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The recent developments in EV batteries and REEs recovery processes from spent NiMH batteries

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Abstract

Electric vehicles (EVs) are becoming more popular than internal combustion engines for reasons such as ease of use, durability, efficiency, and speed. Electric motors aim to improve the efficiency of energy storage systems and be more environmentally friendly. Due to the growing population, the number of vehicles in use is also increasing, leading to higher carbon dioxide (CO_2) and hydrocarbon emissions. This demand has positively affected the battery market in the EV industry. Battery electric vehicles (BEVs), hybrid electric vehicles (HEVs), and plug-in hybrid electric vehicles (PHEVs), along with lead-acid batteries, nickel–metal hydride (NiMH) batteries, and lithium-ion batteries, are frequently preferred in the EV industry. NiMH batteries contain approximately 35–50% nickel (Ni), 5–20% rare earth elements (REEs), and 10% iron (Fe) by weight. Considering the supply and demand gap for REEs, often called "vitamins of modern industry", the recovery of REEs from waste NiMH batteries, which contain high amounts of these elements, becomes important. In this review article, literature on EVs, their types, and EV battery types is presented. In addition, the recycling process of NiMH batteries, which are very rich in REEs and precious metals, such as Ni, cobalt (Co), and manganese (Mn), is discussed, and the enrichment methods effective in the recycling process of NiMH batteries are examined.

Keywords: *electric vehicles*, *electric vehicle batteries*, *nickel–metal hydride batteries*, *rare earth elements*, *recycling*

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1. Introduction

The release of greenhouse gases, which cause global warming, is a critical issue that demands worldwide action. Renewable energy sources are being proposed as substitutes for fossil fuels. As a result, advancements like cleaner gasoline engines, electric vehicles (EVs), plug-in hybrids, fuel cell cars, and other environmentally friendly transportation options are viewed as future possibilities [1]. The transportation sector is one of the major contributors to the increase in greenhouse gas emissions. The automotive industry is closely monitored as one of the largest contributors to global carbon emissions into the atmosphere. Therefore, researchers have had to consider a paradigm shift due to the unstable prices of fossil fuels and environmental issues. Today, EVs are emerging as a revolutionary option for transportation in the automotive sector, and a lot of research is being done on this subject. They offer advantages such as ease of use, fewer moving parts, and less heat generation [2]. The higher efficiency of EVs relative to conventional vehicles is largely due to the fact that the conversion efficiency in electric motors specifically, from the battery to the motor, is nearly 100%. In contrast, internal combustion engines achieve an efficiency of only about 30–40%. When assessing efficiency from the standpoint of energy transmission from the battery or fuel tank to the wheels, this process is typically referred to as "tank-to-wheel" [3, 4]. In addition, EVs have higher torque and offer fast starting and stopping capabilities. With developments in renewable energy systems, the EV market is expected to become a significant market over time [2]. EVs' impact on the energy industry, policy, technical barriers, and power systems is also notable [3].

Figure 1 shows global sales of EVs between 2010 and 2022 [5].

Global sales of EVs are projected to increase significantly, from 10.5 M in 2022 to over 31 million units in 2027, representing a growth of nearly three times. This growth is projected to more than double from the 2027 level to over 74.5 M by 2035. Despite this significant increase in EV sales, the share of EVs in the global vehicle fleet will remain limited due to the existing large fleet of internal combustion engine vehicles (ICEVs) [6–8]. Given the estimated 1.33 billion light vehicles in circulation worldwide and accounting for standard scrappage rates, EVs are expected to account for only 15% of the global vehicle fleet by 2030. As the transition to EVs accelerates, this is expected to increase to around 30% by 2035 and 50% by 2042. Despite these positive growth projections, further technological development of EVs is vital for their wider adoption and increased efficiency in the global vehicle fleet [6, 8, 9].

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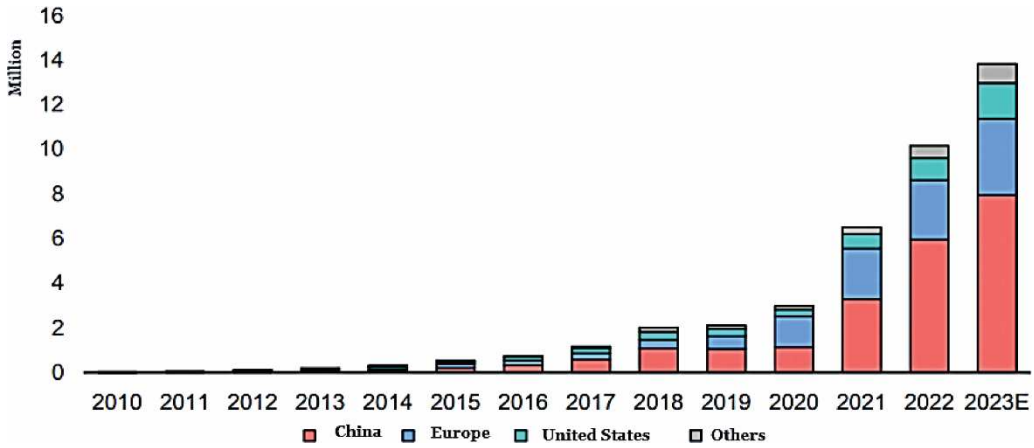

Figure 1 • Global sales of electric vehicles from 2010 to 2022 [5].

To assess the technological development of EVs, it is possible to measure their efficiency in terms of kilowatt-hours (kWh) consumed per 100 miles (161 km), which is comparable to efficiency measurements of gasoline-powered vehicles in miles per liter [6]. According to Wang et al. [10], the efficiency of EV batteries is the ratio of the amount of energy required to charge a battery to the energy obtained during discharge. The efficiency of EVs, i.e., how far they can travel with the same amount of electricity, depends on several factors. The current fleet of EVs, which have no internal combustion engine and run on small batteries, typically offers a range of 150–200 km. Theoretically, the range of EVs could be increased by doubling or tripling the battery mass. However, this would significantly increase the cost of EVs. Moreover, given the weight of the batteries, such an increase would require a more robust chassis. Nevertheless, most EVs are designed to be small and light, suitable for urban traffic, rather than large and heavy. The ability of EVs to achieve longer ranges depends to a large extent on adopting new material systems, increasing energy density, and improving production capacities through research and development (R&D) activities [6].

The upcoming generation of EVs may feature distinct vehicle architectures and weight distributions, which could influence the vehicle's overall crash performance. Central to this shift in vehicle design is the electric powertrain. The battery system, in particular, due to its substantial volume, distributed configuration, and significant mass, serves as a key factor in driving the adoption of new materials, assembly techniques, and methods for dissipating crash energy within the vehicle structure. Compared to ICEVs, battery electric vehicles (BEVs) face challenges such as higher costs and limited driving range. Both of these factors are largely influenced by the vehicle's battery system. The costs associated with the battery system are divided among various components. **Figure 2** illustrates the projected long-term evolution of battery system costs as outlined by Roland Berger [11, 12].

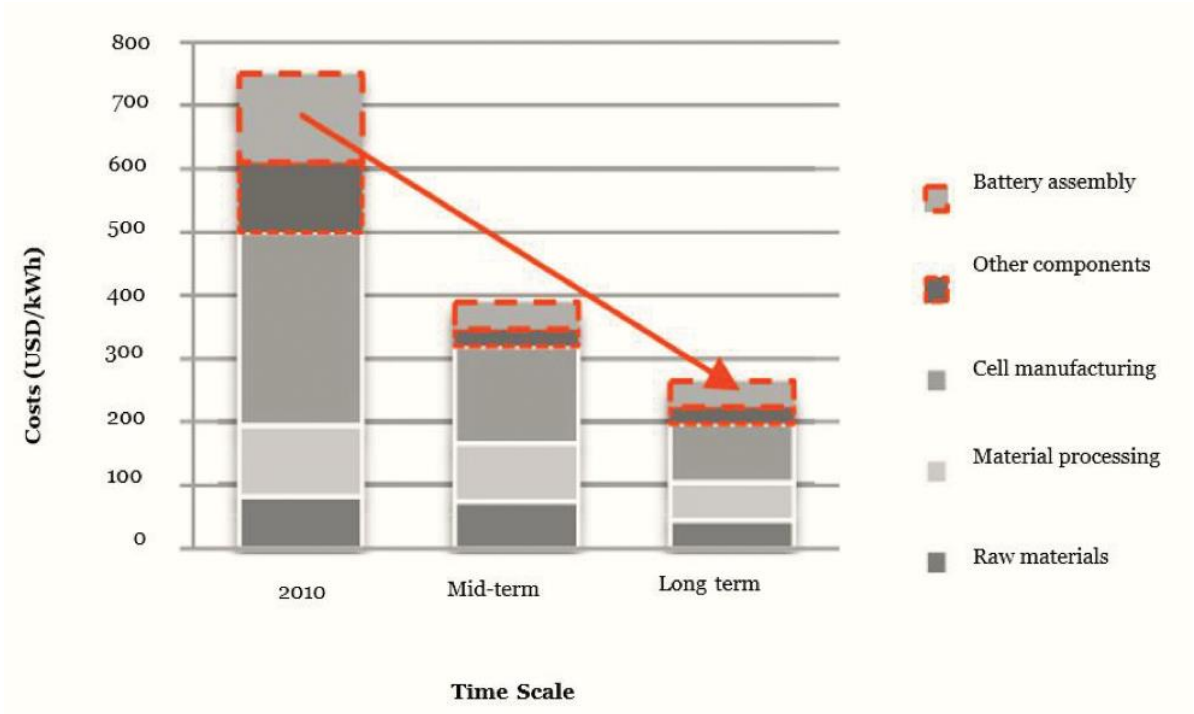

Figure 2 • Improvement of battery system costs [11, 12].

