology. Hence, a critical assessment of Li-ion chemistries is essential to comprehend the potential of LIBs in electric mobilities and to realize the prospects in EVs.

KEYWORDS: Li-ion Battery Technology; Electric Vehicles; Energy density; Well-to-Wheel; Battery Chemistry; High Voltage Cathodes; Safety

Introduction to Electric Mobility – Battery on Wheels

Electric Mobility in the 20th Century

Transportation and vehicular mobility have been significant aspects of modern civilization, ensuring connectivity and facilitating socio-economical development. Today, most automobiles on the road run with conventional fossil fuels like gasoline and diesel. In an Internal Combustion Engine (ICE), incomplete combustion results in greenhouse gas (GHG) emissions contributing to 25% of the total GHG emissions worldwide. As the global consensus grew, green and sustainable alternatives emerged towards the electrification of transportation sectors worldwide. However, electric vehicles (EVs) are historically older technologies as compared to ICE-based automobiles [1]. Battery-powered car first appeared in the early 1800s, followed by electrified locomotives in the 1830s [2]. Patents were granted in England and America in 1840 and 1847 to use electrified rails [3]. Almost a decade later, Gaston Plante invented rechargeable lead-acid batteries in 1859 [4], making battery-operated vehicles viable. However, the limitations of a naïve technology, the people's obsession for "powerful machines," and the lack of an appreciable range of EV vehicles with low energy density rechargeable batteries made the technology obsolete [5].

Due to the limited energy/power storage and low driving range, battery-operated vehicles had never gained popularity in the 19th century and were replaced by gasoline-fueled engines in the early 1900s. ICE as the power generator dominated the automobile market till the 1970s. The petroleum embargo in the 1970s resulted in a rapid increase in fuel cost, [6], [7] forcing the world into more sustainable and green solutions for mobility. The global consensus about the depletion of fossil fuel resources and global warming (0.4°C from 1970 to 1984) due to GHG emissions has fueled the revival of electric automobiles. In 1996, rechargeable Lead-acid batteries (PbA) and Nickel-metal hydride (NiMH) batteries were deployed in EVs by General Motors. [8], [9], [10] Compared to ICE-based vehicles, the low-range EVs failed to spark consumer confidence, and the battery-powered EVs remained more as future prototype vehicles.

The development of rechargeable Li-ion batteries in the 1990s (LIBs) has led to a battery revolution in consumer electronics. Since then, the evolution of LIBs as sustainable solutions as EV batteries has reduced the usage of existing rechargeable PbA and NiMH batteries

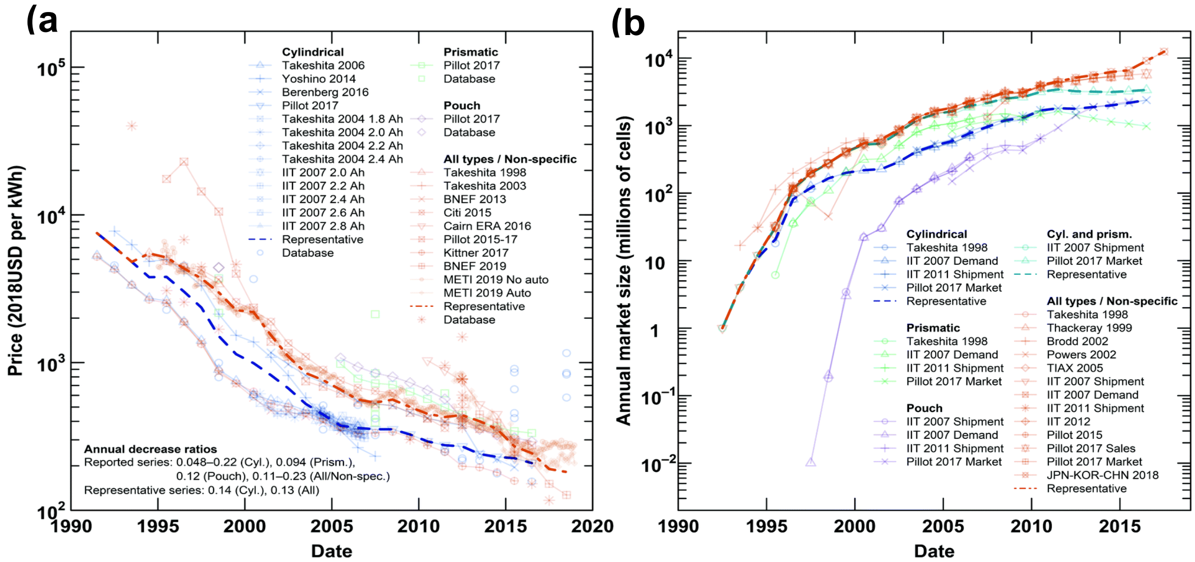

Fig. 1. The progress trends of LIB in terms of (a) price ($/kWh), and (b) Market growth volume (millions), since 1990 [Reproduced with permission from ref. 11. Copyright 2021, Royal Society of Chemistry].

due to their higher energy density (~130-220 Whkg^{-1}) and shallow self-discharge rate (~5% per month). LIBs provide the best choice in terms of energy and power densities, flexibility, and compact cell designs. Figure 1 illustrates the progress trends of LIB in terms of cost ($) per kWh and its market growth since 1990. R&D and mass production of LIBs has reduced the total cell cost by ~98% in the last three decades, reaching an average value of $140 (kWh)$^{-1}$ in 2021[11]. However, LIBs encounter a few challenges due to the limited abundance of lithium sources and the inadequate infrastructure for widespread EV applications. Cell safety and less service reliability of LIBs impede the growth of EVs. New-edge LIB technologies involving cheaper, safer, and sustainable materials have gathered enough attention in the EV market to eliminate these barriers.

This review provides an overview of EVs and the different battery technologies used in electric vehicles, focusing on the state-of-the-art LIB technology for EVs. The current global e-mobility scenario with OEM in battery technologies and automobile manufacturers for Hybrid (HEV), Plug-in Hybrid (PHEV), and All-Electric Vehicles (AEV) is discussed, along with an assessment of materials used as the anode, cathode, and electrolyte for commercial LIBs. The current status of LIBs for e-mobility from an Indian perspective is also presented.

Classifications of Electric Vehicles

Electric vehicles can be broadly classified into hybrid electric vehicles (HEV) and all-electric vehicles (AEV). A hybrid vehicle sub-class is a plug-in hybrid electric vehicle (PHEV) recharged using an external power source. A classification table of various technologies is shown in **Table 1**. The low carbon emission has caused a paradigm shift in consumer vehicles. The sustainable solutions nature of EVs has resulted in a surge in the EVs market share, reflecting the gain in consumer confidence, quality, and performance. However, the cost-effectiveness of EVs is still a significant challenge compared to conventional ICE vehicles. Figure 2(a) shows the progress of electric vehicles (in millions) among the major countries like China, USA, and Europe since 2010. The light passenger vehicles have drawn significant attention, and the demands have increased over 10 times since 2012 (Figure 2(b)).

TABLE 1:

Comparing EV technologies with ICE vehicles.

