batteries

MDPI

Review

Reviews on the U.S. Patents Regarding Nickel/Metal Hydride Batteries

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Abstract: U.S. patents filed on the topic of nickel/metal hydride (Ni/MH) batteries have been reviewed, starting from active materials, to electrode fabrication, cell assembly, multi-cell construction, system integration, application, and finally recovering and recycling. In each category, a general description about the principle and direction of development is given. Both the metal hydride (MH) alloy and nickel hydroxide as active materials in negative and positive electrodes, respectively, are reviewed extensively. Both thermal and battery management systems (BMSs) are also discussed.

Keywords: metal hydride (MH); nickel/metal hydride (Ni/MH) battery; nickel hydride; electrode fabrication; U.S. patent

1. Introduction

The nickel/metal hydride (Ni/MH) battery is an essential electrochemical device for consumer, propulsion, and stationary energy storages. Since its commercial debut in the late 1980s, many researchers have worked diligently in the Ni/MH battery field. Their contributions were publicized through two routes: academic publications and patent applications. While there are several key reviews available on the former [1–16], there is not one on the latter. Therefore, we have organized and summarized the patents related to Ni/MH battery technology in the current review and its companion—Reviews on the Japanese Patents Regarding Nickel/Metal Hydride Batteries [17].

2. Results

As shown in Figure 1, the current review is divided into six categories in the chronological order of technology development, which are active materials → electrode fabrication → cell assembly → multi-cell construction → system integration → application. Because of the extremely large number of U.S. patents on these subjects, we have selected and presented the most representative patents historically and technologically in the current review. More related U.S. patent documents can be found on each U.S. patent presented here.

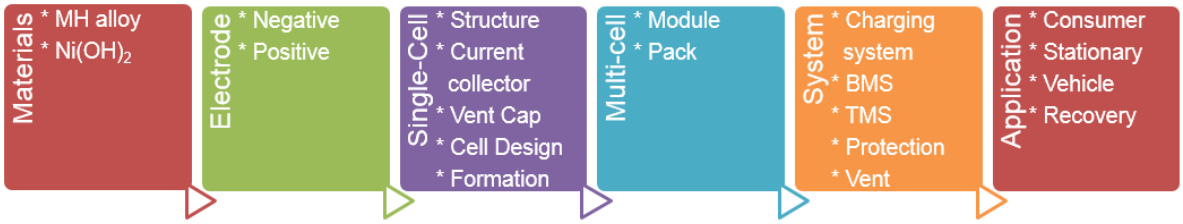

Figure 1. Content of this paper. MH: metal hydride.

2.1. Active Materials

The half-cell electrochemical reaction for the negative electrode is:

$$M + H_2O + e^- \leftrightarrows MH + OH^- \,(\text{forward}: \text{charge, reverse}: \text{discharge}) \quad (1)$$

where M is a hydrogen storage alloy capable of storing hydrogen reversibly and MH is the corresponding metal hydride (MH). During charge, applied voltage splits water into protons and hydroxide ions. Driven by the voltage and diffusion caused by the concentration difference, protons enter into the bulk of the alloy and then meet with electrons from the current collector, which is attached to a power supply. During discharge, protons at the alloy surface recombine with hydroxide ions in the electrolyte. Electrons are injected into the circuit load through the current collector in order to maintain charge neutrality in the negative electrode.

The counter reaction for the negative electrode is:

$$Ni(OH)_2 + OH^- \leftrightarrows NiOOH + H_2O + e^- \,(\text{forward}: \text{charge, reverse}: \text{discharge}) \quad (2)$$

where $Ni(OH)_2$ is the active material going through +2/+3 oxidation state change during the electrochemical reactions. During charge, protons deprived of $Ni(OH)_2$ move to the surface of the positive electrode because of the applied voltage and recombine with hydroxide ions in the electrolyte. In order to maintain charge neutrality, electrons are injected into the power supply through the current collector. During discharge, water at the surface of the positive electrode splits into protons and hydroxide ions. Protons then enter into NiOOH and neutralize with electrons through the load to complete the circuit with the negative electrode. Shown in the net reaction Equation (3), charge/discharge reaction does not change the overall hydroxide concentration/pH value, which is different from the working principle of Ni/Cd battery Equation (4).

$$M + Ni(OH)_2 \leftrightarrows MH + NiOOH \,(\text{forward}: \text{charge, reverse}: \text{discharge}) \quad (3)$$

$$Cd(OH)_2 + 2Ni(OH)_2 \leftrightarrows Cd + 2NiOOH + 2H_2O \,(\text{forward}: \text{charge, reverse}: \text{discharge}) \quad (4)$$

Both the inventions and further developmental works of MH alloy and $Ni(OH)_2$ are presented in the next two sections.

2.1.1. Metal Hydride Alloys in the Negative Electrode

Klaus Beccu contributed to the earliest patent using MH alloy as an active material in the negative electrode to construct a Ni/MH battery in 1970 [18]. The alloy used had a composition of $Ti_{85}Ni_{10}Cu_3V_2$ (in wt%), which was later expanded to any hydride of the third, fourth, or fifth group of transition metal (for example, Ti) [19]. Two types of MH alloys were used in the first commercialized Ni/MH batteries introduced in 1989. While Japanese companies such as Matsushita [20–22] (later changed its name to Panasonic), Toshiba [23], Sanyo [24,25], and Yuasa [26] used the misch metal-based (mixtures of light rare earth elements such as La, Ce, Pr, and Nd) AB_5 MH alloy, the Ovonic Battery Company (OBC, Troy, MI, USA) chose the transitional metal-based AB_2 MH alloy as the active material in the

negative electrode. General properties of these two alloy families are listed in Table 1. The AB_5 MH alloy eventually dominated the market due to the substantially lower price of misch metal since 2000.