Based on this cost projection, in 2010, a 20 kWh battery system was projected to cost $15,000. Approximately two-thirds of these costs were attributed to raw materials for the cells, material processing, and cell production, while the remaining one-third

covered system assembly and the manufacturing of peripheral components. The IWF (Institute of Machine Tools and Production Technology) identified reducing assembly costs as a key focus within the proposed methodology. Lithium-ion (Li-ion) batteries have a gravimetric energy density 100 times lower than gasoline, making it necessary to design large, high-mass systems to achieve adequate energy capacity in BEV battery systems. The lightweight design of the battery system is underpinned by the consideration of the relationship between total vehicle mass and energy consumption. This relationship exhibits an almost linear trend for vehicle masses ranging from 700 to 1,700 kg [11]. For instance, decreasing the vehicle mass from 1,600 to 1,280 kg can lead to approximately 15% energy savings. This reduction has the potential to lower the required drive power and battery capacity of the vehicle [11, 13]. Consequently, the overall structural mass of both the battery and the vehicle can be minimized. This approach involves an iterative process aimed at reducing vehicle weight across four primary stages. **Figure 3** shows this relationship [11].

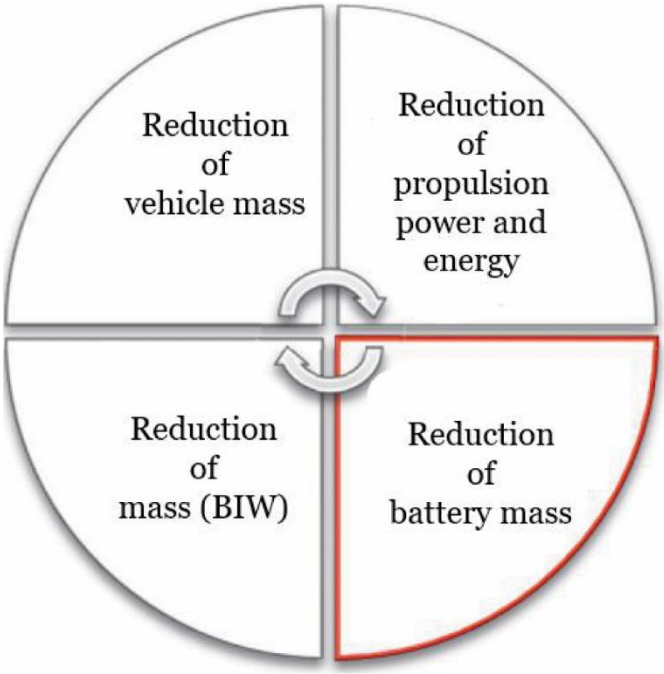

Figure 3 • Lightweight design for electric vehicles [11].

Product-level carbon emission accounting for automobiles encompasses the carbon emissions associated with all vehicle and automotive components, including both direct and indirect emissions across the entire life cycle of automotive products. This includes processes such as vehicle use, assembly, component manufacturing, raw material procurement, logistics, and recycling. For instance, prior studies have explored the life cycle carbon emissions of components like the ignition coil, fuel tank, and body steel in ICEVs. In the case of EVs, particularly BEVs, researchers have focused on analyzing the life cycle carbon emissions of power batteries [11].

The carbon footprint of EVs primarily arises from electricity consumption during vehicle operation. Assessing the carbon footprint of automotive products across their entire life cycle allows for a comprehensive comparison of the environmental impacts of vehicles utilizing various energy sources and offers a theoretical and data-driven foundation for future emission reduction strategies in the automotive industry [14].

Road transport is the predominant source of carbon emissions within the transportation sector. Data from the International Energy Agency (IEA) indicates that in 2021, global road transport emitted 5.86 Gt of carbon dioxide (CO_2), representing 76.1% of the total emissions of 7.7 Gt. Petroleum products, which serve as the primary energy source for road transport, accounted for 90% of the sector's energy consumption, whereas electricity contributed less than 1% to the energy supply [14–16].

The evaluation of vehicle carbon emissions should extend beyond exhaust emissions observed during the operational phase to encompass the environmental impacts of vehicle products from a life cycle perspective. Currently, the primary types of road vehicles include BEVs, ICEVs, and hybrid electric vehicles (HEVs) [14].

As of 2021, life cycle CO_2 emissions, encompassing production, manufacturing, material production, and maintenance processes are estimated at 22.4 tons for EVs. In contrast, the life cycle CO_2 emissions for gasoline ICEVs are 39.7 tons. Substituting gasoline ICEVs with EVs could reduce carbon emissions per vehicle by 43.4%. The electrification of vehicles plays a crucial role in diminishing life cycle carbon emissions. Historical data from 2012 to 2021 indicates that each additional million EVs could lead to a reduction of 7.28 Mt in life cycle carbon emissions and 12.8 Mt indirect carbon emissions from fuel combustion in road transport. Consequently, vehicle electrification is increasingly recognized as a necessary trend for achieving carbon neutrality targets, and EVs are anticipated to progressively replace ICEVs within the road transport sector [14, 17, 18].

However, EVs may not be as environmentally friendly as anticipated, as their life cycle carbon emissions remain relatively high, accounting for 56.4% of those from gasoline ICEVs. While EVs exhibit significantly lower carbon emissions during their operational phase compared to gasoline ICEVs, contributing to their overall reduced carbon footprint, the emissions associated with EVs during this phase are heavily influenced by the cleanliness of the electricity grid. Furthermore, EVs incur substantially higher carbon emissions than ICEVs during the material production phase. Research indicates that batteries play a crucial role in the carbon emissions of EVs. The elevated energy consumption and emissions associated with battery production result in higher life cycle carbon emissions for EVs during the material production stage compared to conventional ICEVs [14].

EVs require increased attention to carbon emissions during material production and component manufacturing, whereas ICEVs tend to focus on economic and environmental performance during operation. The lower carbon emissions of EVs during vehicle use are offset by higher carbon emissions associated with battery production in the material production phase. As the degree of vehicle electrification increases, it is anticipated that the average proportion of carbon emissions from vehicle material production will also rise. However, for EVs, the carbon emissions during material production could decrease due to the large-scale production of power batteries and related components. In the short term, the automotive industry may see an increase in carbon emissions due to the production of power batteries for EVs. Nonetheless, the benefits of reduced carbon emissions from EVs are expected to be reflected in their future operational use. As the operational phase of vehicles expands, the advantages of lower carbon emissions associated with EVs are becoming increasingly evident [14].

The transportation sector accounts for about 25% of CO_2 emissions worldwide. Motor vehicles are responsible for 75% of these emissions. Cars and buses are the largest contributors, accounting for 45.1%, while freight trucks account for 29.4%. For this reason, authorities in both developed and developing countries are encouraging consumers to replace conventional vehicles with EVs to reduce the concentration of air pollutants, such as CO_2, carbon monoxide (CO), sulfur dioxide, and other greenhouse gases [19].

As the international community aims to achieve net zero emissions by 2050 and focus on Sustainable Development Goal 13, the transition to sustainable transport solutions, such as EVs, is imperative. EVs have the potential to reduce greenhouse gas emissions and improve air quality but face barriers, such as high initial costs, inadequate charging infrastructure, range limitations, and fossil fuel-generated electricity. To achieve net zero emissions by 2050 and reduce climate change, EVs need to be adopted as a credible alternative to ICEVs [20].

In sustainable urban design, the advantages of EVs make them a preferred choice for transportation. Various alternative fuel vehicles have been proposed, such as HEVs, plug-in hybrid electric vehicles (PHEVs), BEVs, and fuel cell electric vehicles (FCEVs). EVs have attracted considerable attention in recent years, and their popularity continues to grow. Although BEVs have recently become quite common, the concept has been around for some time. The first experiments with the EV concept were conducted by Jedlik Anyos in 1828, but a rechargeable EV did not appear until 1881, after the invention of practical secondary batteries by engineer and inventor Gustave Trouvé. By 1900, EVs were gaining traction, and the number of EV manufacturers and models increased. In 1912, there were 33,842 EVs registered in the United States, their peak, but ICEVs soon took over [2].

Thanks to advances in energy storage system technology, EVs have gained an important place in a world dominated by ICEVs. EVs are available in various types, such as road and rail vehicles, surface and underwater vessels, electric airplanes, and electric spacecraft. There are two main reasons for the growing importance of EVs: the first is to reduce demand for fossil fuels such as crude oil, natural gas, and coal. Due to the high demand for fossil fuels and an annual consumption rate of 22 billion barrels, oil reserves are estimated to last only 45–100 years. The second is to reduce greenhouse gas emissions. With the increasing number of ICEVs, the amount of CO_2 emitted into the atmosphere has also increased. Greater adoption of EVs can significantly reduce CO_2 emissions. Furthermore, HEVs emit less CO_2 compared to conventional ICEVs [21].

In general, this article provides an overview of global EVs and their various types. It includes information about the battery types used in EVs and a review of studies on the utilization of waste nickel–metal hydride (NiMH) batteries, which are seen as a potential source of rare earth elements (REEs).

2. Electric vehicle types

EV types can be categorized into four classes: BEVs, PHEVs, HEVs, and FCEVs [19].

2.1. Battery electric vehicles

BEVs are defined as “fully EVs” because their powertrain consists of a rechargeable battery. Such batteries are more environmentally friendly than conventional energy conversion systems. Charging of these batteries can be done using grid power or any power generation media via a charging plug. Unlike conventional internal combustion engines, the charging process for BEVs takes slightly longer. Using a slower charger, it can take 5–10 hours to fully charge the battery, whereas with a fast charger, it takes about 15–45 minutes. Heat management and battery capacity are the main factors affecting the overall performance of a BEV. The driving range of a BEV is between 160 and 250 km, while some EV models can travel up to 500 km on a single charge. The time required to fully charge the battery depends on the battery capacity, the charging system, and the type of connection used (parallel or serial). HEVs have been proposed by various research groups to replace BEVs to increase travel distance [2, 19].