Type of vehicle	Fuel	Advantages	Disadvantages
ICE-based vehicle	Gasoline, Diesel, CNG	Proven technology and established infrastructure, Consumer confidence	High emissions, High maintenance
HEV	Gasoline, Diesel, CNG based fuel and Electricity	Lower emissions, Hybrid Technology and utilization as per need	Emissions, No direct charging from the grid
PHEV	Gasoline, Diesel, CNG based fuel and Electricity	Lower emissions, Hybrid Technology and utilization as per consumer need, Direct grid charging	Emissions, Lack of charging infrastructure
AEV	Electricity	No fossil fuel, Zero emissions, Sustainable with electricity from renewable energy, Low maintenance	High electricity demand, Non-renewable electricity generation grid

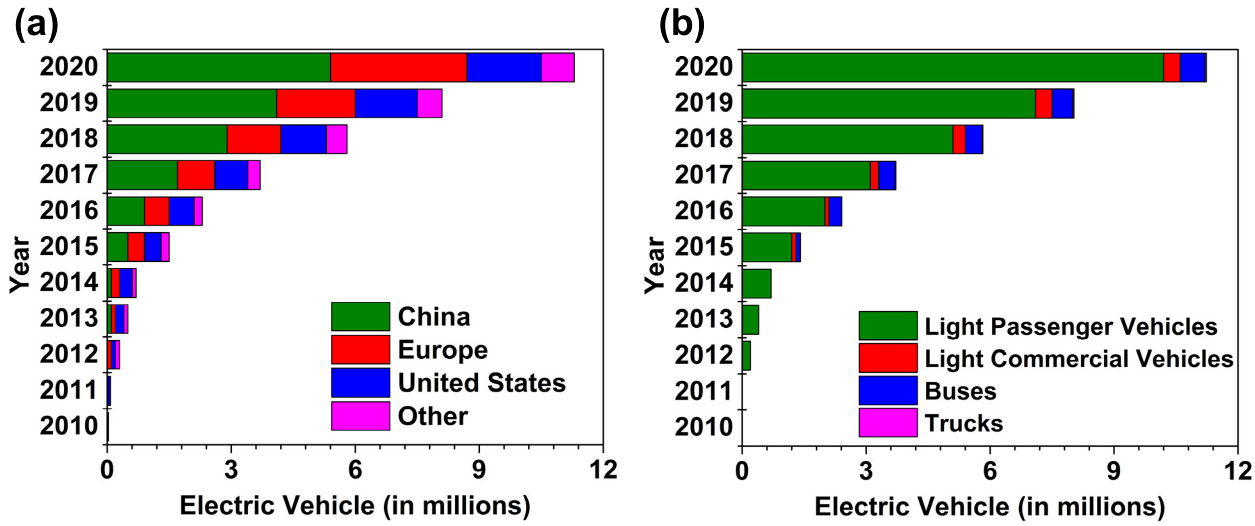

Fig. 2. (**a**) Progress in EV global market share since 2010, (b) The demand of different types of EV (in millions) since 2010 [12].

Hybrid Electric Vehicles (HEVs)

Hybrid electric vehicles (HEVs) combine traditional internal combustion engines (ICE), and electric motors as an alternate fuel for better customizable vehicle performance. Although the target vehicles for HEVs are battery-operated electric cars, various other vehicles, including pickup trucks, tractors, buses, are also beneficiaries in the EV market. A full-hybrid electric vehicle can run with just the ICE, the electric motor, or both. The regenerative braking in modern HEVs convert kinetic energy to electrical energy, and an in-built energy storage device (battery/ supercapacitor) stores the power for later use as employed in Toyota Prius [13]. After the Toyota Prius market's inception in 1997, a surge in full-hybrid electric vehicles has been evident in the automotive market. Over 17 million HEVs are currently operational worldwide. The notable examples of HEVs are Toyota Prius by Toyota, Silverado Hybrid pickup truck by Chevrolet, Saturn Aura Greenline and Malibu Hybrid by General Motors, Ford Fusion Hybrid by Ford, and Honda Accord Hybrid by Honda [14]. Figure 3(a) shows the global market share of HEVs by different automotive manufacturers till 2020. However, the simultaneous hydrocarbon fuel consumption in HEVs will not be environmentally sustainable.

Plug-in Hybrid Electric Vehicles (PHEVs)

Unlike classic HEVs, Plug-in hybrid electric vehicles (PHEVs) can be charged from an electrical power station when needed. PHEVs consist of an onboard charger and a charge port connected to the traction battery pack and electric motor. PHEVs are also recharged by regenerative braking, similar to conventional HEVs. PHEVs are common in commercial vehicles - passenger cars, trucks, buses, trains, and two-wheelers run with PHEV versions. As PHEVs can withdraw power directly from the electricity grid, a sustainable approach for powering the grid can be accepted by considering renewable energy generators compared to traditional HEVs.

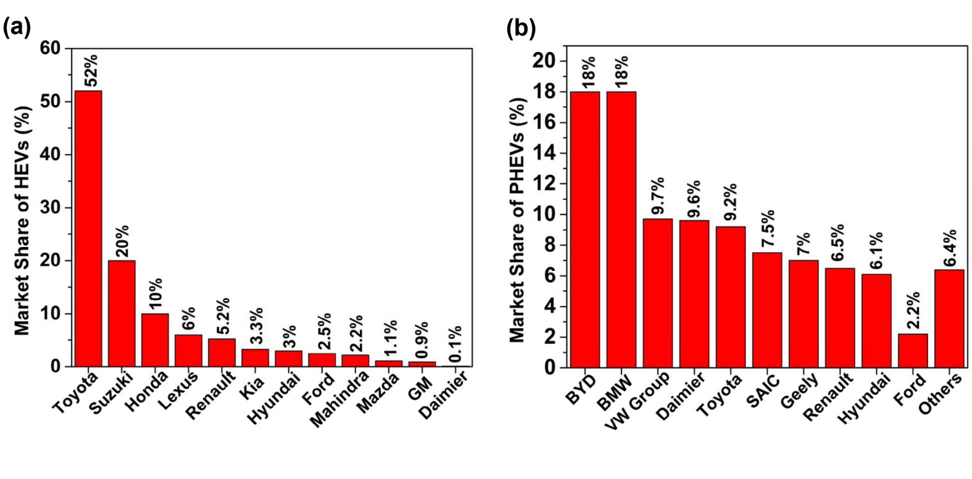

Fig. 3. Global market share (%) of (a) HEV, and (b) PHEV, till the year of 2020. [15].

China, USA, Canada, Japan, Norway, Germany, France, UK, and the Netherlands are the primary markets for PHEVs. The PHEV market models include Mitsubishi Outlander P-HEV, Chevrolet Volt, and Toyota Prius PHV. Five bestselling Cumulative sales of PHEV models have been recorded as ~8,00,000 units until 2018 [16], [17]. PHEVs can save 60% and 40% of the energy costs compared to gasoline-fueled vehicles and classic vehicles HEVs, respectively. The vehicle operating costs are minimized by plugging the battery into charging stations instead of utilizing the onboard charger's power. The onboard charger acquires power from ICE, which adds more tailpipe emissions. However, a larger battery pack to store the energy eventually increases the battery cost. The battery cost for the Chevrolet Volt model PHEV-40 (Li-ion battery pack of 8.0 kWh) has been estimated as US$14,000 in 2010, which can still save around 55% gasoline consumption compared to a classic HEV. PHEVs's global market share (till 2020) is dominated by BYD and BMW, as shown in Figure 3(b).

All-electric Vehicles (BEVs)

All-electric vehicles (AEVs) or pure electric vehicles run on electrical power generated by high-performance batteries. The stored chemical energy in the battery packs is converted into kinetic energy with the help of electric traction motors. The energy supply is controlled and monitored by the power electronics controllers. Numerous applications of AEVs are reported to date, including motorized two-wheelers, passenger-carrying four-wheelers, busses, trains, goods carriages. Based on their applications, AEVs are broadly classified into two groups; heavy and light AEVs. All-electric trains, trucks, and buses are included in heavy AEVs. While light AEVs include electric cars, motorcycles, scooters, and rickshaws. The global emerging AEV market is currently dominated by Tesla, Mitsubishi, General, Nissan, GM, BMW, and Audi, as shown in Figure 4(a). Since 2011, the market growth of AEV and PHEV has been expanding, and AEV has been popular in terms of annual sales in the last decade (Figure 4b).