Table 1. Property comparison between the AB_2 and AB_5 MH alloy families. FCC, 1C/1C, and DOD denote face-centered-cubic, 1C charge and discharge rates, and depth of discharge, respectively.

Property	AB_2 MH alloy	AB_5 MH alloy
Basic crystal structure	Hexagonal C14 and FCC C15	Hexagonal $CaCu_5$
Composition example	$Ti_{12}Zr_{21}Ni_{38}V_{10}Cr_5Mn_{12}Co_{1.5}Al_{0.5}$	$La_{10}Ce_5Pr_{0.5}Nd_{1.5}Ni_{60}Co_{12}Mn_6Al_5$
Discharge capacity	340–440 mAh· g^{-1}	300–330 mAh· g^{-1}
High-rate dischargeability	Acceptable	Excellent
Cycle life (1C/1C, 100% DOD)	800	1200
Self-discharge	Acceptable (V-free alloy)	Acceptable
High-temperature storage	Acceptable due to leach-out	Bad due to surface passivation
Activation	Modest	Easy
Low temperature	Good	Modest
Fabrication method	Casting, hydriding, and grinding	Casting and grinding
Raw material cost	V is expensive	Volatile due to rare earth price fluctuation
Low-cost version	V and Co-free alloy	Pr, Nd, and Co-free alloy

Discovery of the hydrogen storage capability of the rare earth-based AB_5 intermetallic alloy was accidental [27]. Guegen [28] filed the first patent using $LaNi_5$ modified with Ti, Ca, Ba, Cr, and/or Cu with improved capacity as negative electrode material in 1978. In 1984, Willems *et al.* [29] filed a patent adding Co and/or Cu in $LaNi_5$ to reduce the lattice expansion during hydride process and consequently extending the cycle life. Based on the idea of utilizing misch metal in AB_5 for gaseous phase hydrogen storage originated by Osumi and his coworkers in 1979 [30] and 1983 [31], Kanda and Sato from Toshiba [32] filed a U.S. patent based on Mn, Al-modified misch metal-based AB_5 MH alloy for Ni/MH battery in 1986. Subsequently, the Japanese companies filed many patents optimizing the AB_5 formula (examples can be found in [17,33–42]) and finalized on using Co, Al, Mn, and Ni as the B-site elements. OBC also filed a patent for adding Zr or Si to extend the cycle life of Cu-containing AB_5 MH alloy [43].

In the field of AB_2 MH alloy, Gamo and his coworkers [44] from Matsushita filed a patent about its gaseous phase hydrogen storage application in 1979, and Sapru and her coworkers [45] at OBC filed the first patent for battery application in 1985. Since then, many patents have been filed on the AB_2 MH alloys for electrochemical applications by OBC [46–55], Matsushita [56–58], and SAFT [59].

Other MH alloy families, including body-centered-cubic (BCC) solid solution [60–62], BCC-AB_2 composite [63,64], Mg, Ni-based alloy [65–68], TiNi [69], and composite [70,71], were also proposed as negative electrode active materials; however, they were not commercialized. The only MH alloy that successfully succeeded AB_2 and AB_5 in commercialization is the A_2B_7 superlattice alloy [72–76]. Development works of the A_2B_7 MH alloy done in academia can be found in a previous review article [10]. Several non-conventional MH alloy fabrication processes, such as centrifugal casting [77], double-roller casting [78], melt-spin [34,79], gas atomization [80], strip casting [81,82], and a chemical process using misch metal oxide and/or hydroxide as raw material [83], have been patented as well, but only strip casting is used for mass production besides the conventional induction melting.

Besides patents on a specific alloy family, there are several patents that contribute across the board and independently of alloy selection. Sapru *et al.* [84] filed a patent about the advantage of increasing the degree of disorder in a multi-component MH alloy; as a result, the electrochemical performance is improved. Such material is disordered and characterized as a composite of amorphous, microcrystalline, and polycrystalline components lacking long-range compositional order and with three or more phases [85]. Another group of patents not confined to a specific alloy family addresses

the structure designs of surface oxide, which is a catalyst for the electrochemical reaction [52,86–88] (Figure 2).

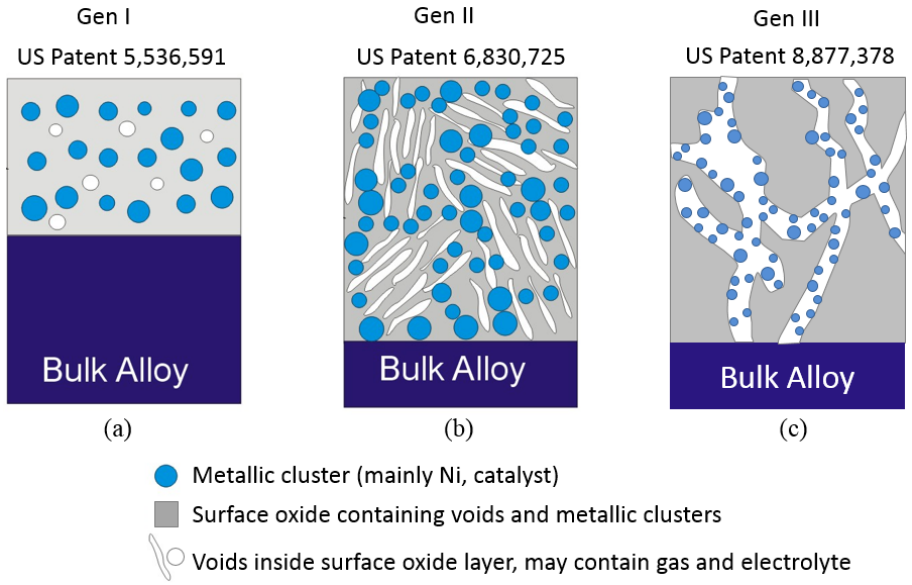

Figure 2. Schematic diagrams of the evolution in MH alloy surface oxide from (**a**) metallic clusters and voids randomly distributed in the surface oxide to (**b**) metallic clusters and channels randomly distributed in the surface oxide and (**c**) metallic clusters only on the surface of channels.