2.2. Plug-in hybrid electric vehicles

These vehicles can run on fossil fuels, electricity, or a combination of both and offer several advantages, including reduced dependence on oil, increased fuel economy, improved power efficiency, and lower greenhouse gas emissions. PHEVs can be superior to HEVs in terms of efficiency because the limited and selective use of the internal combustion engine increases overall efficiency. Furthermore, the internal combustion engine only operates at high speeds, allowing it to achieve the highest efficiency. PHEVs usually run on gasoline, but diesel or ethanol can also be used. PHEVs do not rely heavily on the internal combustion engine to charge the battery like HEVs. Instead, they have a battery pack that can be fully charged by plugging into a standard 120/240 V AC electrical outlet. They also feature regenerative braking, providing an alternative way to recharge the battery. Several studies have shown that PHEVs emit less CO_2 and other pollutants when charged from the electricity grid compared to conventional ICEVs and HEVs. PHEVs can therefore reduce emissions in the transport sector in many regions, as long as the electricity grid provides a cleaner energy source than gasoline or diesel. This means that the fuel mix used to generate electricity must be lower than the average emissions of a conventional gasoline car [22].

2.3. Hybrid electric vehicles

In HEVs, both a gasoline internal combustion engine and a battery-powered electric motor are used to power the vehicle. When the battery is low, the gasoline engine is used to start the vehicle and recharge the battery. These vehicles are less efficient compared to fully electric or plug-in hybrid vehicles. The combination of the battery and electric motor system aims to achieve better fuel economy or enhanced performance compared to a traditional ICEV. This is primarily accomplished by using the less efficient internal combustion engine alongside a much more efficient power source, such as the residual battery [19, 22].

2.4. Fuel-cell electric vehicles

FCEVs are also referred to as zero-emission vehicles. These vehicles operate on electricity, which is produced from a fuel source. Hydrogen is the primary fuel used in the fuel cells of these vehicles. Fuel cell engines are more efficient for long-distance

travel. They offer an extended range, and refueling only takes a few minutes [19].

3. Battery types used in electric vehicle

An essential component of EV systems is a device that stores energy and manages its distribution. Recently, the significance of energy storage systems has been growing. Key factors include the amount of energy stored and the efficiency of energy storage systems. Selecting the appropriate energy conversion method is also crucial. Various energy storage systems are becoming prominent, such as fuel cells, supercapacitors, primary batteries, secondary batteries, and hybrid systems. **Figure 4** illustrates the different types of batteries used in EVs [1].

A battery, an electrochemical device, converts electrochemical energy into electrical energy and is crucial for EVs. In recent years, the use of rechargeable batteries in EVs has surged in popularity due to the inconsistency of renewable energy sources, which are unsuitable for situations requiring a continuous and reliable supply. Ideal characteristics for a good EV battery include high specific energy, high charge acceptance rate, high specific power, long life cycle, low self-discharge rate, long calendar life, recyclability, and low cost. Various types of batteries are used in EVs, including lead-acid, nickel-based, and Li-ion batteries [23].

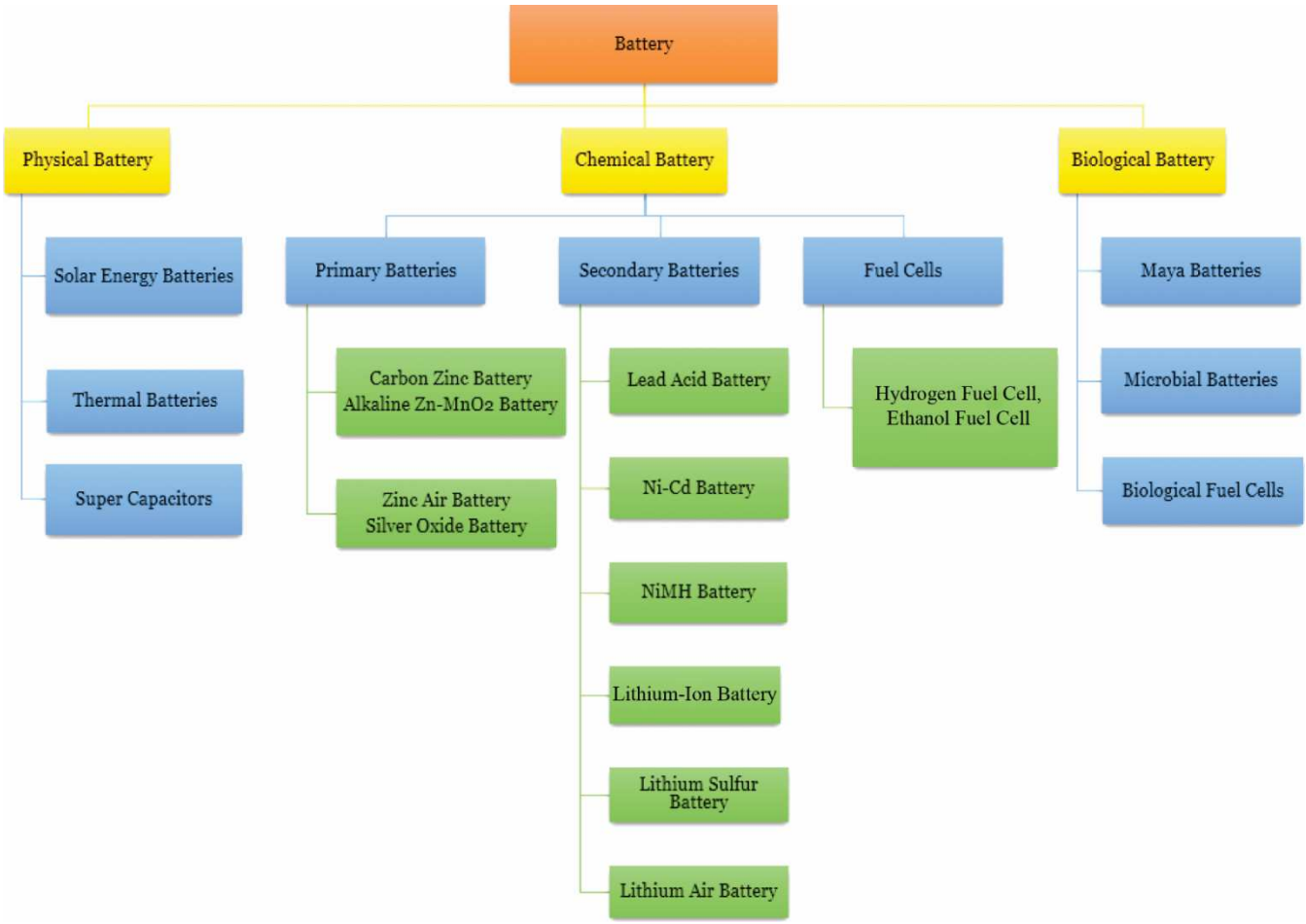

Figure 4 • Classification of electric vehicle batteries [1].

Automotive manufacturers globally are making significant efforts to develop vehicles with electric powertrains. Additionally, substantial efforts are being focused on extending vehicle range and energy density while cutting costs. Li-ion batteries have taken the place of NiMH batteries in electric and hybrid vehicles and are now also being utilized in "stop–start" vehicles [21]. **Figure 5** illustrates the economic trends for lead-acid, NiMH, and Li-ion batteries within the EV market. These three battery types are the predominant choices for EV applications, and their market share is anticipated to experience substantial growth throughout the forecast period [21, 24].

Figure 6 illustrates the average costs of lead-acid, NiMH, and Li-ion batteries from 2014 to 2022. According to the figure, the average cost of a Li-ion battery pack decreased to $181 in 2022. Advances in battery technology over the past decade have resulted in average cost reductions of 72% for lead-acid batteries, 68% for NiMH batteries, and 82% for Li-ion batteries since 2014. It is anticipated that the cost of Li-ion batteries will decline to $100 per kWh by 2024 [21, 25].

From the perspectives of manufacturers and buyers, battery price and battery cost are defined differently. As the battery market is relatively new to the automotive industry, these values are subject to variation due to numerous factors. Two primary methods are used to evaluate battery cost: the market data survey, which indicates the prices at which manufacturers sell battery packs, and the bottom-up cost model, which encompasses all production-related expenses. Manufacturers then set a price that includes a profit margin. Market analysis suggests that both the market data approach and the cost modeling method can be utilized for determining battery pricing [21].

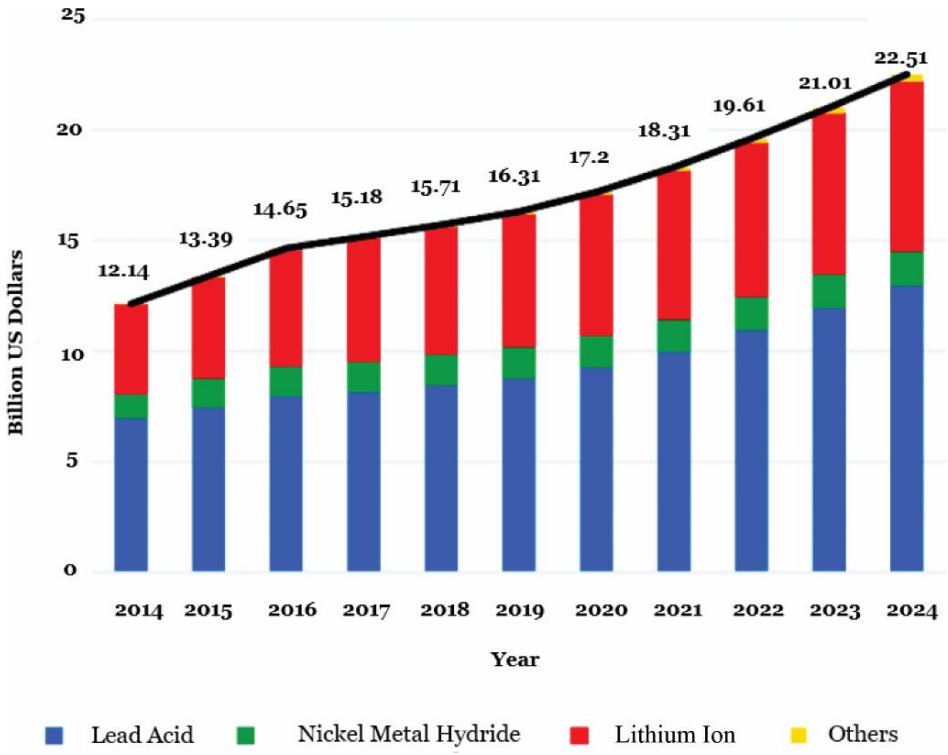

Figure 5 • The evolving trends in the use of various battery technologies in electric vehicle applications in the United States [12].