Lead-acid battery-operated passenger-carrying three-wheelers currently occupy the majority of the Indian AEV market. The current Indian major market players for four-wheeler AEV's are Tata Motors Limited, Maruti Suzuki India Limited, Mahindra & Mahindra Limited, MG Motor India, Toyota Kirloskar Limited. Recently, Infraprime Logistics Technology (IPLT) launched a heavy-duty 60-tonne electric truck, claiming a mileage of 400 km (without load) and 200 km (with load) [17].

Batteries for EVs

As an integral part of the EV system, energy storage devices can consist of batteries, supercapacitors, or fuel cells. Nowadays, batteries impart one of the best performances as the state-of-art lithium-ion battery (LIB) technologies exert high power and energy densities with compact, lightweight designs. Compared to traditional Lead-acid (Pb-A) batteries and a few decades-old Nickel-metal hydride batteries (NiMH) technologies, LIBs extend much higher economic and environmental benefits. Figure 5 shows the performance parameters (energy density, single-charge range, and battery mass for EVs) for different battery technologies. A comparison shows the advantages and disadvantages of different battery technologies (Table 2). However, in either case, the expensive and high emitting gasoline-based fuel consumption is restricted to quite an extent, making the EV technology more attractive.

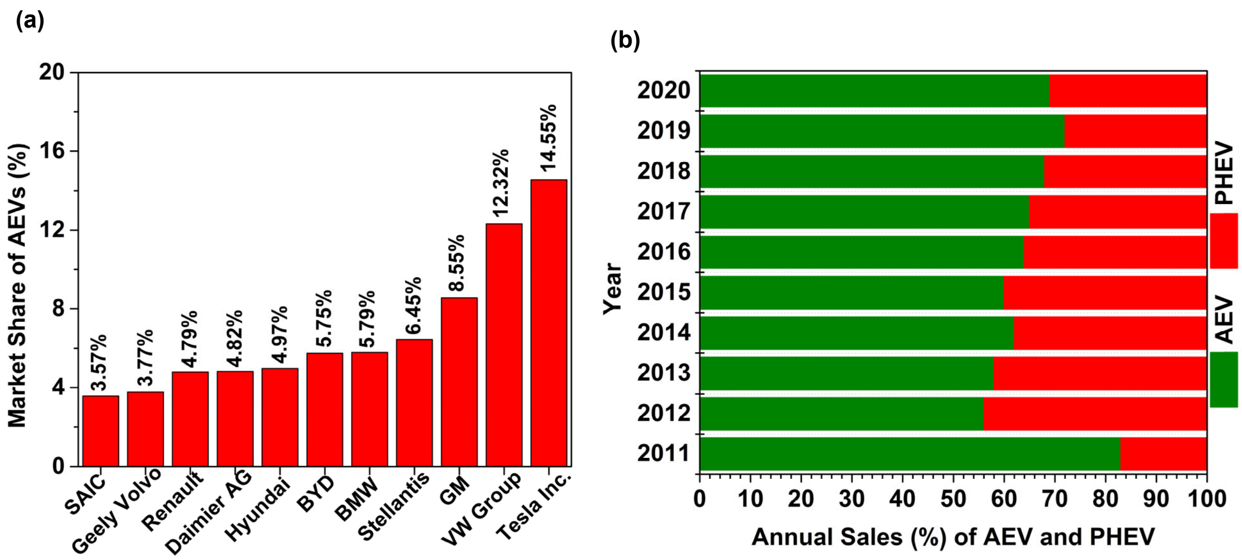

Fig. 4. (a) Global market share (%) of AEV till the year of 2020; (b) Annual sales (%) comparison of AEV and PHEV from 2011 to 2020. [15]

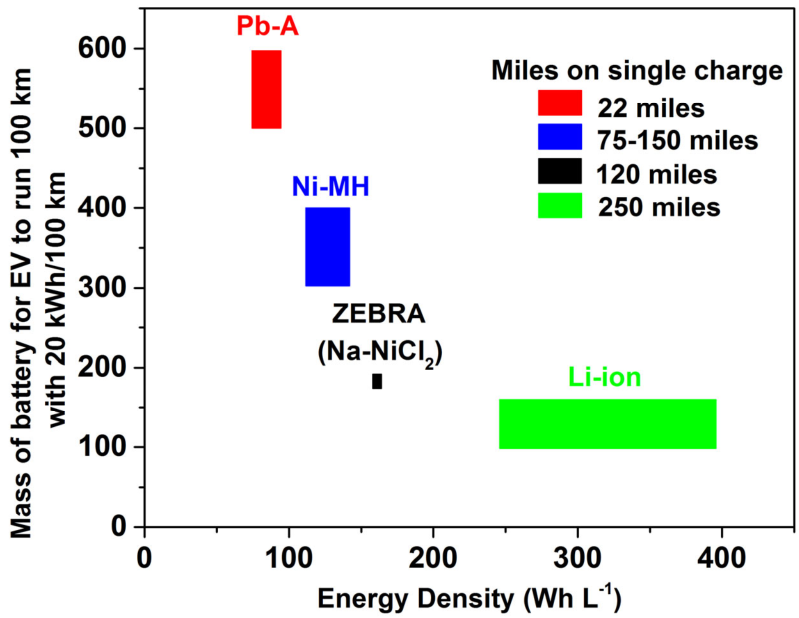

Fig. 5. Comparison of battery technologies in terms of energy density, battery weight, and range (miles) on a single charge.

TABLE 2:
Comparison of different Battery Technologies for EVs

Battery System	Nominal Cell Voltage (V)	Capacity (Wh/kg)	Advantages	Disadvantages
Pb/Acid	2	35	Low cost, Rugged and proven technology	Low specific energy, poor charge retention
Ni-MH	1.4	55	High storage capacity, good cycle life, resistance to overcharge	High initial cost, high self-discharge rate
Li-ion	up to 4.2	135	High energy and power density, matured technology, durable, flexible	High cost, Safety concerns, poor charging infrastructure

Lead-acid Batteries

Rechargeable lead-acid (Pb-A) batteries were the earliest generation of batteries employed in EV applications. The low cost has made their applications affordable in any stationary or mobile use due to the matured and rugged technology and sufficient raw materials availability. Pb-A batteries were used in most of the conventional battery electric vehicles in the 20th century. However, the low specific energy of 34 $Whkg^{-1}$ has failed to satisfy the extended driving range requirements. Also, heavy-weight Pb-A batteries occupy at least 20-25% of the total vehicle weight. Therefore, Pb-A batteries usage has been restricted into two separate areas in the current scenario. The Pb-A batteries are being used as start lighting-ignition batteries in automobile [18] to start the engine. Simultaneously, the deep-cycle Pb-A batteries are expected to store sizeable electrical energy for moving the wheels. In the Indian market, over 1.5 million three-wheelers (E-rickshaw) based on Pb-A batteries are on the road. A gasoline-powered rickshaw has a running cost of about 4/km, while the running cost of its e-rickshaw is coming around 0.5/km [19]. Working temperature conditions play a vital role in the usage of Pb-A batteries. Flooded Pb-A batteries deployed to start the engine need to be monitored regularly. The electrolyte of these batteries needs to be replaced periodically as at moderate/higher temperatures, the evolution of hydrogen, oxygen, and sulfur gases are pervasive during charge/discharge. In lower temperatures, the efficiency of deep-cycle Pb-A batteries drops quite an extent. All-electric Pb-A powered EVs were introduced to the market by GM. The 1st generation Pb-A [9] operated GM EV 1 has provided charge storage for 130-160 km mobility. Low energy storage capacity and poor service life of Pb-A batteries have indicated its restricted use in the current EV market.

