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Review

Reviews of European Patents on Nickel/Metal Hydride Batteries

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Abstract: Patent applications in the field of nickel/metal hydride (Ni/MH) batteries are reviewed to provide a solid technology background and directions for future developments. As the fourth review article in the series of investigations into intellectual properties in this area, this article focuses on 126 patent applications filed by European companies at the European Patent Office, while the earlier articles dealt with those from USA, Japan, and China. The history and current status of the key companies in the Ni/MH battery business are briefly discussed. These companies are categorized by their main roles in the industry, i.e., battery manufacturer, metal hydride alloy supplier, separator supplier, and others. While some European companies are pioneers in bringing the Ni/MH product to customers, others have made significant contributions to the development of the technology, especially in the button cell, bipolar cell, and separator areas.

Keywords: nickel metal hydride battery; European patent; metal hydride alloy; nickel hydroxide; European battery manufacturer

1. Introduction

Nickel/metal hydride (Ni/MH) batteries are widely used in consumer electronics, stationary energy storage, and hybrid electric vehicles (HEV). Their advantages are in energy density (compared to the replaced NiCd), cycle stability at deep discharge (>3000), robust construction, ability to be quickly charged, environmental friendliness, and higher abuse tolerance (compared to the Li product) [1]. Two types of Ni/MH batteries are available: the small consumer type (usually cylindrical with a capacity <4 Ah) and the larger transportation type (usually prismatic with a capacity >6 Ah). Nowadays, most consumer type and HEV Ni/MH batteries are produced in China and Japan. Nevertheless, European companies still have a small portion of the market share (for example, about 10% in 2013 [2]) (Figure 1a). On the other hand, European countries are a large market for consumer type Ni/MH batteries. For instance, in 2013, Germany, the Netherlands, Belgium, and the UK accounted for a combined 8.3% of the Ni/MH batteries exported from China [2] (Figure 1b), and in February 2014 Europe received 32.7% of the Ni/MH batteries from Japanese manufacturers (mainly FDK, Tokyo, Japan) [2] (Figure 1c). European companies made great contributions to the debut of Ni/MH batteries in their early days. For example, Klaus-Dieter Beccu from Battelle Memorial Institute (Geneva, Switzerland) contributed to the earliest patent using MH alloy as the active material in the negative electrode to construct a Ni/MH battery in 1970 [3]; Annick Percheron-Guégen and her coworkers from the Agence Nationale de Valorisation de la Recherche (Neuilly sur Seine, France) filed the first patent of using $LaNi_5$ modified with Ti, Ca, Ba, Cr, and/or Cu with improved capacity as

negative electrode material in 1978 [4]; and Johannes Willems, Johann van Beek, and Buschow Kurt from Philips (Eindhoven, Netherlands) filed a USA patent adding Co and/or Cu in $LaNi_5$ to reduce the lattice expansion during hydride process and consequently extend the cycle life in 1984 [5]. In the past, we have reviewed the Ni/MH-related patents (or patents applications) filed in the USA [6], Japan [7], and China [8]. In this review, we focus on patent applications to the European Patent Office submitted by European companies; these applicants are grouped as battery manufacturers, metal hydride (MH) alloy suppliers, separator suppliers, and others. The patent applications can be found online through the website of World Intellectual Property Organization (WIPO) [9]. In the case of a non-English description, a machine translation option supplied by WIPO is available free of charge. A brief introduction of European companies contributing to the European Patent Application (EPA) is included in Table 1.

Table 1. European companies related to Ni/MH battery manufacturing included in this review, in order of their presence.

Company Name	Headquarter	Products Related to Ni/MH	Trademark	Website
Saft	Bagnolet, France	C, D, F-size cells		http://saftbatteries.com/
Arts Energy	Nersac, France	C, D, F-size cells		http://www.arts-energy.com
Varta	Ellwangen, Germany	AAA to FA, button cell		http://www.varta-microbattery.be/
Nilar	Täby, Sweden	Prismatic, bipolar cell		http://www.nilar.com/
Alcatel	Paris, France	Cell phone battery		http://www5.alcatel-lucent.com
Hoppecke	Brilon-Hoppecke, Germany	Prismatic cell		https://www.hoppecke.com/
GfE	Nuremberg, Germany	MH alloy powder		http://www.gfe.com
Treibacher	Althofen, Austria	MH alloy powder		https://www.treibacher.com
SciMat	Swindon, UK	Separator		Not available now
Freudenberg	Weinheim, Germany	Separator		http://www.freudenberg.com