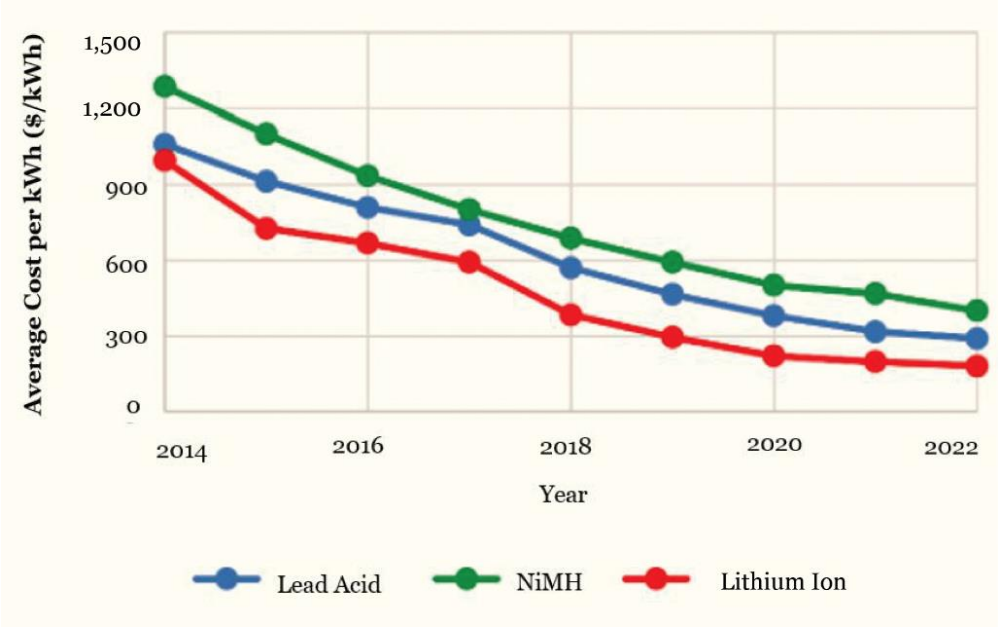

Figure 6 • Average cost per kWh of different battery types between 2014 and 2022 [21, 25].

3.1. Lithium-ion batteries

Li-ion batteries, similar to lead-acid and NiMH batteries, are well-known examples of rechargeable batteries that can be recharged whether fully or partially discharged and are used in EVs and HEVs. Since their introduction to the battery market in 1991, Li-ion batteries have gained significant importance. They are considered the most promising batteries for EVs due to their high energy density and long cycle life. Historically, a major challenge with Li-ion batteries was their limited operational temperature range, which affected performance. However, in recent years, Li-ion batteries have offered superior battery economy due to their high power and energy density ratios. They have become increasingly vital in industrial applications, providing higher voltage per cell, improved life cycles, and greater energy density. Additionally, Li-ion batteries are distinguished by their excellent low-temperature performance, low self-discharge rates, and straightforward charging methods, setting them apart from other battery types. These characteristics make Li-ion batteries the preferred choice for commercial applications. The Li-ion battery market was valued at over USD 24 billion and is projected to grow by over 12% from 2017 to 2024. Stringent government regulations on lead usage, along with the increasing demand for electronic devices, have propelled the growth of the Li-based battery market. **Figure 7** illustrates the usage trend of Li-ion batteries across various application areas in 2016 and 2024 [21, 26].

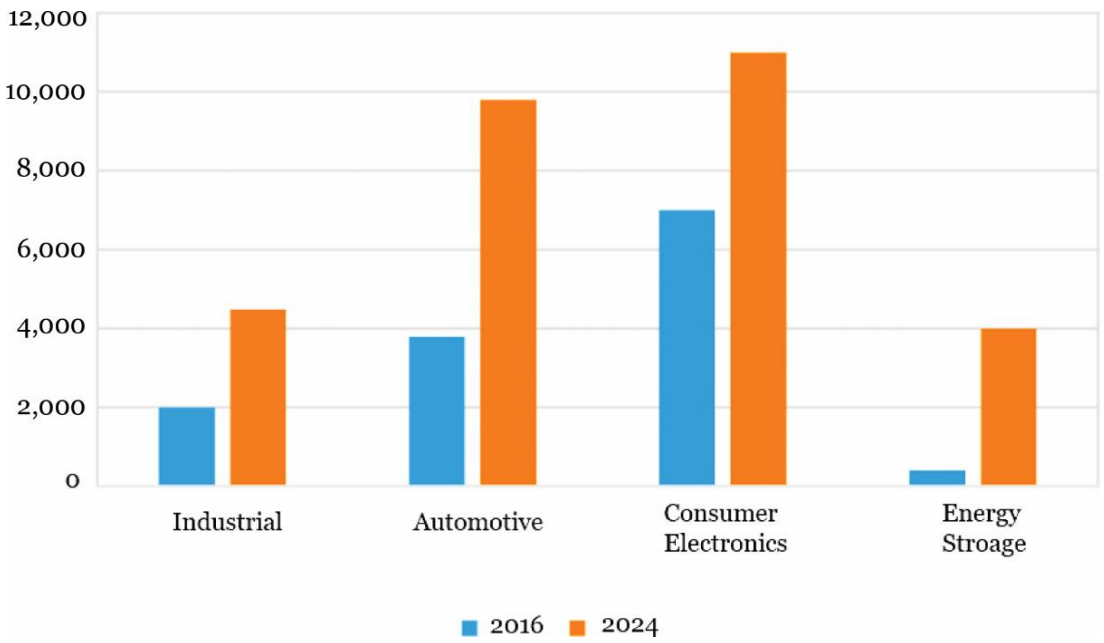

Figure 7 • The use of lithium-ion batteries in different applications in 2016 and 2024 [21, 26].

Li-ion batteries are recognized as the most efficient type of battery for various portable electronic devices due to their high energy and power density per unit weight. These batteries typically consist of a cathode, anode, organic electrolyte, and separator enclosed in a metal case and plastic cover. The cathode is usually composed of Li metal oxides such as $LiCoO_2$, $LiNiO_2$, $LiMn_2O_4$, and $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$, while the anode is typically graphite. Since the anode is usually composed of graphite or activated carbon (C), most of the precious metals in Li-ion batteries are found in the cathode [27]. Batteries are also classified according to the type of cathode material. The first commercially developed cathode materials include LCO (lithium cobalt oxide) ($LiCoO_2$), LFP (lithium iron phosphate) ($LiFePO_4$), NCA (lithium nickel cobalt aluminum oxide) ($LiNiCoAlO_2$), LMO (lithium manganese oxide) ($LiMnO_2$ or $LiMn_2O_4$), and NMC (nickel manganese cobalt oxide) ($LiNi_xMn_yCo_zO_2$, where $x + y + z = 1$). Today, NMC batteries dominate around 72% of the market and are the most widely used battery type. NCA batteries represent 26% of the market share, with LFP and LMO batteries following [28]. LCO batteries have achieved considerable success in portable electronics due to their high volumetric energy density. These batteries also power EVs such as the Tesla Roadster and Smart ForTwo. However, the high cost driven by limited cobalt (Co) availability has constrained their broader adoption in the EV market. Another cathode material utilized in EVs, particularly in electric buses in China, is LFP. The primary benefits of LFP batteries include stable voltage, high cycling efficiency, and thermal stability, though their lifespan is significantly impacted by moisture. LMO is another commercially used cathode material, valued for its thermal stability and cost-effectiveness. Nonetheless, its low energy density limits its standalone use in EVs, leading to its common combination with NMC to enhance energy density. Among the most prevalent cathode materials for EVs are ternary-layered oxides, particularly nickel-rich variants like NMC and NCA. While nickel-rich NMC offers higher capacity, it faces challenges with thermal stability and cyclic performance, which can be addressed through a full concentration gradient approach. Similarly, nickel-rich NCA provides high specific energy but requires additional safety measures due to its lower safety profile before being incorporated into EVs; NCA is notably used in vehicles, such as those produced by Tesla [2]. In general, the raw materials for Li-ion batteries are lithium carbonate and lithium hydroxide, which are used in the production of Li cathode materials. In addition, Li salt forms can also be included in the battery structure as electrolytes [29]. It is shown in **Table 1** that Co, Li, Ni, and manganese (Mn) dominate the cathode structure and require special pyrometallurgical or hydrometallurgical processes for recycling [27, 30–36].

Table 1 • The precious metals in the most common commercial lithium-ion batteries [27, 30–36]

Battery type	Metal type (elemental weight %)							
	Co	Li	Mn	Ni	Cu	Al	Fe	Ref.
$LiNi_xCo_yMn_zO_2$	14.88	5.75	16.96	21.87	0.05	10.82		[30]
$LiCoO_2$	16	2	-	-	10	3	19	[31]
$LiCoO_2$	5–20	5–7	-	5–10	-	-	-	[32]
$LiCoO_2$	16.5	2	-	-	7.1	4.1	-	[33]
$LiCoO_2$	29.49	3.14		0.02	16.48	8.02	-	[34]
$LiCoO_2$	27.5	14.5	-		24.5	14	-	[35]
$LiNiMnCoO_2$	8.45	1.28	5.86	14.84	16.6	22.72	8.79	[36]

In general, Li-ion batteries consist of positive and negative electrodes and electrolytes that conduct Li-ions. **Figure 8** shows the schematic structure of Li-ion batteries [37, 38].