Nickel-metal Hydride Batteries

The commercialization of Nickel-metal hydride (Ni-MH) batteries in EV applications started in the late 1990s. Toyota has introduced Toyota Prius EV as the world's first commercial HEV in Japan. Prius model functions with a Ni-MH pack providing a significant driving range. General Motors and Ovonic Battery's joint venture emerged as GM Ovonic L.L.C. in 1994, which has developed various generations of Ni-MH batteries for electrified GM vehicles. GM Ovonic has served impressive and extensive research on Ni-MH batteries, which pioneered the EV evolution [20]. The first-generation GMO-1 batteries were assembled using 11 Ni-MH cells with 90 Ah cell capacities in a series connection. Each Ni-MH cell could exert 70 Whkg^{-1} of specific energy and 170 WhL^{-1} of volumetric energy density. The battery pack has supplied 13.2 V voltage with 1.2 kWh capacity. In the second generation, GMO-2 batteries, the ultimate power and energy densities were kept constant while compacting the cell design. A total no of 10 individual Ni-MH cells well assembles into a battery pack. GMO-2 batteries have provided 12 V voltage with 100 Ah capacity. The overall specific energy was noted as 80 Whkg^{-1} per battery pack. The second-generation GMO-3 batteries were assembled with Zr–Ti–Ni-based metal hydride alloy as the electrode leading to a storage capacity of 380-400 mAhg^{-1}. Such improvement in cell-level energies has eventually led to obtaining a battery pack with >95 Whkg^{-1} specific energy.

GMO-3 batteries are remarkably tolerant of aggressive conditions. The 160,000 km driven GMO-3 batteries have shown high thermal stability even at 60°C and excellent retention of 80-90% of the energy efficiencies. GM EV1 e-cars launched in 1999 with Ni-MH batteries have furnished a driving range of 225 km [10]. Several other fully electric and PHEV models from automobile manufacturers like Daimler Chrysler, Ford, General Motors, and Honda were functional with Ni-MH batteries as the last century's charge storage device. However, high-performance LIBs have gradually acquired the EV market, replacing Pb-A and Ni-MH technologies.

Lithium-ion batteries (LIBs)

Lithium-ion batteries (LIBs) furnish many applications, from mobile phones to transport vehicles (Figure 6). Each application requires a different set of electrochemical performances and material components. The choice of materials in LIBs for consumer electronics may not be ideal for stationary storage or electric vehicle application. High gravimetric and volumetric energy densities are prerequisites for EV applications. The choice of a battery system's cathodes, anodes, and electrolyte materials have varying impacts on the application's targeted needs.

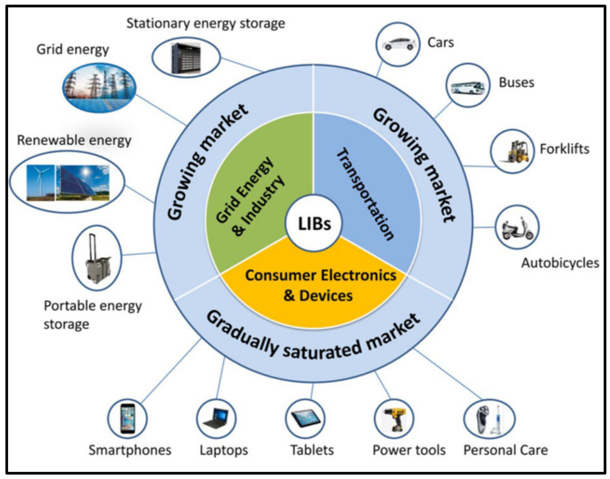

Fig. 6. Lithium-ion battery technology devices for various energy sectors [Reproduced with permission from ref. 21, Copyright @2019 Springer Nature]

LiB Technology

Electrode Materials for LIBs

Lithium cobalt oxide (LCO) is the most widely used cathode in consumer electronic devices. It falls short as a suitable cathode in EVs as it inherently suffers from structural instabilities in the overcharged state at the cost of battery lifespan. In contrast, Lithium manganese oxide (LMO), Lithium nickel cobalt aluminum oxide (NCA), Lithium nickel manganese cobalt oxide (NMC), and Lithium iron phosphate (LFP) materials are widely accepted cathodes as a replacement for LCO. Graphitic anodes are still indispensable in LIBs for EV applications on the anode side.

Cathodes

The cathode contributes to the maximum cost in manufacturing LIBs, making optimal material and cell design essential to achieve high performance, as shown in Figure 7. Batteries for EVs must be small, light, and portable. Hence, such small and lightweight batteries should contain cathodes of high gravimetric and volumetric energy densities. In reality, only a few cathodes qualify these criteria. LMO, LFP, NCA, and NMC cathodes are notable examples employed in current technologies.

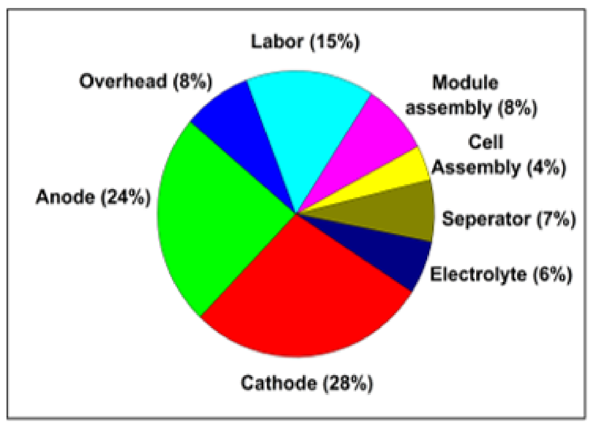

Fig. 7. The cost breakdown of various components in LIB fabrication technology [Reproduced (adapted) with permission from ref. 22 under a Creative Commons Attribution License Copyright © 2020 MDPI].

Lithium Iron Phosphate (LFP)

In 1996, JB Goodenough and co-workers first explored LFP as a potential replacement of LCO cathodes. [23] LFP cathodes provide high thermal stability, long cycle life, and significant power density. A polyanionic-based cathode can also extend the stability range at a higher charged state, unlike LCO. However, for the same reason, LFP suffers low electronic conductivity compared to oxide-based cathodes. Such low electronic conductivity reduces the fast charge capabilities, making its EV applications critical. Also, LFP cathodes exert a low average voltage (3.4 V), leading to a comparatively low gravimetric energy density of ~120 $Whkg^{-1}$ and volumetric energy density of ~220 WhL^{-1} [24]. For high-performance EV applications, smaller-size, compact, and high energy density battery packs are essential. Such low volumetric energy density in LFP cathodes qualifies them as suitable systems in larger vehicles such as buses and goods carriages. BYD e6 is an AEV that runs on prismatic cell types based on LFP cathode and provides a range of 300 driving kilometers. A123 pouch cells fabricated for Chevrolet spark e-cars employ carbon-coated LFPs as cathodes and extend the volumetric energy density up to ~250 WhL^{-1}. However, much lower battery energy (21 kWh) and range (130 km) unfortunately do not meet the current EV requirements [25].