2.1.2. Nickel Hydroxide in the Positive Electrode

Soon after the Swedish inventor Waldemar Jungner filed a number of Swedish and German Patents on the concepts of Ni/Cd and Ni/Fe batteries in 1899 [89], a famous American inventor, Thomas A. Edison, filed several U.S. patents on Ni/Fe and Ni/Cd rechargeable batteries utilizing nickel hydroxide as active material in the positive electrode in 1901 [90,91]. The same chemistry, $Ni(OH)_2$, has been used as a positive electrode active material for the entire alkaline battery family because of its high electrochemical reversibility, appropriate voltage (the highest among several options but lower than the oxygen gas evolution potential), and low cost. The original positive electrode used in Ni/MH batteries was inherited from those in Ni/Cd and Ni/Fe batteries [92–95]. The electrode is manufactured by first pasting filamentary Ni onto a substrate (such as perforated foil) and then "sintered" under a high temperature annealing furnace in a nitrogen/hydrogen atmosphere, where binders used in the pasting process are burned away to leave a conductive skeleton of nickel with a typical average pore size of about 30 μm [96]. Next, a chemical impregnation process is carried out to produce the active $Ni(OH)_2$ by consecutively dipping the electrode between the $Ni(NO_3)_2$ and NaOH baths [97,98]. This kind of $Ni(OH)_2$ electrode is the sintered type. A new pasted type of positive electrode using spherical $Ni(OH)_2$ particles together with the CoO additive (for forming a CoOOH conductive network on the particle surface) was first patented by Oshitani and his coworkers in Yuasa in 1989 [26,99], and soon became the mainstream for Ni/MH battery due to its high volumetric and gravimetric energy densities. The spherical particle is originally produced by a two-step batch chemical precipitation process, which consists of ammonia complex formation and precipitation in two different reactors. Later on, Fierro and his coworkers at OBC [100] developed a continuous stirring single reactor process, and the smaller particles near the bottom of the reactor and the larger ones are separated by the centrifugal force from a bottom blade, leaving the container with an overflow (Figure 3). This method has been widely adopted in today's modern manufacturing. The produced spherical $Ni(OH)_2$ powder is then mixed with binders and pasted onto the Ni foam substrate to form the pasted type of positive electrode. The main advantage of using the pasted type of positive electrode is the increase in gravimetric energy density; however, rate-capability, cycle stability, and cost are sacrificed.

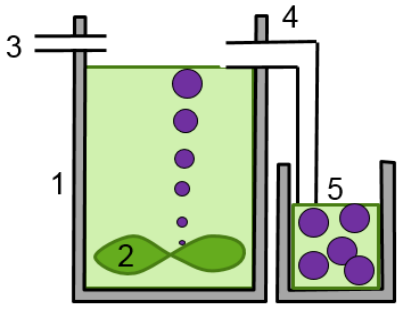

Figure 3. Schematic diagram of a continuous stirring tank reactor used to co-precipitate hydroxides of metals with similar solubilities, such as Co, Ni, and Zn. 1: Stainless-steel vessel; 2: mixer blade; 3: raw material inlet; 4: product overflow; and 5: product container.

There are many patents modifying the basic $Ni(OH)_2$ for performance improvement, such as increasing utilization rate by chemical modification through co-precipitation [25,101–111], promoting higher electron transfer per Ni atom by introducing the gamma phase with an oxidation state between +3 and +4 [112–117], alternating the microstructure by various process changes [118–120], and adding a conductive coating on the $Ni(OH)_2$ core [121]. Moreover, OBC filed four patents regarding the incorporation of fine Ni fiber in the co-precipitation reactor for the purpose of increasing the native conductivity of $Ni(OH)_2$ [122–125]. The crystalline size is an important indicator for the degree of disorder (density of stacking fault along the *c*-axis) of $Ni(OH)_2$ and affects the electrochemical performance, and an evaluation method was patented by OBC [126]. In order to increase the conductivity or resistance to poisoning, a core-shell type of structure obtained by sequential precipitation or other coating methods was invented [108,124,125,127–129].

2.2. Electrode Fabrication

There are two electrodes in a typical Ni/MH battery, namely the negative and positive electrodes, and they are made of MH alloy and nickel hydroxide, respectively. During service (discharge), MH in the negative electrode is oxidized into metal, and therefore the negative electrode is also called an anode. Furthermore, the positive electrode can be termed a cathode since NiOOH is reduced to $Ni(OH)_2$ during discharge.

Patents regarding electrode fabrications of both the anode and cathode, including component, substrate, and construction, are discussed in the following sections.

2.2.1. Negative Electrode (Anode)

The negative electrode in a commercial Ni/MH battery can be fabricated by any of the following methods: dry compaction, wet paste method, or dry paste method (Figure 4).