The electromechanical charging and discharging process for Li-ion batteries operates as follows: during charging, the battery is connected to a charger. The positive electrode releases negatively charged electrons. To maintain charge balance at the negative electrode, an equivalent amount of positively charged ions is dissolved into the electrolyte solution. As lithium ions migrate to the positive electrode, they are absorbed by the graphite in the anode. This absorption process not only involves the incorporation of lithium ions into the graphite but also includes the

deposition of electrons to "lock" the lithium ions in place. During discharge, when a load is connected to the battery terminals, the negative electrode releases lithium ions, which move toward the electrolyte. The positive electrode then absorbs these lithium ions. Concurrently, electrons are released from the negative electrode and travel through the external circuit to the positive electrode, thereby supplying electrical current to the circuit [37].

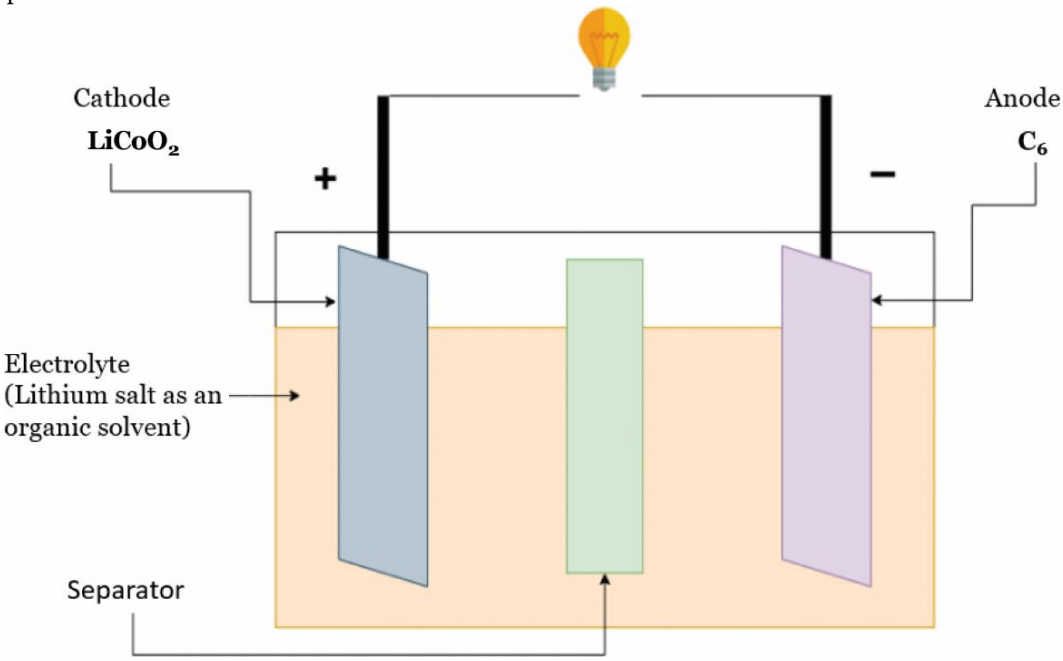

Figure 8 • Schematic structure of lithium-ion batteries [37, 38].

Figure 9 shows the size of the global Li-ion battery market and its projected growth in US$ billion from 2015 to 2024. The Li-ion battery market is expected to grow from USD 22.5 billion in 2015 to USD 58.6 billion by 2024, primarily driven by demand in light- and medium-/heavy-duty vehicles [21, 39].

Figure 10 shows the recycling process of used Li-ion batteries, consisting of three main stages. During the pretreatment stage, the batteries undergo discharging, disassembly, crushing, classification, and separation of materials, such as plastics, C, aluminum (Al), iron (Fe), and copper (Cu), from the cathode and anode components. The metal extraction stage can employ pyrometallurgical, hydrometallurgical, electrometallurgical, and bio-metallurgical methods. This stage utilizes energy, reagents, reducing agents, water, and bacteria or microorganisms and produces outputs like concentrates, slag, wastewater, and waste gases. The final stage of product preparation includes the separation and recovery of various materials and the synthesis of cathode active materials for the production of new Li-ion batteries [40].

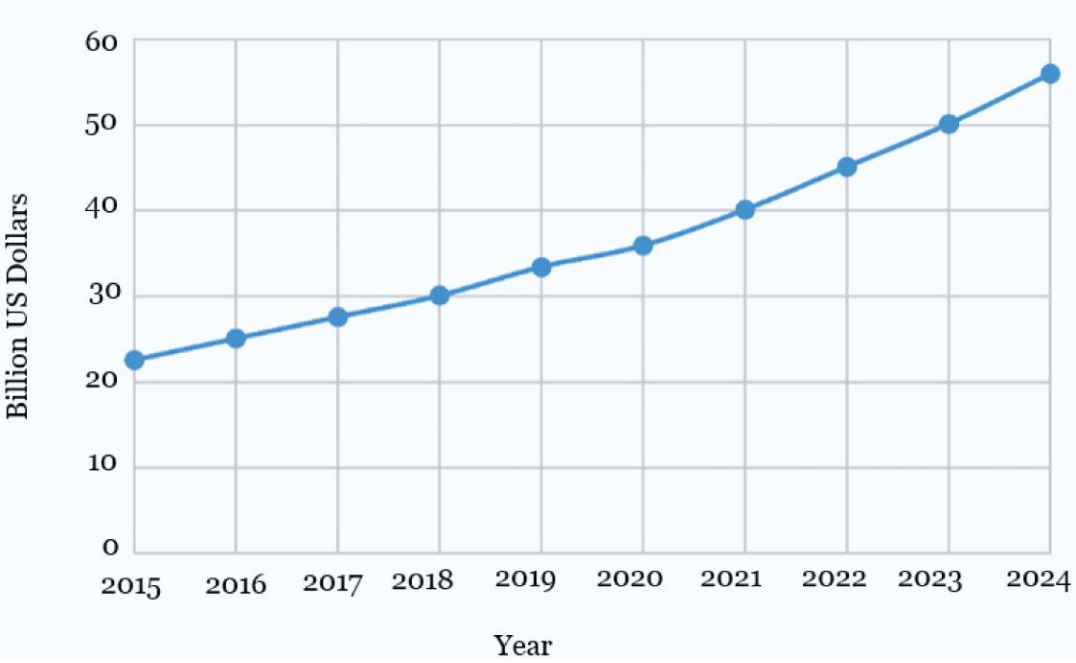

Figure 9 • The growth in the global lithium-ion battery market [21, 39].

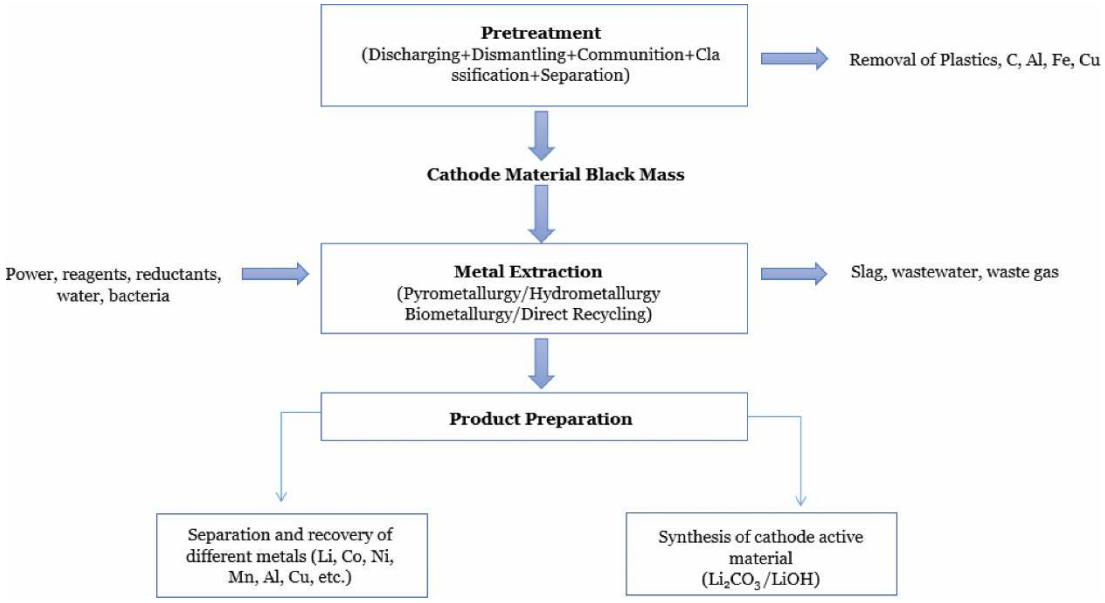

Figure 10 • Primary steps in the recycling of active materials from spent lithium-ion batteries [40].

Typically, EVs use three main types of batteries: lead-acid, NiMH, and Li-ion batteries. A comprehensive comparison summarizing the features, advantages, and disadvantages of each of these battery technologies is shown in **Table 2** [21, 41–51].

Table 2 • Main characteristics of different battery types [21, 41–51]

Battery type	Lead acid	NiMH	Lithium ion
Main components	Metallic lead, lead dioxide, lead sulfate, and sulfuric acid (H_2SO_4)	Hydrogen, nickel hydroxide ($Ni(OH)_2$), and potassium hydroxide (KOH)	Li, Fe, Al, Cu, Co, graphite, and Mn
Energy density (J/kg)	30–50	40–110	100–250
Battery voltage (V)	~2.00	1.35	2.50–5.00
Efficiency	75–80%	-	~100%
Service life (years)	2–3	> 5	5–6
Life cycle (cycles)	500–1,000	300–500	>1,000
Fast charging time (hour)	8–16 h	2–4 h	≤1 h (for manganese and phosphate based ones) and 3–4 h (Co-based)
Advanced charging ability	Good	Good	Weak
Working temperature (°C)	18–45	−30 to 70	20–65
Environmental impact	Harmful	Less impact	Less impact
Recyclability	Good	Good	Weak
Cost benefit	Low	Sealed maintenance and free design	Low maintenance cost

3.2. Nickel-based batteries

Alkaline batteries, which are Ni based, use an alkaline solution as their electrolyte. Batteries, such as nickel–zinc (Ni-Zn), nickel–iron (Ni-Fe), nickel–cadmium (Ni-Cd), and NiMH, are produced with nickel oxyhydroxide (NiOOH) as the cathode material. The Ni-Zn battery family typically has a maximum nominal cell voltage of 1.6 V and surpasses Ni-Cd batteries in specific energy and environmental friendliness due to their nontoxic nature. Additionally, Ni-Zn batteries offer excellent charge and discharge rates and a broad operating temperature range. However, their commercialization is significantly restricted by a short life cycle of approximately 300 cycles, caused by the preferential solubility of zinc species in the electrolyte. Since 1992, the EV market has extensively adopted NiMH batteries, which have a nominal cell voltage of 1.32 V and greater specific energy than lead-acid batteries [23].