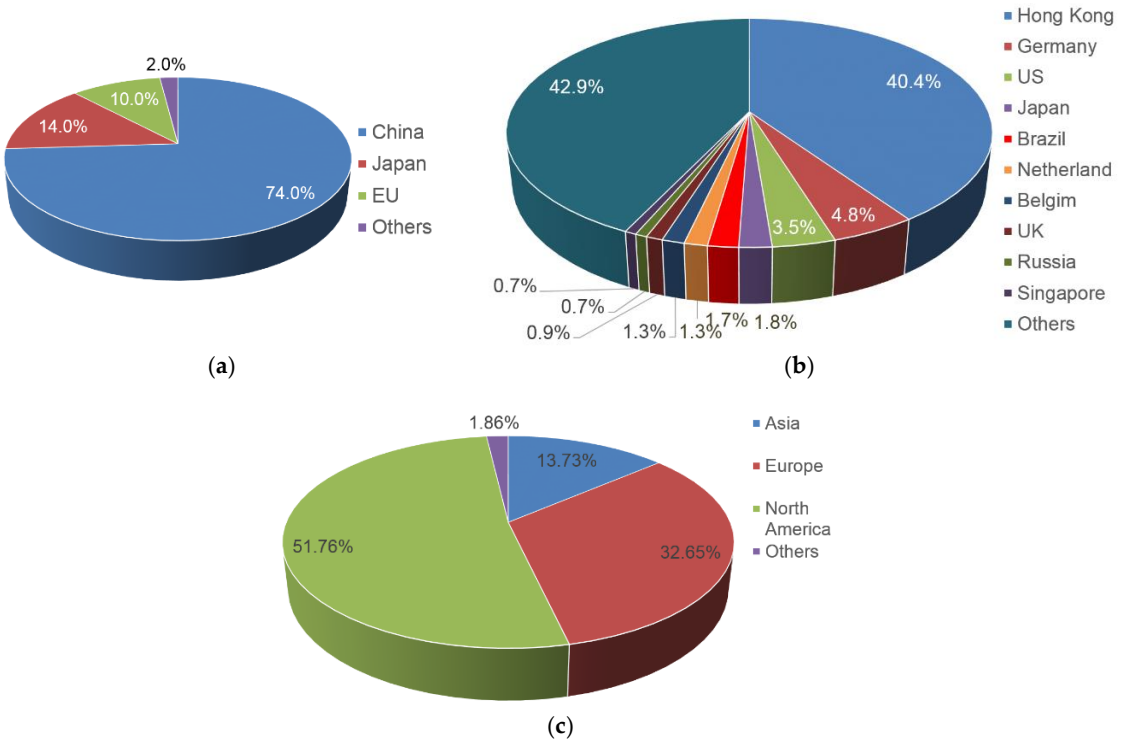


Figure 1. Consumer Ni/MH battery market shares for (**a**) Sources in 2013, (**b**) Exports from China in 2013, and (**c**) Exports from Japan in February 2014. Data originate from [2].

2. Ni/MH Battery Manufacturer

Patent applications from five European battery/chemical companies related to the fabrication of Ni/MH batteries are reviewed in this section. There are two large European companies, Varta (Ellwangen, Germany) and SAFT (Bagnolet, France), that manufactured consumer-type Ni/MH batteries and submitted patent applications in related fields over the past two decades. In 2002, Varta microbattery spun off from Varta and focused on the button cell business. In 2013, SAFT sold its Small Nickel Battery division to Arts Energy (Nersac, France). Alcatel (later merged into Lucent Technology, Paris, France) made Ni/MH batteries for cell phones and was active in filing patent applications. Recently, Nilar (Täby, Sweden) began to offer bi-polar prismatic Ni/MH batteries for transportation applications. Hoppecke (Brilon-Hoppecke, Germany) made prismatic flooded-type Ni/MH batteries for transportation applications.