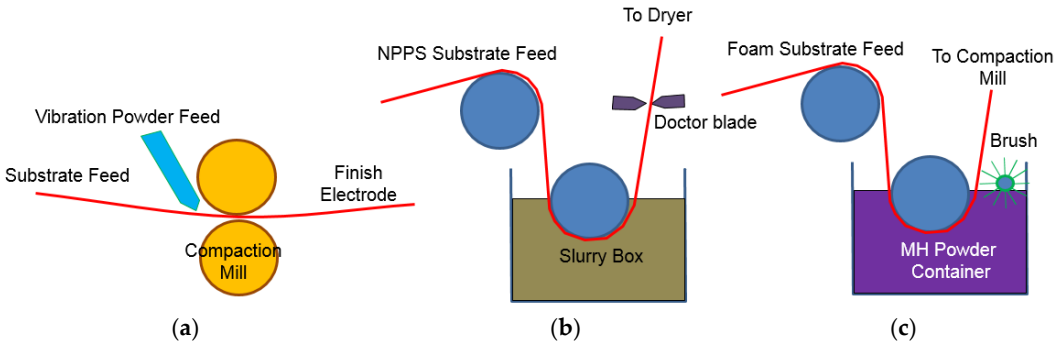

Figure 4. Schematic diagrams of three commonly used nickel/metal hydride (Ni/MH) battery negative electrode fabrication methods: (**a**) dry compaction; (**b**) wet paste method; and (**c**) dry paste method.

OBC filed a patent for its dry compaction manufacturing process in 1989 [130]. While the wet paste method was inherited from the conventional alkaline battery technology, negative electrode fabrication by dry pasting onto the Ni foam through brushing has gained popularity in just the last few years [131]. The pros and cons of each method are summarized in Table 2.

Table 2. Comparisons among three commonly used Ni/MH battery negative electrode fabrication methods. Both the dry compaction and dry paste method can be processed without a binder, and therefore the power density is higher from both cases compared to that from the wet paste method.

Methods	Pros	Cons
Dry compaction	• Higher power density • Lower cost and simple process	• Difficult to control the thickness precisely
Wet paste method	• Longer cycle life • Lower cost	• Lower power density • Complicated process
Dry paste method	• Simple process • Higher power density	• Higher cost

Expanded Ni substrate is typically used for the dry compaction method to reduce the burr that happens with the use of Ni mesh substrate. Ni-plated perforated steel (NPPS) plate [132] and Ni foam are commonly used for the wet paste and dry paste processes, respectively. Other types of substrates, such as porous Ni [133], porous Cu [134], welded Cu [135], cellular metal [136], and conductive ceramic [137], were also patented. Substrate-less electrode fabrication was also proposed, where the active material is first mixed with a conductive metal and subsequently forms the electrode without a substrate [138].

Additives such as carbon and metal powder can be added in the negative electrode to increase the conductivity [139]. Other additives such as rare earth oxides and GeO_2 can be used to reduce the oxidation of the negative electrode [140,141]. Moreover, binders are required for the wet paste method, and many candidates were patented in the past [142–144]. Polyvinyl alcohol is the most popular binder material used in the negative electrode of Ni/MH battery.

In order to remove the passive oxide formed during powder fabrication and form a thin, porous, and electrochemically active oxide on the surface of MH alloy, activation is needed and can be executed at the powder or electrode level with acid [145] or alkaline [146] or in the sealed cell during formation. Furthermore, performing a controllable oxidation on MH alloy before fabricating it into electrode is preferred for the purpose of reducing the native oxide thickness [147,148]. Also, oxygen gas evolution during fast charge was found to be reduced by increasing the final surface roughness [149]. A surface fluorination process was invented for performance enhancement as well [150]. In terms of construction, an auxiliary negative electrode was added to reduce the cell pressure of button cell [151].

2.2.2. Positive Electrode (Cathode)

The most common type of positive electrode in today's Ni/MH battery is the pasted type. Spherical $Ni(OH)_2$ co-precipitated with one or more additional metal hydroxides (Co and Zn being the most common) is mixed with additives, binder, and solvent (water, methanol, or ethylene glycol) to form a paste. The paste is then applied to the Ni-foam substrate, calendared, and dried [152]. A continuous positive electrode fabrication method was patented by OBC in 1994 [97]. The active material in the tab area can be removed or avoided by water wash or ultrasonic cleaning, using a pre-taped foam substrate [153] or reduction by hot hydrogen [154]. Besides the commonly used Ni foam patented by Inco in 1990 [155], metal foil [156], metal mesh [157], Ni mesh/fabric/metal fiber composite [158], and Ni-plated porous Cu/Cu alloy skeleton [159] were also proposed as substrates. Both non-conducting [160] and conducting binders [161] can be used. Since the intrinsic conductivity of spherical particle is poor, a CoOOH conductive network on the surface has been indispensable since the beginning of spherical $Ni(OH)_2$ application [99], an idea originated from its use in the sintered

electrode [162]. Conversion from Co^{2+} to Co^{3+} is usually completed in the sealed cell during the formation process. Pre-oxidation of Co at the electrode level was also patented [163]. Modifications and improvements on the basic Co-compound additives were proposed [23,102,127,164–169]. Other additives such as Ca compound [170], Mn compound [171,172], Al compound [173], carbon [174], and silicate [175] were patented as well. For high temperature application, oxides of rare earth elements (Y_2O_3 and Yb_2O_3 being the most common) are added [176–178]. In today's commercial Ni/MH battery, Y_2O_3 is a must additive in the positive electrode. Besides the pasting method, an electrodeless plating technique was proposed for applying the active material onto the substrate, but it is not commercialized [179,180].

2.3. Cell Assembly

Several key areas such as components (negative and positive electrodes, current collector, venting cap, case, electrolyte, and separator), construction type, design of capacity ratio, and formation process need to be considered in order to complete the cell assembly. The following sections summarize the patents concerning the aforementioned items.