3.2.1. Ni-Cd batteries

Ni, as a strategic metal, is used in a wide variety of applications, including the production of stainless steel and nonferrous alloys, chemical or petrochemical catalysts, military industries, and battery production. Recently, Ni-Cd batteries have become a popular rechargeable power source for many portable devices, such as laptops, cameras, and music players. These used Ni-Cd batteries contain valuable materials, including cathode and

anode materials made of Ni and Cd (approximately 43%), plastic separators (5%), a steel container (27%), and a Ni mesh plate that supports the anodic powder (25%). The significant value of the metallic Ni content, along with the toxic and carcinogenic nature of Cd, has led to increased interest in recycling these batteries for both academic and industrial purposes [27].

In 1908 and 1909, Edison developed Ni batteries as a portable and versatile power source. The five main types of rechargeable Ni batteries are Ni-H_2, Ni-Zn, NiMH, Ni-Cd, and Ni-Fe. All these batteries use NiOOH as the cathode material and Cd as the anode material. Ni-Cd batteries, which use nickel oxide hydroxide (NiO(OH)) and metallic cadmium as electrodes, were invented in 1899 and were widely used in portable electronic devices and EVs until the 1990s. They have since been largely replaced by newer battery technologies due to their lower energy density and the environmental concerns related to cadmium. Despite this, Ni-Cd batteries excel in high-rate capacity and longevity, lasting up to ten years under optimal temperatures of 30–80°C. However, they quickly lose capacity and have a high self-discharge rate when stored at room temperature. Their energy density ranges from 1.2 V to 50 J/kg [1]. **Figure 11** illustrates the charging principle of a Ni-Cd battery [37].

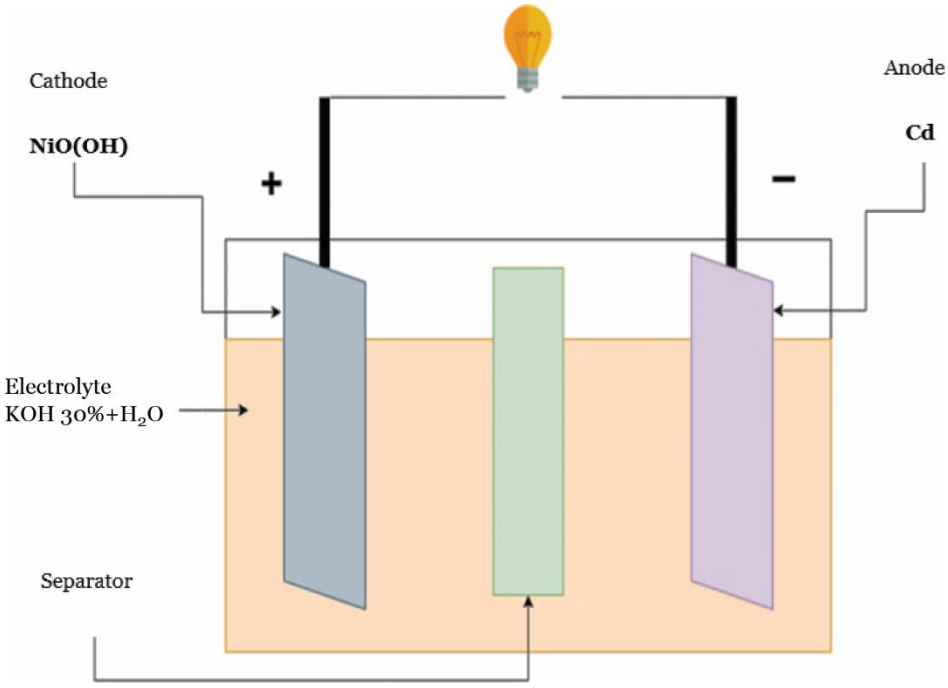

Figure 11 • Ni-Cd battery charging principle [37, 38].

The charging and discharging mechanism of Ni-Cd batteries is illustrated in Eq. (1) [41]:

$$2NiOOH + 2H_2O + Cd \Leftrightarrow 2Ni(OH)_2 + Cd(OH)_2 \; E^0 = 1.29V \quad (1)$$

The electromechanical charging and discharging process for Ni-Cd batteries operates as follows: upon connecting a load to the battery terminals, the battery begins to discharge. The KOH electrolyte dissociates into positive potassium ions (K^+) and negative hydroxyl ions (OH^-). The K^+ migrate toward the positive electrode (anode), while the OH^- move toward the negative electrode (cathode). The negative electrode releases electrons, which travel to the positive electrode through an external circuit, generating current that flows from the positive electrode to the negative electrode through the load. During charging, the OH^- migrate toward the anode, and the K^+ move toward the cathode. To discharge, the reaction is reversed: $Ni(OH)_2$ at the positive electrode is converted to NiO(OH), releasing electrons, while $Cd(OH)_2$ at the negative electrode is reduced to Cd as it receives electrons from the external circuit [37].

3.2.2. NiMH batteries

Research on NiMH batteries began in 1967. However, issues with the stability of metal hydride (MH) led to the exploration and development of nickel–hydrogen (NiH) batteries. The discovery of new hydride alloys in the 1980s significantly enhanced the stability of these batteries. NiMH batteries were subsequently developed and brought to market in Japan between 1989 and 1990, and they have become some of the most widely used rechargeable batteries today. They offer numerous advantages, including low self-discharge rates, excellent electrochemical performance, reasonable environmental compatibility, safety, and efficient operation across a broad temperature range. While Ni-Cd batteries dominated the portable rechargeable battery market until 1992, the introduction of NiMH batteries led to their replacement of over half of the Ni-Cd batteries in use. NiMH batteries avoid the issue of Cd toxicity and outperform in terms of energy density and storage effect. Currently, NiMH batteries deliver 40% higher specific energy than conventional Ni-Cd batteries. These rechargeable batteries are widely utilized as power sources for various electrical applications, including mobile devices, stationary energy storage systems, and notably, transportation systems. Despite their robust performance and longevity, NiMH batteries have a relatively lower specific energy density compared to Li-ion batteries. To bridge this energy density gap, ongoing research on NiMH batteries is being conducted in the United States, China, Japan, and Europe. Over 10 million HEVs currently in operation rely on NiMH batteries for propulsion. Consequently, given the significant contribution of NiMH batteries to the battery market, a comprehensive economic analysis of this battery type is essential. The anodes of NiMH batteries incorporate less harmful elements, such as

lanthanum (La), cerium (Ce), praseodymium (Pr), and neodymium (Nd). Typically, NiMH batteries consist of 36–42 wt% Ni, 25 wt% Fe, 4 wt% Co, and 8–10 wt% REEs, such as La, Ce, Pr, and Nd. As NiMH batteries are a major application for REEs, the global supply of these resources is experiencing considerable pressure. Currently, the supply of lanthanide group elements is severely constrained due to inefficiencies in mining processes and the scarcity of ore resources. Consequently, recycling used NiMH batteries is becoming increasingly attractive due to the substantial quantities of Ni and REEs they contain [21, 27].

MH alloys are active materials in the negative electrodes of NiMH batteries that can reversibly store hydrogen in electrochemical media. MH alloys, which exhibit metal bond strengths suitable for electrochemical applications at room temperature, can be classified as solid-solution and binary intermetallic alloys. These alloys are specifically categorized as A3B, A2B, AB, AB2, AB3, A2B7, and AB5, where "A" represents one or a combination of REEs, alkaline earth elements, and light transition metals [e.g., titanium (Ti) and zirconium (Zr)] and "B" represents transition metals, mainly Ni [52].

The NiMH battery market reached its peak between 2012 and 2013, with approximately 1.5 million HEV batteries. However, it is now gradually declining as higher-performance Li-ion batteries take over. In 2013, more than half of all HEVs utilized NiMH batteries, and a substantial number of these batteries remain on the market. Consequently, a significant quantity of used NiMH batteries is being discarded, posing a hazardous waste disposal challenge. Additionally, these used NiMH batteries contain valuable metal elements that can serve as secondary resources. For instance, each Toyota Prius HEV contains 10–15 kg of REEs, and large-scale recovery of these elements could provide significant resources for new battery production. REEs are among the most critical raw material groups in certain regions, particularly in Europe, and the recovery of Ni is becoming increasingly attractive due to its high market value [53].