Lithium Nickel Cobalt Aluminum Oxide (NCA)

As a cheaper, stable alternative of LCO, $LiNiO_2$ (LNO) cathodes have been identified with a high theoretical capacity of 275 $mAhg^{-1}$. Despite the advantages, LNO cathodes undergo structural deformations while charging. Similar size Ni^{2+}-ions (0.69Å) diffuses into Li-layer (ionic radii of Li^+ is 0.76 Å), causing an irreversible phase transition [26]. Substitution in the transition metal site has significantly improved the phase stability in LNO cathodes. One of the modern EV battery cathodes, NCA ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$), is synthesized via substituting Ni with 15 mol% Co and 5 mol% Al ions [27]. Tesla EV models employ NCA cathodes provided by Panasonic batteries. Al^{3+}ions substitution helps improve the structural and thermal stability, increasing the voltage of operation and subduing the structural deformations at high voltage charging [28].

On the other hand, the inclusion of Co^{3+}ions into the Ni-site can suppress Ni-migration into Li-layer and provide extra electrochemical support during the battery's charging/discharging [29]. NCA cathodes can successfully extend gravimetric and volumetric energy densities of 236 $Whkg^{-1}$ and 673 WhL^{-1}, making them one of the most potent cathodes in EV industries. Despite its toxic and expensive Co-component, NCA cathodes are typical examples in EV batteries due to their long service life of more than 15 years.

Lithium nickel manganese cobalt oxide (NMC)

NMC cathodes have emerged as economically viable solutions of LCO cathodes as expensive Co-transition metal is replaced by Ni and Mn in the transition metal layer. A variety of NMC compositions are currently used in the EV market. NMC majorly includes low or medium Ni-containing cathodes. $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC-111) is counted as a low Ni-containing cathode. Medium Ni-containing cathodes are typically $LiNi_{0.4}Mn_{0.4}Co_{0.2}O_2$ (NMC-442) and $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC-532) [30]. The notable producers of NMC cathode-based batteries are Panasonic, LG Chem, Sanyo, and Toshiba, which have successfully been employed in EVs such as Tesla 3, Chevrolet Bolt, Renault Zoe, VW e-Golf, and Honda Fit EV. NMC cathodes can furnish sufficient energy densities, good cycle stability, and high thermal stability. Tesla 3 EVs can provide a 350-500 km driving range with 75-100 kWh battery packs. Additionally, new-edge Ni-rich NMCs, NMC-622 ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$) and NMC-811 ($LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$), have shown their potential to be the future of EV technologies due to enhanced specific energy and low cost [31]. High energy composite-based NMC cathodes were initially considered an alternative solution to minimize cell manufacture costs involving expensive Co-metal. They also provided excellent specific energy of ~900 $Whkg^{-1}$ considering the cathode formula of $xLi_2MnO_3.(1-x)LiMO_2$ (M=Ni, Mn, Co) [32]. A hybrid system, composite NMC/LMO cathode has proven its worth by delivering 150-160 km driving range. Various oxide-spinel composite cathodes are efficiently utilized in EV models such as Mitsubishi i-MIEV, Ford Focus, and Nissan Leaf. The NCA cathodes can prove the best specific capacity of 200 $mAhg^{-1}$ and the highest thermal stability (even at 60°C). However, the cells suffer from economic constraints. Composites of NMCs are the most suitable cathodes for high-performance EVs. Figure 8a summarizes the different cathode materials in LIBs technology regarding specific energy, energy density, energy efficiency, and working potential.

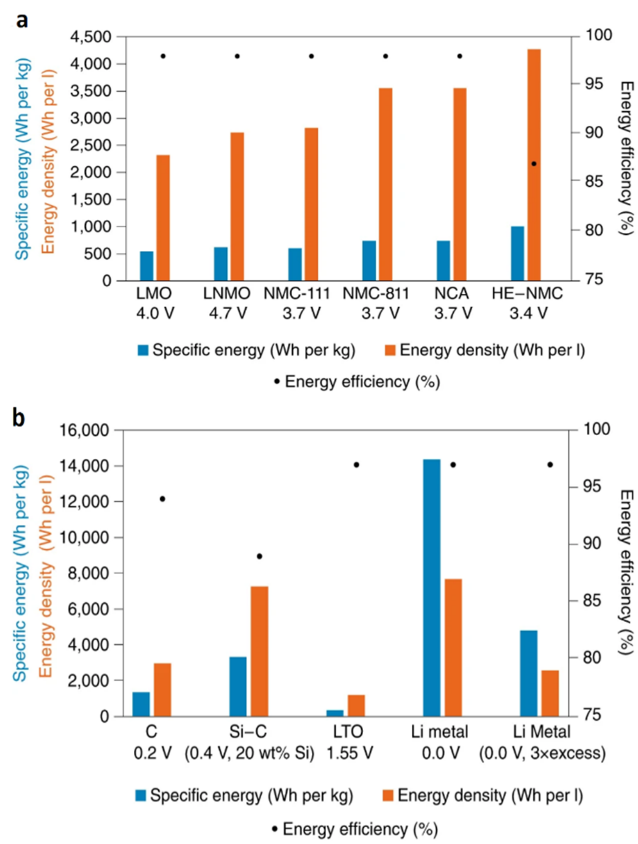

Fig. 8. Specific energies, energy densities, and average energy efficiencies of cathodes (**a**) and anodes (**b**) materials at materials level [Reproduced with permission from ref. 33, Copyright 2018 Springer Nature]

Anodes

Anode materials for LIBs are categorized based on their lithiation/ de-lithiation process. First are intercalation-type anodes materials, in which ions are inserted into the layered structures. Graphite is one of the most common intercalation-type anodes used in LIBs and forms LiC_6 intercalated compound providing a high theoretical capacity (Q_{th}) of 372 $mAhg^{-1}$ with a low volume expansion of about 9%.[34],[35] Many cell manufacturers such as AESC, LG-Chem, Li-Tech, Li Energy Japan, Samsung, and Panasonic use graphite as an anode for EV applications (Table 3).

TABLE 3:

Material Characteristics of different OEM for anode materials in EVs [36].

Anode Materials	Plateau potential vs Li/Li^+ (V)	Theoretical Capacity ($mAh\ g^{-1}$)	Advantages	Disadvantages	Battery Manufacturer/ company for EVs
Graphite	0.1	372	Abundant, cheap, long cyclic stability	SEI formation, low energy density, lithium plating	AESC LG Chem Li-Tech Samsung Panasonic
LTO	1.5	175	High coulombic efficiency, negligible volume expansion	Low capacity, low energy density higher plateau voltage	Toshiba Aptiv Altairnano
SiO_x	0.4	4200	Very high capacity for next-generation LIBs.	Low ICE Low electronic conductivity Sudden volume change	Enevate Enovix Huawei Amprius Nanotek

Since 1980, lithium titanate (LTO) has replaced graphite for Li-ion batteries. The spinel structure of LTO is regarded as a precious material due to negligible volume changes during the charge-discharge process, resulting in the electrode's durability [37]. In graphite, lithiation occurs at lower potential (~0.1V) close to metallic lithium. In comparison, LTO, due to higher lithiation potential at around 1.5 V, reduces the chance of metal plating and increases the batteries' safety [38][39]. Lithium titanium oxide (LTO) has been used in LIBs (Toshiba Cell) markets for EV application by Honda and Mitsubishi Company due to advantages of no SEI formation, high rate capability, negligible volume expansion/contraction during cycling. The serious shortcoming of LTO is low Q_{th} ($\approx$ 175 $mAh\ g^{-1}$), and high operational voltage ($\approx$ 1.55 V vs. Li/Li^+) results in inadequate energy density [40],[41]. The Toshiba cells have the lowest specific energy and energy density for Li-ion batteries due to the high voltage plateau and low specific capacity of the LTO anode. The specific energy and energy density of Li-ion batteries come in the range of 90-160 Wh kg^{-1} and 200-320 Wh $L^{-1,}$ respectively, when an individual cell is concerned. Panasonic cells using cylindrical design provide the highest specific energy of 248 $Whkg^{-1}$ and energy density of 630 WhL^{-1} [42]. The electrochemical performance characteristics of anode materials such as specific energy, energy density, working potential, and energy efficiencies are summarized in Figure 8 b.