2.3.1. Cell Construction

There are five conventional designs of Ni/MH batteries, and a comparison among them can be found in Table 3. Moreover, a new pouch type of Ni/MH battery providing high energy density was recently proposed [181]. The cylindrical type of Ni/MH battery is the most common and mainly used to replace the disposable primary battery. While the typical cylindrical cell is made by winding the electrodes/separator spirally, a patent from Matsushita used the technique of layering hollow cylindrical electrodes in the form of concentric circles to increase the mutual facing area between the negative and positive electrodes [182]. Examples of prismatic Ni/MH batteries can be found in patents filed by Energy Conversion Devices Inc. (ECD) [183,184] and Toshiba [185–187] and show the most scalability up to 1 kWh. The typical prismatic cells are composed of a stack of electrodes with tabs welded together. Toyota filed a patent using a single piece of folded electrode stack (one separator and multiple positive and negative electrode) in a rectangular cell [188], similar to the design of GIGACELL developed by Kawasaki Heavy Industry [189]. The case and electrode assembly may not be in the same shape. For example, optimizing the amount of electrolyte by placing a spirally wound electrode assembly in a rectangular casing was proposed [190]. A flat wafer cell, similar in design to the coin cell [191], was patented with the use of conductive carbon-filled polymeric outer layers as electrode contacts [192,193].

Table 3. Comparison among five common types of Ni/MH battery. SS: stainless steel. EV: electric vehicle; and HEV: hybrid electric vehicle.

Type	Description	Pros	Cons
Consumer	• Cylindrical • SS case	• Mass production • Compatible with other alkaline batteries	• Size limitation
Stick	• Prismatic • SS case	• Mass production • Easy for integration	• Higher cost • Lower energy density
EV	• Prismatic • SS case	• Large format (100 Ah)	• Hand assembly • High cost
HEV	• Prismatic • Plastic case	• High-volume mass production	• Lower pressure rating • Poor heat transfer
Coin	• Button-shaped • SS case	• High-volume mass production • Compact for medical application	• No vent • Cannot be scaled up

While only separators are placed between the adjacent negative and positive electrodes in the typical Ni/MH cell construction, a special "bipolar" design (originated from its use for a lead-acid battery [194]) also uses a metal sheet as substrate for both electrodes (one side as negative electrode substrate and the other side as positive electrode substrate) (Figure 5). This design substantially reduces the contact resistance between adjacent cells [195–198]. Placing a hydrophobic layer between two adjacent negative electrodes to reduce the hydrogen pressure buildup in the cell is another special arrangement [199]. A monoblock of multi-cell assembly was designed to reduce the contact resistance and improve heat dissipation by integrating flow channels within, which allows the gas or liquid coolant to flow between adjacent cells [200]. A lightweight design where the electrode stacks are connected together in a honeycomb structure was also proposed [201].

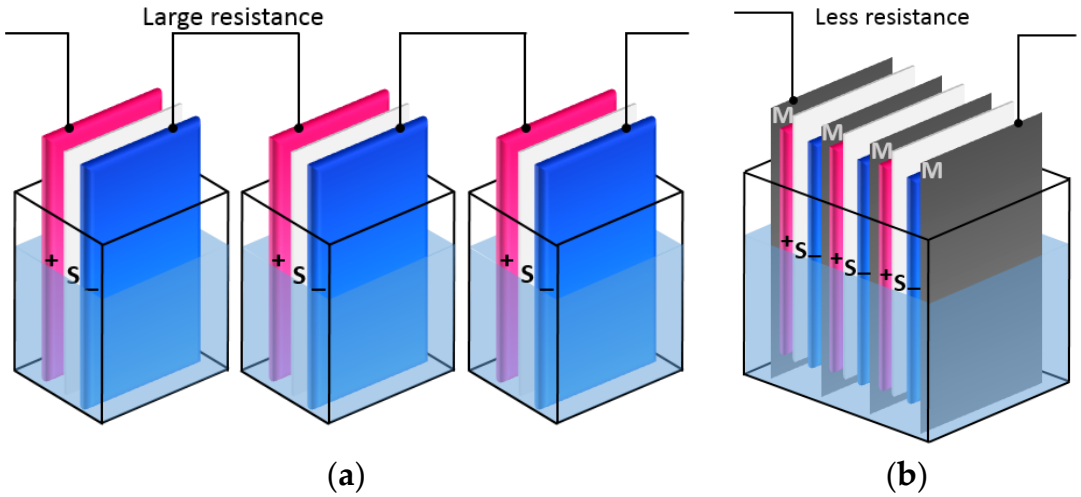

Figure 5. Schematic diagrams of (**a**) regular three cells in series; and (**b**) a bipolar design with three internal cells. +, −, S, and M denote the positive electrode, negative electrode, separator, and metal sheet, respectively.

2.3.2. Current Collector

Electrodes are connected to the terminals located outside of the cell casing through current collectors. Conventional consumer-type of Ni/MH battery (for application of up to 0.5C rate) uses a single Ni tab (or Ni-plated stainless steel (SS) tab) to collect the current from each electrode. The negative tab is stapled onto the negative electrode, and the positive tab is welded to the part of positive electrode without active material (tab area cleaned by ultrasonic cleaning or water jet). Cu was also proposed as the tab material [135]. Generally, the tab only connects to one side of the electrode: in a cylindrical cell, the positive tab is on the top and electrically connected to the safety valve, and the negative tab is on the bottom and grounded to the case; in a prismatic cell, both the positive and negative tabs are on the top. However, in a patent filed by Park, tabs on both sides of the electrode were proposed to reduce the impedance [202]. Usually, the tab is directly connected to the substrate, but there is also a patent suggesting the technique of folding the end of the electrode, which provides a coupling surface for the connection to the tab or terminal [203]. Matsushita [204–206], Sanyo [207], Yuasa [208], and Johnson Controls [209] patented special current collector designs for high-power cylindrical cells (for applications in power tool and hybrid electric vehicle (HEV)). Both laser welding [210] and electron beam welding [211] are popular techniques for welding multiple pieces of tabs together.