As with batteries, NiMH has four main components: cathode, anode, electrolyte, and separator, as well as case (steel). The cathode of a NiMH battery consists of a Ni core coated with sodium hydroxide (NaOH), while the anode is an alloy made up of misch metal (primarily Ce, La, Pr, and Nd), Ni, Co, Mn, and Al. The anode alloy is usually represented as $MmNi_{3.5}Co_{0.8}Mn_{0.4}Al_{0.3}$, where Mm denotes misch metal. A separator made of polyamide or polypropylene is placed between the two electrodes, and a KOH solution is used as the electrolyte [54]. The chemical composition and reactions of NiMH batteries are illustrated in **Figure 12** [37, 55–57] and detailed in Eq. (2) [41]:

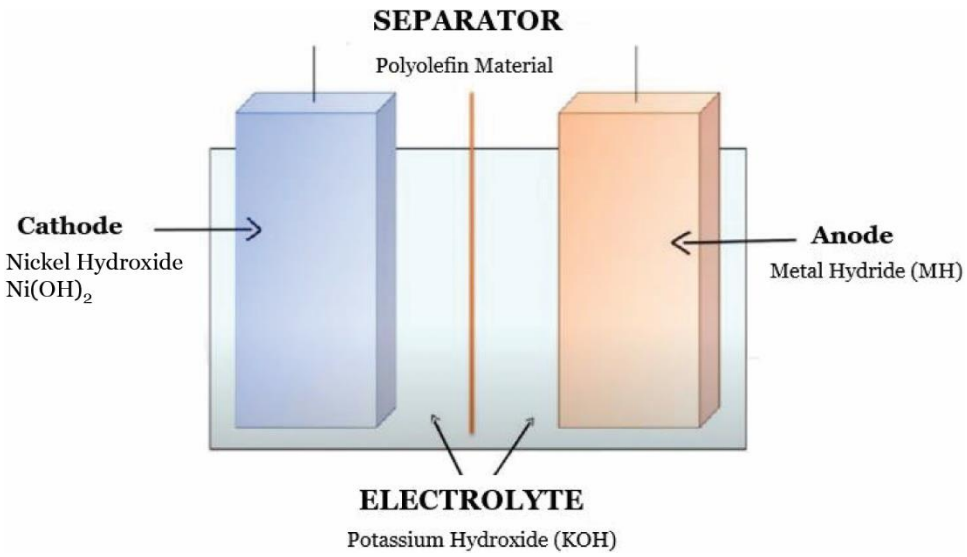

Figure 12 • The principle of operation of a NiMH battery [37, 55–57].

$$MH + NiOOH \Leftrightarrow M + Ni(OH)_2 \; E^0 = 1.35\,V \; . \qquad (2)$$

The electromechanical charging and discharging process for NiMH batteries operates as follows: during the charging phase, the cathode is composed of $Ni(OH)_2$, and the anode consists of a metal. When the charger is connected across the electrodes, a current flows through the cathode. KOH serves as the electrolyte, dissociating into positive K^+ and negative hydroxide ions (OH^-). The OH^- react with $Ni(OH)_2$ at the cathode to produce NiO(OH) and water. This reaction generates electrons that travel through the external circuit to the anode. At the anode, water reacts with the metal to form MH, which releases OH^-. During discharge, NiOOH is formed at the cathode, and MH is formed at the anode. A load is connected across the electrodes. The KOH electrolyte again splits into K^+ and OH^-. The OH^- react with the MH to produce the metal and water, releasing electrons in the process. These electrons travel through the external circuit, powering the load and returning to the cathode. At the cathode, water reacts with NiOOH to regenerate $Ni(OH)_2$ and release OH^-. The reactions that occur during discharge are the reverse of those in the charging phase [37].

As in NiMH batteries, both the anode and cathode are porous structures with a large surface area, providing low internal resistance and the ability to perform at high speeds. The cathode is usually produced by embedding or bonding Ni compounds into foam Ni or a highly porous sintered substrate, while the anode is an alloy-coated perforated Ni foil or grid that stores hydrogen. In practice, NiMH batteries can be used in telecommunications, uninterruptible power supplies, and powering HEVs due to their high energy and power (40–110 J/kg, 200–1,200 J/kg), environmental friendliness, wide operating temperature range (–30°C to

70°C), high tolerance to overcharge and over discharge, long life, low maintenance, and low cost [41].

4. A potential source of rare earth elements: end-of-life NiMH batteries

NiMH batteries are widely used in HEVs, cell phones, and laptops. However, NiMH batteries are slowly being replaced by Li-ion batteries, resulting in a higher amount of used NiMH batteries. Known for its low toxicity and high energy density, NiMH battery consists of a cathode, usually made of $Ni(OH)_2$, and an anode made of a hydrogen storage alloy based on misch metal (mostly REEs). The typical composition of NiMH is 36–42% Ni, 3–5% Co, and 5–25% REEs, such as La, Ce, and Nd. Reuse and recycling of end-of-life NiMH batteries are important issues as they lead to minimal environmental impact in terms of CO_2 emissions, resource depletion, and nonrenewable energy needs, as analyzed by life cycle assessment. Meanwhile, spent NiMH batteries are recognized as one of the most promising secondary sources of REEs for sustainable use [58].

REEs comprise 17 chemical elements in the periodic table, as defined by the International Union of Pure and Applied Chemistry (IUPAC): the 15 lanthanides, plus yttrium and scandium. Yttrium and scandium are classified as REEs because they are commonly found in the same ore deposits as lanthanides and share similar chemical properties. Although all REEs occur naturally, they are not found in pure metal form. Due to their distinctive magnetic, phosphorescent, and catalytic properties, REEs have become essential for modern technology. These elements are critical for a wide range of technologies, including cell phones, televisions, LED light bulbs, and wind turbines. The estimated average concentration of REEs in the Earth's crust is between 130 and 240 mg/g, which is significantly higher than that of many other commonly used elements. REEs are crucial components in all high-tech devices. Nd is extensively used in super magnets for disk drives, Ce is vital in auto-catalysts, and all REEs are utilized in the production of flat-panel TVs. Various REE compounds are present in smart batteries that power every EV and HEV. Owing to their unique physical, chemical, magnetic, and luminescent properties, these elements facilitate numerous technological advantages, such as reduced energy consumption, enhanced efficiency, increased speed, durability, and thermal stability. Recently, there has been a rising demand for energy-efficient devices (green technology), particularly those that are faster, lighter, smaller, and more efficient. These advancements are also contributing to the miniaturization and improved efficiency of analytical instruments [59].

Considering the demand and supply gap for REEs, there is a need to adopt the sustainable approach of the circular economy system. To ensure the sustainable use of REEs, it is essential to develop recovery technology from secondary sources. Recycling of spent batteries can be achieved through pyrometallurgical and hydrometallurgical processes, but hydrometallurgy is preferred due to its lighter conditions, higher selectivity, and sensitivity [58].

NiMH batteries are a significant secondary source of REEs, Co, and Ni. Recycling these batteries properly not only contributes to their economic value but also protects the environment. Various authors have described different recycling methods for extracting REEs and other valuable nonferrous metals from used NiMH batteries. The three primary methods employed are mechanical processing, pyrometallurgy, and hydrometallurgy. Mechanical processing typically includes steps such as grinding, screening, and bed elutriation to recover valuable materials. In pyrometallurgy, the thermal decomposition of REEs, such as La, Ce, Nd, and Pr, from spent NiMH batteries is performed in their oxidized forms [53].

Pretreatment methods for the recovery of NiMH batteries, typically categorized as end of life and waste, aim to minimize the quantity of waste NiMH batteries, separate them into different fractions, and produce a black powder, known as battery powder, ready for further processing (**Figure 13**). Early research on NiMH battery recycling primarily focused on recovering Ni-based alloys through ore dressing techniques [53]. Pyrometallurgical processes involve melting the battery material, either after pretreatment to reduce Fe content or as a whole unit, sometimes including plastic components. In the presence of slag (an oxide liquid phase), these processes result in the reduction of Ni and Co elements to form a liquid nickel–cobalt (Ni-Co) alloy, while the REEs are oxidized and dissolved in the slag (**Figure 14**) [53].

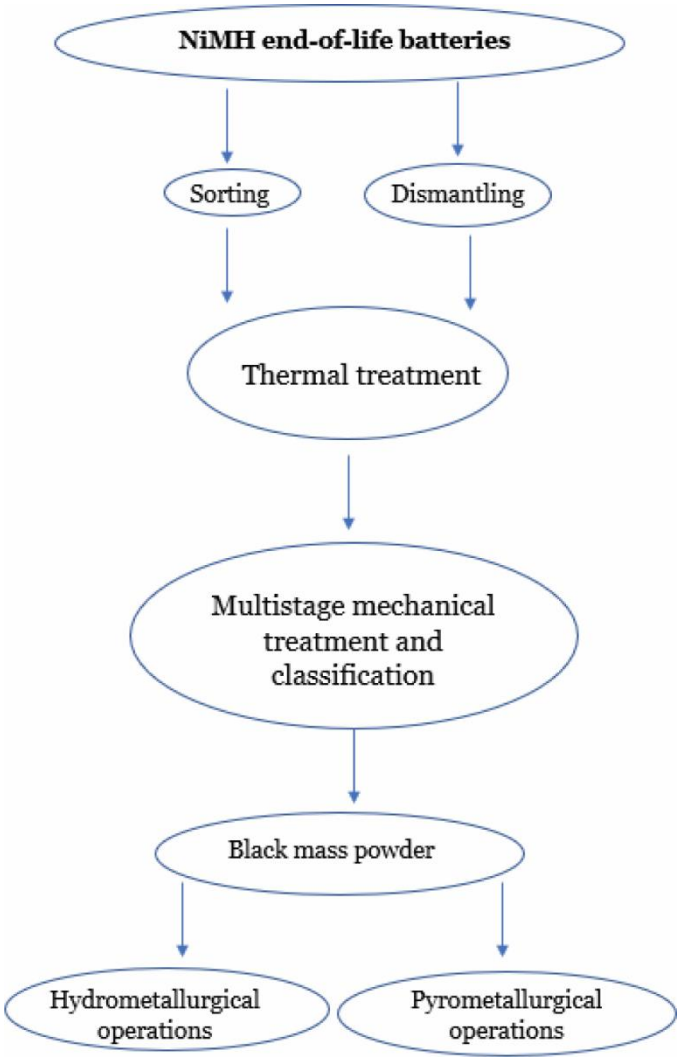

Figure 13 • Flowchart of typical physically based pretreatment of used NiMH batteries (modified from [53]).