Alloying-type anode materials (Si, Ge, Sn, Sb metals and metal oxides, sulfides, and phosphides) offer very high specific discharge capacity. However, they suffer colossal volume expansion of more than 300% resulting in poor cyclic stability limiting its practical application [43]. Low-cost silicon has a high theoretical capacity of 4200 $mAhg^{-1}$ dominating graphite and metallic lithium (Q_{th} = 3860 $mAhg^{-1}$) as an anode material. Silicon stored a more significant number of Li-ions upon charging than graphite, giving more range for the EVs on a single charge [44]. Recently, Enevate's silicon-based LIBs deliver a 400 km run on a 5 min charge.[45] Also, to high internal strain, silicon displays low Li^+-ionic mobility and high resistance in the electrical circuit.

The conversion reaction-based anodes such as Fe_2O_3, Co_2O_3, and CuO (transition metal compounds) deliver a higher specific capacity than graphite [46]. Anions (oxides and sulfides) have shown great potential as a high theoretical capacity ranging from 500 to 1500 $Ahkg^{-1}$ is evident [47]. The major drawback of these anodes is the volume expansion during cycling from conversion reactions between products and reactants. It leads to ~~a~~ substantial irreversible capacity loss, low rate performance, and voltage hysteresis due to poor electronic conductivity and loss of contact upon cycling [48].

The alternative anode materials have been explored towards high energy density, long-term cyclic stability, volume expansion, and improved reversible capacity for EV applications. Due to the safety concern and poor cyclic stability, intercalation-type anodes materials attract more attention in LIBs for EV applications. For safe LIBs anode for EVs, SEI's thermal stability is critical to guard lithiated graphite against being exposed to the electrolyte. The coated graphite with Al_2O_3 effectively avoids undesirable reactions to protect lithiated graphite and showed excellent capacity retention even at a higher temperature [49] [50]. The addition of electrolyt~~es~~ additives also provides thermally stable SEIs.

Electrolytes

Electrolyt~~es~~ are the ion-conducting medium sandwiched between two electrode components. The high ionic conductivity, low viscosity, and high cationic transference numbers are essential requirements for a smooth shuttling of Li^+-ions across the electrolytic medium. Four major electrolytes have been explored in LIB systems: organic, aqueous, solid-polymer, and ceramic electrolytes. Among them, organic electrolytes are the most widely investigated systems to date. Organic electrolytes present the maximum electrochemical stability window of 5 V vs. Li/Li^+ when ionic conductivity maximum is noted as ~8 $mScm^{-1}$. Also, standard organic electrolyte systems are preferred over the other electrolytes due to their high wettability to maintain good surface contact with the electrodes. Organic electrolytes are typically a combination of Li-salt ($LiClO_4$, $LiPF_6$, LiTFSI) and one or more organic carbonate-based solvents such as ethylene carbonate (EC) dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), propylene carbonate (PC) [51]. However, organic solvents are highly flammable and can cause fire hazards when overcharged or used in high-temperature conditions.

Also, the formation of a solid electrolyte interface (SEI) by decomposing electrolytes is critical in batteries' longer cycle life. A stable SEI performs as a protective layer onto the electrode surface whose micro-channels allow the Li^+-ions to traverse the electrode and prevent the electrodes from chemical dissolution. An unstable and outgrown SEI hinders the ionic diffusion leading to poor/zero surface contact. Dendritic growth on the anode surfaces is one of the significant examples of SEI failure [52]. One of the pioneers in the EV market, Tesla, has mentioned its challenges working with classic organic electrolytes due to poor carbon-based electrode stability. Due to organic electrolytes' high flammability and low thermal stability issues, aqueous and solid electrolytes have gained enough interest as green and sustainable solutions [53-56]. Aqueous electrolytes inherently suffer from a low electrochemical stability window (1.23 V) and inefficient SEI formation. However, concentrated aqueous electrolytes in the form of a "water-in-salt" scenario can remarkably improve the electrochemical stability window > 2 V by offering a stable SEI layer onto the anode surface.

Similarly, reliable electrolyte systems can improve battery safety by eliminating organic solvents. Polyethylene oxide (PEO) based polymers and Lithium Super Ion Conductors (LISICON) are widely accepted solid electrolytes in LIB systems.

Current Status of Battery Electric Vehicles

LIBs as high energy density batteries have initially been developed for small-scale stationary storage devices, and the utilization in EV industries was targeted to cut down on conventional fossil fuel usage. Unfortunately, the low earth abundance of LIB raw materials and the high cost slowed down mobility applications during the late 1990s and early 2000s. A sharp reduction of cell cost after 2010 has made the EVs economically viable (Figure 9a). Recent developments in LIBs have significantly dropped the overall battery cost by ~85% in the last decade, reaching a \$176 kWh^{-1} in 2019. To this extent, the base price for Honda Clarity e-car has come down below \$20,000. However, the high driving range (~300 km) of Tesla X AEVs is still above \$100,000. Tesla EVs typically employ high-performance Panasonic batteries with a large-scale production GWh factory established in Nevada, USA. Since the incubation and first production in 2017, the manufacturing cost fell by 30%, and the final production capacity is to reach 150 GWh per year[57].

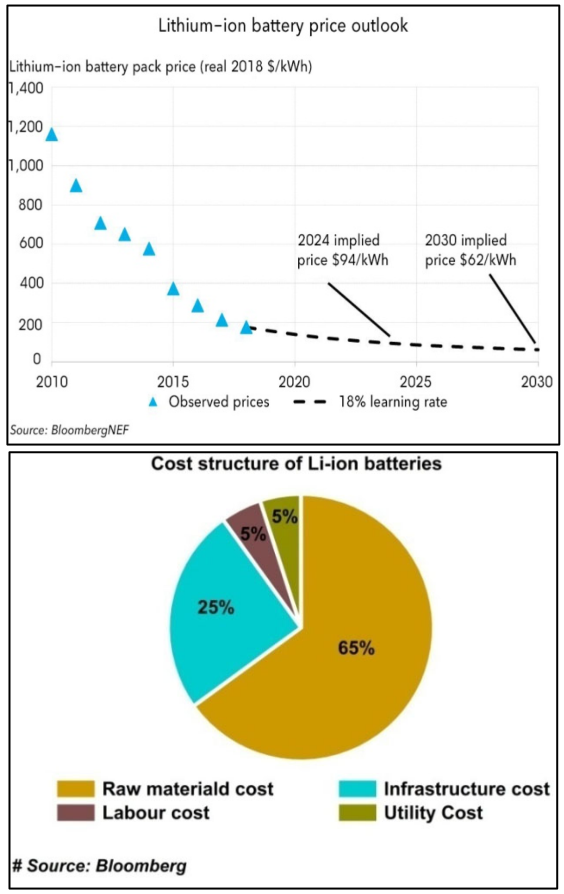

Fig. 9. (a) Actual and projected LIB pack price over the years and (b) Cost break-up of LIBs sourced from Bloomberg NEF [58].