2.3.3. Other Components

Other components essential to the construction of Ni/MH battery include a safety vent, case, separator, and electrolyte. Most types of Ni/MH batteries require a safety vent to manage the extra pressure buildup during high-rate charge and also near the end of service life except a coin cell because of its special medical application requirement. The original safety vent designs were inherited from those available for the Ni/Cd battery. Through the years, companies such as ECD [212], Moltec [213], Sanyo [214], Toyota [215,216], and Johnson Controls [217–219] proposed a variety of improvements in the safety vent design. While the pressure limit in a cylindrical cell is around 350–400 psi, in a prismatic cell it is reduced to 100–120 psi because of the easy deformation of the case. Panasonic and Toyota together patented the idea of using a hydrogen-permeable case to release the overpressure within of the cell [220]. Another special case design with multiple ridges on the outside was also developed to improve the cooling performance and mechanical strength of the case [221].

Several different non-woven fabrics were proposed as separator materials previously [222–229], and the most commonly used is the grafted polypropylene/polyethylene type. Recently, a sulfonated separator [230] has become very popular for low self-discharge application since it can trap the nitrogen-containing compound in the cell, which is the main cause of the shutting effect that results in self-discharge [231]. A ceramic, polymer, or composite separator capable of conducting alkali ions was also proposed by Ceramatec [232,233], which can be used to eliminate the cross-contamination of ions leached out from the negative electrode onto the separator and positive electrode. Metal-organic framework, coordination polymer, or covalent-organic framework can serve as separator as well [234]. While most of the electrolyte works were performed in academia [235,236], there are several patents in the area of electrolytes for the Ni/MH battery. For example, Toshiba filed a patent for adding W- and Na-containing compounds to the electrolyte [237]. Certain separators can also serve as electrolytes, and they can be categorized into the gel/polymer and solid types. Electrolyte leakage can be reduced by the use of the gel/polymer type electrolyte. Motorola filed a patent with a KOH-dispersed polyvinyl alcohol or polyvinyl acetate polymer electrolyte [238]. Furthermore, Matsushita filed a patent on a gel electrolyte composed of a water absorbent polymer, a water repellent, and an aqueous alkaline solution [239]. In the solid-state battery field, silicon nitride [240], lithium nitride [241], halide [241], oxysalt [241], phosphorus sulfide [241], and oxide with a perovskite structure [242] were suggested. Among these, perovskite is the most promising. $ABO_{3-\delta}$ perovskite, where A is a rare earth or alkaline earth element with a large radius and B is an element with a smaller radius, is capable of proton conduction. The structure of such an oxide, if prepared under proper conditions (e.g., acceptor-doped), is prone to a large density of oxygen vacancies. When it reacts with water, oxygen from water occupies the vacancy, and the remaining two protons are then attached to two separate oxygen ions, which creates a proton-conducting path. The proton conductivity depends on the vacancy density and also the distance between neighboring oxygen ions.

2.3.4. Cell Design

In a typical Ni/MH cell, an oversized negative electrode is needed to create an overcharge reservoir (OCR) and an overdischarge reservoir (ODR) (Figure 6) [243,244]. OCR is created to absorb the hydrogen gas produced during overcharge and also to provide a recombination center to react with the oxygen gas evolved from the positive electrode. ODR is created to prevent oxidation of the negative electrode during overdischarge (cell reversal). Typical negative-to-positive ratios (N/P) are around 1.2 for a high-capacity cell, 1.4–1.6 for a consumer cell, and 1.8–2.2 for a high-power cell [245]. Methodology for the N/P design can be found in several patents [245–247].

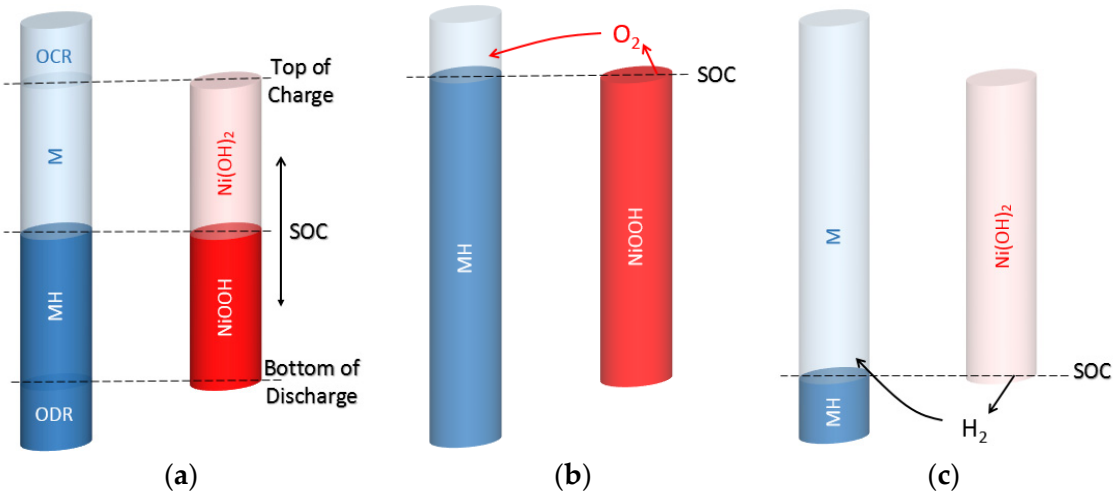

Figure 6. Schematic diagrams demonstrating the positive-limit cell design in Ni/MH battery: (**a**) during regular operation (operating within the limit); (**b**) in the state of overcharge, where the oxygen gas evolved from the positive electrode recombines with the excess hydrogen stored in the negative electrode (OCR prevents hydrogen evolution from the negative electrode); and (**c**) in the state of overdischarge, where the hydrogen gas evolved from the positive electrode is stored in the negative electrode (ODR prevents oxygen gas evolution from the negative electrode). SOC: state of charge.