2.1. SAFT and Arts Energy

The *Société des Accumulateurs Fixes et al Traction* (SAFT) was founded in 1918 and manufactured batteries for the luggage carts and lighting in locomotives. In 2006, Saft and Johnson Controls (Milwaukee, WI, USA) formed a joint-venture pursuing batteries for electric vehicles (EV) and HEV. It was acquired by Total (Courbevoie, France) and delisted from the stock exchange in 2016. SAFT filed 24 and 19 European patent applications (EPAs) from 1990 to 2013 in the name of Accumulateurs Fixes and SAFT SA, respectively. In the positive electrode area, SAFT filed EPAs on the following subjects: a fabrication method of co-precipitated metal hydroxide [10], a description of characteristic of β-$Ni(OH)_2$ [11], a fabrication method of pasted electrode [12,13], and uses of several additives including Celestine [14], CoO with a spinal structure [15], active carbon [16], $Li(OH)_2$ [17], $Ba(OH)_2$ and $Sr(OH)_2$ [18], Nb_2O_5 [19], non-conductive fiber [20], and plastic binders [21–23]. Out of these inventions, those with additives and binders are particularly helpful in improving the performance of positive electrodes. In the negative electrode area, SAFT filed EPAs on the following subjects: active

materials made from a single phase TiNi-based alloy [24], a Zr-based C14 AB_2 alloy [25], a mixed-metal based AB_5 alloy [26], superlattice alloys containing Y [27], Mg [28], or Zr (Ti) [29] and A_2B_7 [30] and A_5B_{19} [31] phases, a catalytic substrate [32], a hydrophilic additive [33], and a paste process [34]. Among these EPAs, the superlattice alloy-related patents lead to a new direction of material research in MH alloy field. SAFT filed multiple EPAs on the subject of a foam substrate and associated current collector design [35–41] (Figure 2), which built a strong foundation for further research in the area. In the separator area, SAFT filed EPAs on microporous grafted membranes [42,43], a tri-layer composite [44], a double-layer with different orientations [45], and a N-retaining substrate [46]. SAFT also filed three EPAs on a charging algorithm [47–49] and one on a leak-tight box for holding battery modules [50].

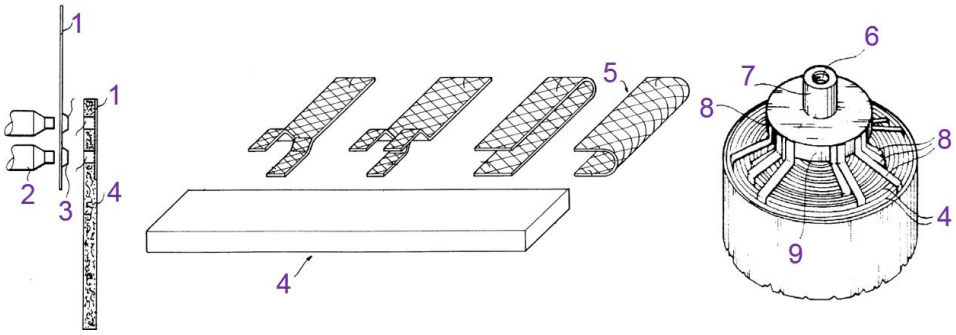


Figure 2. Schematics of three difference designs of current collectors by SAFT. The parts are 1: metal plate, 2: plug, 3: indent, 4: electrode [37]; 5: expanded metal or perorated metal strip [38]; 6: threaded holes, 7: terminal assembly, 8: conducting strip, and 9: shoulder [40].

Arts Energy filed two EPAs in the early days; one on the re-sealable safety valve [51] and the other one on a battery cooling module design [52]. Arts Energy continues to perform research and produce Ni/MH batteries after it was purchased from SAFT in 2013, but has not filed an EPA since then. Its products are mainly cylindrical NiCd and Ni/MH batteries (Figure 3).

Figure 3. Cylindrical cells from Arts Energy [53].

2.2. *Varta and Varta Microbattery*

Varta (originally *Vertrieb*, *Aufladung*, *Reparatur Transportabler Akkumulatoren*) was founded in 1887 and produced the batteries that powered the first EV in Germany. For the production of Ni/MH batteries, Varta formed a joint-venture, 3C Alliance, with Toshiba (Tokyo, Japan) and Duracell (Bethel, CT, USA) in 1996. Varta filed 10 EPAs in 1994 and 1995, which cover an oxidation-resistant graphite additive in the positive electrode [54,55], a use of foam or felt electrode in the button cell [56], a multi-segment cylindrical cell design [57] (Figure 4a), a button cell design [58,59] (Figure 4b), Fe- [60], Ti(Zr)- [61] containing AB_5 MH alloys, and metal recovery from spent batteries [62,63]. Out of these EPAs, those related to the new cell structure designs are important to extend the applicability of Ni/MH into various fields.