Tesla has been the most prominent service provider in terms of driving range among AEVs. The EV models can run for ~300 km on a single charge with a battery pack energy of 60 kWh. BMW i3, Nissan Leaf, Volkswagen e-Golf, Toyota RAV4EV serve the moderate driving range 170-200 km per single charge. While Fiat 500e, Kia Soul EV, Ford Focus EV, Mercedes B-class Electric, and Mitsubishi I are the low driving range (100-160 km) EVs. Low and moderate driving range EVs cost between \$20,000 - \$50,000[59]. As expected, the high driving range Tesla models cost ~ \$100,000[60]. However, Nissan Leaf EVs provide a driving range (~230 km) with a 62 kWh battery pack and a low price below \$30,000 since 2019. The current AEV market models and their battery characteristics are listed in Table 4.

The PHEVs are considered as low-driving range vehicles for 20 to 85 km. Battery capacities of PHEVs are much lower (6-18 kWh) than AEVs as additional gasoline-based fuel provides the necessary power. Chevrolet Volt 2nd generation EVs deliver the highest electric range of 85 km along 595 km of gasoline fuel, leading to 680 km (Table 5). Although small PHEVs' overall costs are lower than AEVs, consumers' interest in AEVs has increased in the last decade.

The growth of electrification of vehicles is still catching up despite the dramatic reduction of battery pack costs. The global market share for PHEVs has increased by only 0.79% in 5 years till 2016. While at a similar period, ICE-based car sales have increased by 97%. There is still a preference for ICE-based cars among consumers over EVs due to the high cost and poor infrastructure of EV technologies. The central and federal government policies keenly promote zero-emission vehicles globally to reduce fossil-fuel dependence. Nissan has received a grant of $32.5 million from the British government to set up a plant for the manufacturing of Nissan Leaf. Toyota has sold roughly 600,000 Toyota Prius EVs only in Europe. The EV penetration percentages are 23.5% in Norway 5.1% in the Netherlands, 3.2% in Sweden, 1.3% in the UK, 1.2% in France, 0.7% in Germany, 0.9% in the USA, and 1.3% in China relative to the country's total light vehicles sold.

Table 4:

Battery packs currently used in All Electric Vehicles (AEV)[61]

Automobile Manufacturer	Model	Battery size (kWh)	Battery Chemistry	OEM	Vehicle range (km)
Tesla	S	60-100	C/NCA	Panasonic/Tesla	334-508
Tesla	X	60-100	C/NCA	Panasonic/Tesla	334-508
BMW	i3	22,33	C/NMC	Samsung/Bosch	129-183
Nissan	Leaf	24,30	C/LMO (C/NMC)	AESC and LG Chem†	135-172
Volkswagen	e-Golf	24,35.8	C/NMC	Panasonic (Sanyo)	135-200
Chevrolet	Spark	19	C/LFP	A123	132
Fiat	500c	24	C/NMC	Samsung/Bosch	140
Kia	Soul EV	27	C/NMC	SK Innovation	145
Smart	Fortwo EV	17.6	C/NMC	LG Chem	109
Ford	Focus EV	35.5	C/NMC	LG Chem	160
Mercedes	B-Class Electric	28	C/NCA, (C/NMC)	Panasonic/Tesla and SK Innovation†	137
Mitsubishi	I	16	LTO/LMO	Toshiba	100
Honda*	Fit EV	20	LTO/LMO	Toshiba	132
Toyota*	RAV4 EV	41.8	C/NCA	Panasonic/Tesla	182
*Discontinued Models.					
†In process of changing suppliers.					
Note: NCA= $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, NMC = $LiNi_{1-x-y}Mn_xCo_yO_2$, LMO = $LiMn_2O_4$, C = Graphite, LTO = $Li_4Ti_5O_{12}$.					

Table 5:

Battery packs for current market PHEVs [61]

Automobile Manufacturer	Model	Battery size (kWh)	Battery Chemistry	OEM	Vehicle range (km)
Chevrolet	Volt	18.4	C/NMC	LG Chem	85
Ford	Fusion Energi	7.6	C/NMC	Panasonic	32
Ford	C-Max Energi	7.6	C/NMC	Panasonic	32
BMW	X5	9.2	C/NMC	Samsung/Bosch	22
Hyundai	Sonata Plug-In	9.8	C/NMC	LG Chem	43
Audi	A3 Plug-In	8.8	C/NMC	Panasonic (Sanyo)	26
Volvo	XC90 Plug-In	9.2	C/NMC	LG Chem	40
BMW	i8	7.1	C/NMC	Samsung/Bosch	37
Porsche	Cayenne SE-Hybrid	10.8	C/NMC	Samsung/Bosch	22
BMW	3 Series Plug-in	7.6	C/NMC	Samsung/Bosch	22
Mercedes	S550 Plug-In	6.4	C/NMC	Panasonic (Sanyo)	32
Mercedes	GLE 550E Hybrid	8.8	C/NCA and C/NMC	Tesla and SK Innovation†	30
Porsche	Panamera SE-Hybrid	9.4	C/NMC	Samsung/Bosch	35
Cadillac	ELR	17.1	C/NMC	LG Chem	60
†In process of changing suppliers.					
Note: NCA= $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, NMC = $LiNi_{1-x-y}Mn_xCo_yO_2$, C = Graphite.					

To attract consumers towards electric mobilities, several governments have included incentivizing electric cars. This incentivization consists of both offering subsidies and investments to better charging infrastructures. A study based on the incentivization of EVs in Europe reveals that more than 80% of the new EV registration has taken place in 10 different cities/regions among Germany, United Kingdom (UK), France, the Netherlands, and Norway 2014[62]. Figure 10 indicates the relationship between the tax incentive and charging points per 1,000 registered cars among the above regions around Europe. As expected, the higher market shares of plug-in hybrid electric vehicles (PHEVs) and battery electric vehicles (BEVs) are found in cities/countries where sound incentives and charging infrastructures exist. For example, both of these criteria make Norway's EV share much higher than the rest of Europe. It is also interesting that a few individual cities/regions have a higher market share than their respective countries. For example, the EV development in Bergen, Oslo, and Utrecht is higher than in their respective countries. Indirect incentivization, including preferential treatment of EV consumers, application of EV in public transport, consumer outreach events, are the prime reasons for such a surge in market share.

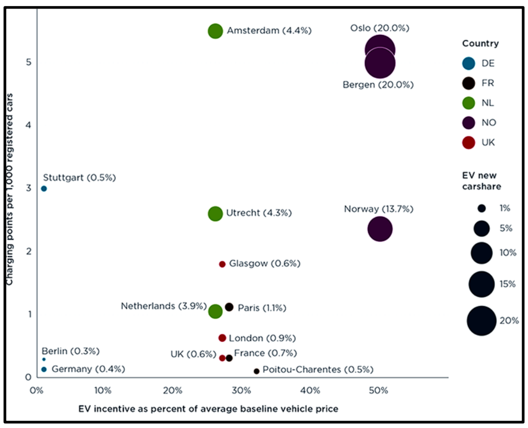

Fig. 10. The plot of fiscal incentives for EVs and charging point density for five European countries and ten cities/regions. The marker's size represented the EV share of newly registered vehicles [62].

Since 2014, a significant increase in EV market share has been observed in Germany. The recent data shows a sharp rise in EV market penetration in September 2020 in Germany. A large number of EVs sold in September (21,188 units) led to an increase of 260% compared to the last year[63]. Battery electric vehicles currently cover 8% of the total car share in Germany. Hybrid electric vehicles have observed an increase of 185%, leading to a total share of 20.4%. Although new registrations of gasoline-based cars have dropped by 24.4% (petrol and diesel engine), their market share remains higher (a total of 71.1%) compared to the EVs. A significant increase in EV share due to new government regulations and subsidies was observed during this period. Also, such growth in EV share has reduced the average carbon dioxide emissions by 13% (1,343 grams per kilometer) as compared to the last year. Besides, a record number of new EV registrations (2,04,251 units) are observed between January and September 2020 despite the economic shrinkage caused by the Covid-19 pandemic. Norway currently holds the largest EV share around the world.