ODR does not exist when the battery is first built. Later on, ODR starts to increase due to oxidations of MH alloy (Equation (5)), $Co(OH)_2$ (Equation (6)), separator, and other metal components in the cell.

$$3M + H_2O \rightarrow MO + 2M + H_2 \rightarrow MO + 2MH \,(\text{assuming metal in } +1 \text{ oxidation state}) \quad (5)$$

$$Co(OH)_2 + M \rightarrow CoOOH + MH \quad (6)$$

With further cycling, ODR continues to grow and suppresses OCR, and finally the cell vents without the protection from OCR. Therefore, it is important to introduce a "pre-charge" to the positive electrode in order to compensate for the increase in ODR during formation when a low N/P is used [248,249].

2.3.5. Formation Process

After the cell is closed, a series of formation/activation is performed. The process is composed of two parts: a thermal activation and an electrochemical formation (Figure 7). Thermal activation dissolves the Co-compound additives in the positive electrode and also dissolves the native oxide formed on the surface of MH alloy during fabrication. Electrochemical formation applies several charge/discharge cycles to crack the MH alloy and create new and electrochemically active surfaces, and it also converts the dissolved Co into a CoOOH conductive network on the surface of $Ni(OH)_2$. Various formation schemes have been patented [250–254], but they may need to be optimized to prevent cell venting. Also, it was shown that keeping a low state of charge (SOC) during battery routine operation helps ensure a long cycle life [251].

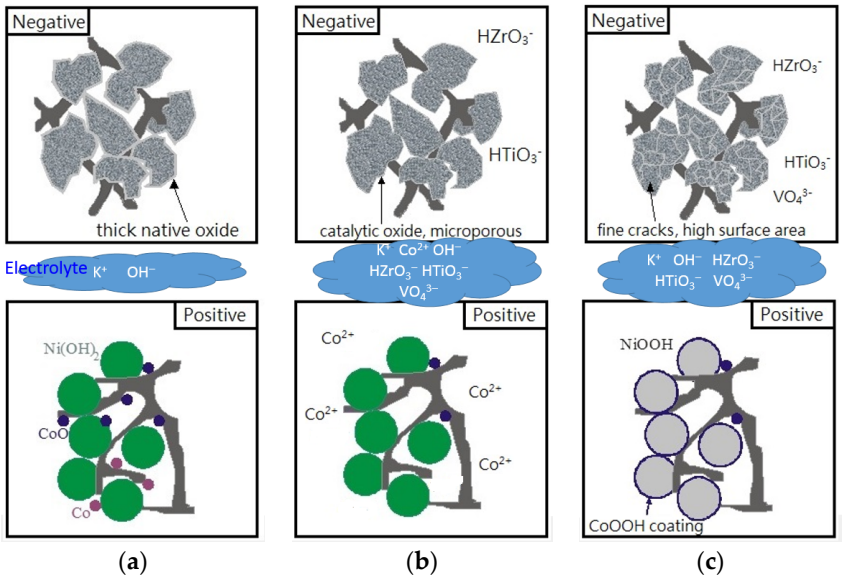

Figure 7. Schematic diagrams showing the formation process of Ni/MH battery: (**a**) before formation, where the MH alloy surface is covered with a thick native oxide; (**b**) after thermal activation, where a new catalytic oxide with metallic Ni embedded within is formed on the MH alloy surface, and bi-valent Co is dissolved; and (**c**) after electrochemical formation, where new surfaces are formed due to MH alloy cracking, and tri-valent Co conductive network is formed on the spherical particle surface in the positive electrode.

2.4. Multi-Cell Construction

The nominal voltage of a single Ni/MH cell is 1.2 V. In order to increase the voltage, several cells have to be connected in series to form a multi-cell module. For consumer use, a module composed of 4 (4.8 V) or 10 (12 V) cells is common. For the propulsion application, multiple modules are assembled together to form a pack. For example, the 1.3 kWh braking energy storage system for the Toyota 2010 HEV Prius (Generation IV) is composed of 168 cells in 28 modules, and each cell is a 6.5 Ah Ni/MH prismatic cell [255]. Patents regarding the module and pack constructions are reviewed in the following sections.

2.4.1. Module Assembly

A module can be made of a series of reparably individual cells with metal [184,256,257] or plastic cases [258], or it can be built with cells isolated by compartment dividers in a single case [259]. Inter-cell conduction is the subject of a number of patents [260,261]. Special module designs for cooling performance improvement include strategic placement of thermally conductive case material and negative electrodes [183] and adding coolant flow channels [262], heat exchanger [263,264], or cooling fins [265,266] between adjacent cells. A compressible two-plate spacer was designed to be placed between adjacent cells for module swelling reduction during operation [267]. For smaller applications in cellphones [268], portable power tools [269], and personal health devices [270], a multi-cell module is the final pack integrated with other system components.

2.4.2. Pack Assembly

When assembling several battery modules into a single pack, both the electrical interconnection between modules and integrity of the connectors are very important. While the former can be ensured by a dedicated welding methodology [271], the latter can be secured by strategically filling the gaps between modules with packaging material [272]. Thermal management of Ni/MH battery pack is also critical to achieve a long service life, which can be done by using either forced air [273,274] or a liquid

coolant [275–277]. Moreover, the use of a heat sink is helpful to increase the thermal dissipation rate of the battery pack [278].

2.5. System Integration

For application of a larger energy storage system (for example, in EV), many sub-systems need to be integrated, such as a charging system, a battery management system (BMS), a protection system, and a thermal management system (TMS), to ensure a smooth and safe operation. Patents on these sub-systems are discussed in the following sections.