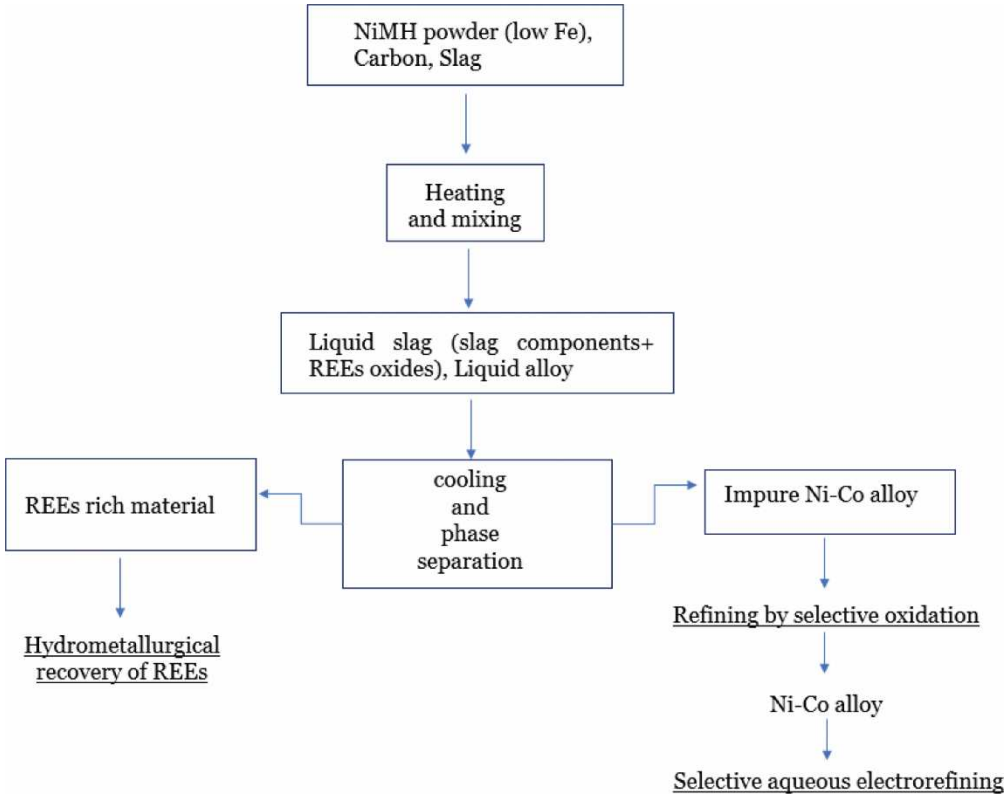

Figure 14 • The principle of pyrometallurgical processing of end-of-life NiMH batteries (modified from [53]).

Various hydrometallurgical processes, including leaching, solvent extraction, and precipitation, have been employed to extract metals from NiMH batteries, utilizing different binders, extractants, and precipitants [60]. Yang et al. [61] conducted leaching of REEs from waste NiMH batteries using hydrochloric acid (HCl), followed by impurity removal with an ammonia solution, achieving a 99% recovery of rare earth oxides. Bertuol et al. [62] reported H_2SO_4 leaching of REEs, followed by selective precipitation with NaOH at pH 1.2. Fernandes et al. [63] reported chloride leaching followed by selective extraction of REEs from NiMH batteries using PC88A. Innocenzi and Veglio [64] also examined H_2SO_4 leaching of REEs from spent NiMH batteries, followed by selective precipitation with NaOH at a pH below 2. Zhang et al. [65] investigated chloride leaching of REEs followed by solvent extraction using D2EHPA. Mei et al. [66] detailed the recovery of REEs via leaching with H_2SO_4, followed by solvent extraction with Cyanex 923, and ultimately obtaining rare earth oxides using oxalic acid. Two different extractants, D2EHPA diluted in kerosene and Cyanex 923, were employed for extracting REEs from spent NiMH batteries. Chemical precipitation is the most commonly used method for REE recovery due to its simplicity and low cost, with anhydrous sodium sulfate and sodium hydroxide being utilized for this purpose. Pietrelli et al. [67] studied H_2SO_4 leaching of REEs, Ni, Co, Cd, and Fe from Ni-Cd and NiMH batteries. Ahn et al. [68] reported the recovery of REEs as mixed oxides through a leaching and precipitation process [60]. Other studies on NiMH batteries, which are the most evaluated among secondary REE sources, are summarized in **Table 3** [68–87].

Table 3 • Studies on the recovery processes of rare earth elements from NiMH batteries [68–87]

Process unit operations	Leaching or *final recovery rates		Ref.
	Metal	Recovery %	
Leaching , precipitation	Y	97% → *54	[70]
	Ce	100 → *99	
	La	100 → *99	
	Nd	99.48	[71]
Leaching	Ce	96.43	
	La	99.14	
	*Nd	85.97	
Leaching (2×),precipitation	*Ce	90.75	[72]
	*La	82.59	
Leaching , precipitation ,oxidation ,solvent extraction	*Ce	84.7	[73]
Leaching ,aqueous biphasic system (3×)	*Ce	8.5	[74]

	*La	47	
Leaching, precipitation, heat treatment	*Nd	97.53	[68]
	*Ce	99.6	
	*La	98.76	
Leaching, precipitation, hydroxide conversion +wet oxidation, selective dissolution	*Ce	97–98	[75]
	La	95.8 → *99.49	
	Ce	89.9 → *99.14	
Leaching, precipitation	Pr	96.3 → *99.12	[76]
	Y	97 → *41.79	
	Sm	98.6 → *95.71	
Mechanical treatment, leaching	La	91.6	[77]
Mechanical treatment, leaching, evaporation and recirculation, precipitation	La	99	[78]
	Ce	99	
Mechanical treatment, leaching	La	68.08	[79]
	Ce	84.61	
	Pr	32.36	
	Sm	61.07	
	Nd	65.95	
Mechanical treatment, thermal oxidation, thermal reduction	REEs separated in the oxide phase		[80]
Leaching, precipitation	La	66.4% → *98%	[81]
	Ce	88.8% → *99%	
	Pr	59.3% → *99%	
Supercritical fluid extraction	*REE	90	[82]
Supercritical fluid extraction	*REE	90	[83]
Leaching	Ce	97.7	[84]
	La	88.7	
Mechanical treatment, leaching, precipitation	La	69.5	[85]
	Ce	89.4	
	Pr	95.5	
	Sm	98.4	
	Nd	98.1	
Leaching, electrochemical purification, precipitation	Ce	96	[86]
	La	96	
	Nd	96	
Mechanical treatment, washing with water, leaching, filtration, precipitation	REEs	98 → *97	[87]

*Final recovery rates. Source: Authors.

5. Conclusions

With the advancement of technology, vehicles powered by internal combustion engines are expected to be replaced by EVs. It is a fact that EVs, especially HEVs, are of crucial contribution to reducing the emission of greenhouse gases, which have long-term harmful effects on human life. Energy storage systems include batteries, fuel cells, solar collectors, and fuel cells. In the last decade, the prominence of energy storage systems, especially batteries, has led to positive effects on human life.

Considering the increase in EVs and the types of batteries used, the demand for certain metals will rise in parallel. The growing demand for critical metals such as Cu, Ni, Co, and Li, used in EVs as part of the clean energy transformation policy, will create a supply problem. The supply problem will also have a direct impact on the rising prices of these metals. Within the scope of countries' net zero emission targets, the concept of recycling comes to the fore. Within the scope of recycling, EV batteries attract attention due to the critical metals they contain.

In this study, the literature on EVs and the types of batteries used is reviewed. In particular, NiMH batteries containing high amounts of REEs are discussed in detail, with an emphasis on the

importance of recycling methods and processes of these batteries. The global supply of REEs has been steadily increasing. Due to their unique properties, REEs find extensive applications, particularly in the energy sector, catalysts, magnets, ceramics, and electronic devices. China, as the dominant nation with the largest reserves and production rates of REEs, has prompted other countries to seek alternative sources due to its export restrictions on these critical materials. Due to declining energy resources and the increasing need for storage systems, the demand for batteries is projected to increase to approximately 10 times the current amount within the next ten years, driven by the anticipated rise in EV production. Recently, the recovery of critical and strategic metals from end-of-life EV batteries, referred to as potential waste, has been gaining importance in order to prevent negative environmental impacts and mitigate economic conditions caused by decreasing raw materials and increasing production costs. The processes applied in beneficiation are generally categorized into three main groups. These include physical (discharge, removal, size reduction and classification, gravity, magnetic, and electrostatic separation), physicochemical (flotation), and chemical (hydrometallurgical and pyrometallurgical) methods. The combination of physical and chemical processes is of absolute importance for the recovery of precious metals from secondary sources. The application of these processes reduces the risk of battery explosion during recycling processes, reduces by-products, avoids complexity in chemical processes, requires smaller capacity chemical beneficiation equipment as pre-concentrates are produced, reduces the need for reagents, and prevents the production of toxic gases from plastics and binders in thermal processes. As a result, a process with numerous economic and environmental advantages can be designed. After discharge, disassembly, size reduction and classification, the resulting coarse battery components can be separated based on their physical properties (conductivity, density, or magnetic susceptibility). Physical methods are a crucial step in the recycling process, as they lead to significant cost reductions and increased recovery rates. Producing a raw material for metal recovery is the primary objective in this case. NiMH batteries used in EVs, especially HEVs, contain high levels of REE and Ni and have much lower amounts of Co, graphite, and Li compared to Li-ion batteries. The recovery of REEs and Ni-Co alloy from NiMH batteries generally involves hydrometallurgical and pyrometallurgical methods following pretreatment processes. It is known that low-purity Ni-Co alloys are formed through pyrometallurgical methods, with REEs dissolving in slags. Further research is required for the recovery of REEs from these slags. For REE recovery using hydrometallurgical methods, battery leaching with H_2SO_4 followed by the selective precipitation of rare earth compounds is also proposed. Subsequently, the extraction of impurities through a combination of precipitation methods and solvent extraction is required. It has been reported that obtaining pure Ni-Co sulfate solutions, which can be further used in electrolysis cells to prepare valuable Ni-Co alloys, is essential [53]. Continued research is also important to determine the final form of the REE products.

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Author contributions

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Conflict of interest

The authors declare no conflict of interest.

Data availability statement

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