The adequate incentives and easy access to charging stations have encouraged the consumers to move towards electric mobility slowly. In the first half of 2020, Norway has experienced 68% new cars[64]. The second and third positions were occupied by Iceland (49%) and Sweden (26%). Figure 11 represents a total of BEV and PHEV sales and percentage growth during the first half of 2020 (from January to June) instead of the last year. Covid-19 pandemic effects were more severe in Europe, as the new sales of EVs have increased to around 57% compared to the rest of the world.

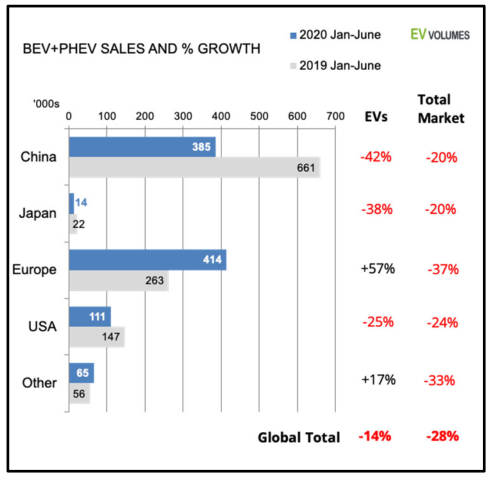

Fig. 11. The total of BEV and PHEV sales and percentage growth during the first half of 2020 (January to June) as opposed to the last year [64]

EVs – An Indian Perspective

The electrification of India's vehicles has experienced remarkable growth after the National Mission for Transformative Mobility launch in 2019. The motive behind this mission was to enforce clean, green, and sustainable vehicles on the road. The Indian EV market is currently dominated by electric three-wheelers (E3W). E3W has already penetrated smaller cities as these vehicles offer income to commercial transport agencies. BYD e-bus, electric two and four-wheelers have also contributed to the total market share[65]. Various Tier 1 city has also acquired electric buses for in-city transport. In 2014, Chinese manufacturers launched the BYD e-bus (K9D) in Bangalore[66]. The government of India's committee has sanctioned 5,645 e-buses for 65 cities in 2019 for intercity operation [67]. Olectra Greentech Ltd (Formerly Goldstone Infratech Ltd) built e-buses and delivered the first e-bus (K7) in Silvassa with a range of 200 km on a single fully charged battery [68]. Figure 12 illustrates multiple EV models currently functional in India and their respective prices in the Indian scenario [69].

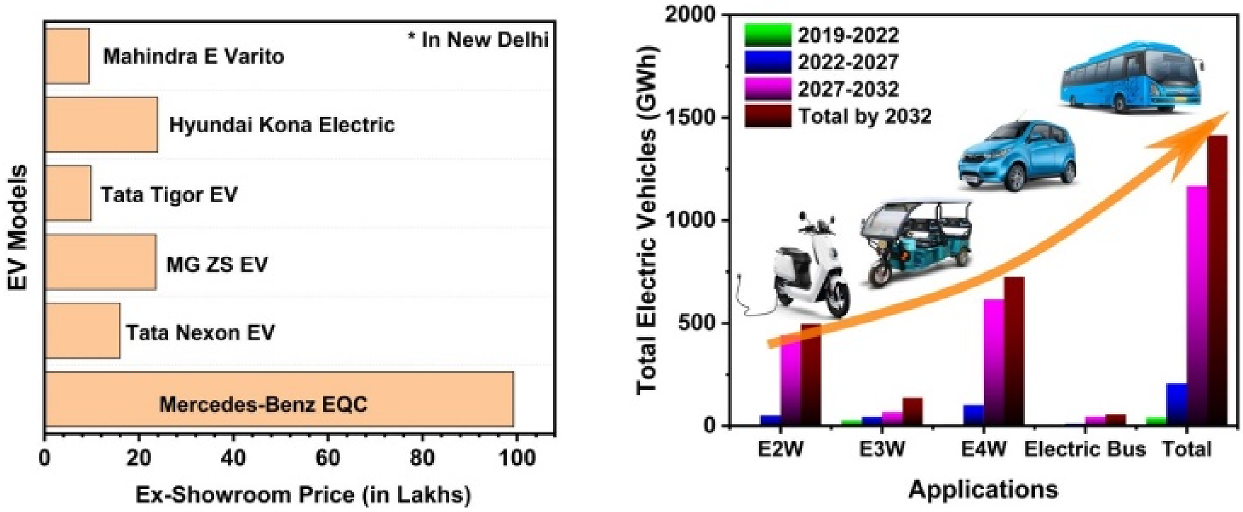

Fig. 12. Electric Cars scenario for India based on Energy Storage for Electric Mobility Applications (India Energy Storage Alliance Estimates) [70]

It is projected that E2Ws and E4Ws will cover most of the EV market shortly. The launch of large-scale LIB industries is anticipated under the Phased Manufacturing Programme (PMP) scheme for cost-effective battery productions. Such growth at the industry level will eventually encourage personal buyers to prefer EVs. In the next five years, EVs' cost is expected to surpass the price of ICE-based vehicles [70]. Figure 12 also presents the projected growth (Indian Energy Storage Alliance, IESA) of EVs across India till 2032.

Summary

LIBs presently dominate batteries in EVs as they provide high energy and power densities with appreciable driving range. However, LIBs inherently experience challenges, including high cost, low safety, and immature infrastructure. Overcharging and overheating of LIBs can cause thermal runway leading to fire hazards or explosion. Declining Li-resources also raise concerns regarding the reliability and shelf-life of LIB technology. Hence, a critical assessment of Li-ion chemistries is essential to comprehend the potential of LIBs in electric motilities and to realize the prospects in next-generation EVs. LIBs must attain a minimum cost of \$125 $(kWh)^{-1}$ and a high driving range of 500 km to achieve significant vehicles' electrification. Ni and Mn-based cathode materials like NCA, NMC, and blended LMO-NMC can considerably reduce the raw material costs. Similarly, moving towards Si-based anodes can also improve the specific energy and cut costs. Next-generation solid-state electrolyte-based LIBs enhance battery safety. Solid-state electrolyte found LIBs can also extend the specific energy range up to 900 $Whkg^{-1}$.

The innovation in the anode and cathode materials for LIBs with low cost, better battery efficiency, durability, and lightweight will be the top priorities to power EVs' demand. The next-generation anode material for LIBs could be silicon, and using these materials could improve the mileage of the EVs on a single charge. As a complementary technique to LIBs, sodium-ion batteries (SIBs) have gained momentum in the stationary storage and transport-based markets. What makes SIBs attractive is the abundant resources and comparable capacity and energy density to LIBs and lead-acid batteries. Recycling spent LIBs for EV applications can also add to sustainability and achieve good economic returns. At present, LIB technologies are almost indispensable in near-future EV applications, while various green and sustainable alternatives can lower their dependency in the far future.

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CRediT authorship contribution statement

Nagmani: Literature review, Conceptualization, Permissions, Figures, TOC and Tables, and Writing - original draft preparation, editing and review. **Debanjana Pahari**: Literature review, Permissions, Figures and Tables, and Writing - draft preparation. **Ashwani Tyagi**: Permissions, Figures and Tables, and TOC. **Sreeraj Puravankara**: Conceptualization, Supervision, Resources, Writing - review & editing, Funding Acquisition, and Project Administration.

Declaration of competing interests

The authors declare no known competing financial interests or personal relationships that could have influenced this review article, and there are no conflicts of interests to declare.

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