2.5.1. Charging System

For consumer application, a charger is a separate unit from the battery. In the early days, a different charger was needed for the Ni/MH battery but with a very similar design principle. Later charger hardware improvements made charging both Ni/Cd and Ni/MH batteries with the same charger possible. Numerous patents regarding the portable charger hardware design are available [279–294]. Most of the charging algorithms are composed of several different cut-off schemes, such as ΔT (temperature change), $-\Delta V$ (negative voltage change), $\mathrm{d}V/\mathrm{d}t$ (voltage change with time), $\mathrm{d}T/\mathrm{d}t$ (temperature change with time), maximum input, and V_t (terminal voltage) [295–320]. End-of-charge can also be detected by electronic circuit alone [321]. In order to fast charge, special hardware [322] and algorithms [323,324] are required. For application in EV, the charging unit is integrated into BMS and is a part of the system.

2.5.2. Battery Management System

When a large number of cells are used together, BMS is indispensable [325]. Even in a portable power tool, the SOC estimation of Ni/MH battery is necessary [326]. Due to its high abuse tolerance, BMS for Ni/MH battery system is much simpler than that for a Li-ion battery system. For example, voltage monitoring and SOC recording at the cell level in Ni/MH battery system are not necessary. Therefore, for Ni/MH battery systems used in vehicles, the main functions of BMS are monitoring charge/discharge [327–332] and voltage [333–336], estimating SOC [337–344] and state-of-health (SOH) [345–351], communicating with the on-board computer system, and handling emergencies. In order to obtain an accurate estimation, several methodologies, such as integral amp-hour [352–354], self-discharge calculation [355], calibration from voltage variation [356,357], and impedance measurement [358,359] are used in combination.

2.5.3. Protection System

Originated from the overcharge protection circuit invented for Ni/Cd battery [360–364], a similar design was patented for Ni/MH batteries [365,366]. Over-temperature protection [367] and under-voltage [368] circuitries for Ni/MH battery were previously patented. The design of putting a capacitor in parallel with the battery was also invented to offer an emergency power source [369–371]. The addition of a high-voltage connector was suggested for electrical shock prevention [372]. Finally, a venting system for automobiles equipped with Ni/MH battery system was proposed to release hydrogen from a strategic location [219].

2.5.4. Thermal Management System

Thermal dissipation for Ni/MH battery is performed either at the cell [184,256,257] or module level [189,262,263,265,266,373–376]. Compared to TMS for Li-ion battery pack, a much simpler TMS is needed for Ni/MH battery pack [373]. For example, the Ni/MH battery system equipped in Toyota HEV Prius requires only a three-speed 12 V DC blower [377], and its motor speed is determined by both the take-in air temperature and cell temperature. Furthermore, Toyota filed a patent regarding the design of cooling control in a battery pack [378].

2.6. Others

As the last part of this review, U.S. patents on the application and recycling of Ni/MH battery are presented.

2.6.1. Application

Key applications for Ni/MH battery are summarized in Table 4 [11]. For a consumer battery, a hybrid Ni/MH battery/capacitor device was patented by Motorola to handle pulse power communication [379,380]. Patents filed by OBC [381] and Delphi [382] described the use of Ni/MH batteries in HEV. Applications in uninterrupted power supply (UPS) [318,383] and grid [384–386] were also patented. A window construction combining Ni/MH battery and solar cell was proposed [387].

Table 4. Key applications for Ni/MH battery in the market. UPS: uninterrupted power supply.

Application	Examples	Main Competitor	Advantages over Competitor
Consumer	AA, AAA, cordless phone, shaver, power tool	Ni/Cd	• Higher energy density • Environmentally safe
Propulsion	EV, HEV	Li-ion	• Long service life • Excellent abuse tolerance
UPS	Cellphone communication hub, vending machine	Lead-acid	• Higher energy density • Longer service life • Environmental safe
Grid	Solar and wind energy storage, voltage equalizer	Lead-acid	• Higher energy density • Longer service life • Environmental safe • Higher instant charge acceptance

2.6.2. Revivification and Recycling

In order to reuse the battery pack, a refreshment method was proposed to replace or re-inject electrolyte into cells with bad performance [388]. At the stage where recovery is impossible, the Ni/MH battery is ready for recycling. Because of the high flammability of the negative electrode, rupturing the battery under anaerobic conditions and flooding the battery interior with CO_2 in an anaerobic chamber is recommended [389]. Recycling can be performed at the component level by separating the MH alloy (AB_2 [390] and AB_5 [391]) and binder [392] or at the individual element level by aiming at rare-earth metals [393–397], Zr [395], Fe [396], Al [396], Co [396,397] and Ni [397,398].

3. Conclusions

U.S. patents on the Ni/MH battery have been reviewed and categorized. With close to 400 U.S. patents cited, this report summarizes the efforts to improve the performance and expand the applicability of Ni/MH batteries undertaken by scientists and engineers working in various companies; these can be used to study the principle and fabrication process systematically. By learning what has been patented previously, we can continue to explore uncharted territory in the science and engineering of Ni/MH batteries.

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Abbreviations

Ni/MH	Nickel/metal hydride
MH	Metal hydride
OBC	Ovonic Battery Company
FCC	Face-centered-cubic
1C/1C	1C charge and discharge rates
DOD	Depth of discharge
BCC	Body-centered-cubic
NPPS	Ni-plated perforated steel
ECD	Energy conversion devices
SS	Stainless steel
EV	Electric vehicle
HEV	Hybrid electric vehicle
OCR	Overcharge reservoir
ODR	Overdischarge reservoir
N/P	Negative-to-positive ratio
SOC	State of charge
BMS	Battery management system
TMS	Thermal management system
ΔT	Temperature change
$-\Delta V$	Negative voltage change
dV/dt	Voltage change with time
dT/dt	Temperature change with time
V_t	Terminal voltage
SOH	State of health
UPS	Uninterrupted power supply

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