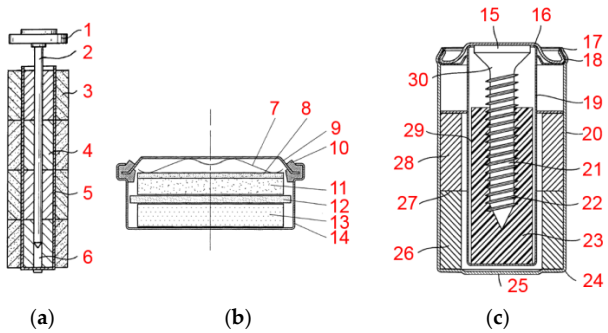


Figure 4. Schematics of (**a**) A multi-segment cylindrical cell, (**b**) A button cell, and (**c**) A hollow-cathode cylindrical cell from Varta. The parts are 1: terminal, 2: conducting pin, 3: positive electrode, 4: negative electrode, 5: separator, 6: central hole [57]; 7: pressure spring, 8: current collector plate, 9: cover, 10: gasket, 11: negative electrode, 12: electrolyte-saturated separator, 13: positive electrode, 14: case [58]; 15: head, 16: disc shape lid with radial profile, 17: rim (bent radially inward), 18: seal, 19: separator, 20: cylindrical wall, 21: shank, 22:thread, 23: negative electrode, 24: can, 25: flat circular bottom, 26: one positive electrode, 27: positive electrode assembly, 28: another segment of positive electrode, 29: cavity, 30: screw shape current collector [64].

In 2002, Johnson Controls acquired the automotive battery division of Varta. Immediately before the sale, Varta Automotive GMGB filed an EPA on an electrolyte containing soluble Al-compound added in the positive electrode to improve high-temperature charge acceptance [65].

Varta Microbattery was a spin-off company from Varta in 2002 and is now owned by Montana Tech Components (Menziken, Switzerland). Their main products are button cells (Figure 5). Between 1998 and 2015, five EPAs were filed with Varta Microbattery as the assignee. They covered the areas of a charging device for Ni/MH battery [66,67], a hollow-cathode cylindrical cell design [64] (Figure 4c), and a battery/supercapacitor composite design [68,69]. The combination of rechargeable battery and supercapacitor increases the high-rate charging capability of Ni/MH battery and opens the door for some power-demanding applications.

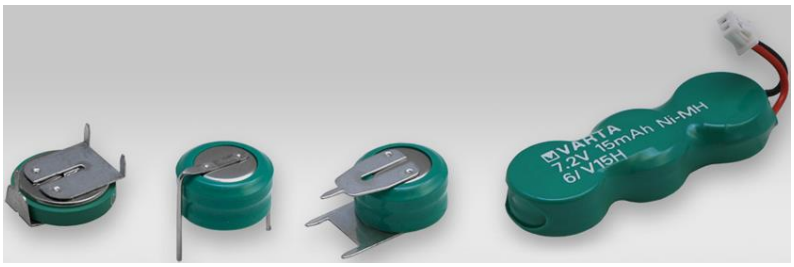


Figure 5. Button cells from Varta Microbattery [70].

2.3. Alcatel

Alcatel (originally short fir the *Société Alsacienne de Constructions Atomiques, de Télécommunications et d'Électronique*) was merged into CGE in 1966, which was later merged again with Lucent Technologies in 2006 and formed Alcatel-Lucent. It was finally merged into the Nokia Group in 2016. Alcatel made Ni/MH batteries for cell phones and cordless phone (Figure 6). Between 1994 and 2003, Alcatel filed 8 EPAs under the names of Alsthom CGE Alcatel and CIT Alcatel. They disclosed two designs for metal current collectors [71,72] (Figure 7), a minus ΔV cut-off charge control method [73], a C14 AB_2 MH alloy [74], a coated, perforated metal substrate for a negative electrode [75], a double-layered separator [76], a cell design with a negative/positive ratio >1.15 [77], and a $Ni(OH)_2$ coating on a MH alloy to improve low-temperature and cycle performance [78]. The last EPA is very important for the use of Ni/MH battery in areas with very harsh winter, Siberia, for example.

Figure 6. Alcatel One Touch 535 with Ni/MH battery [79].

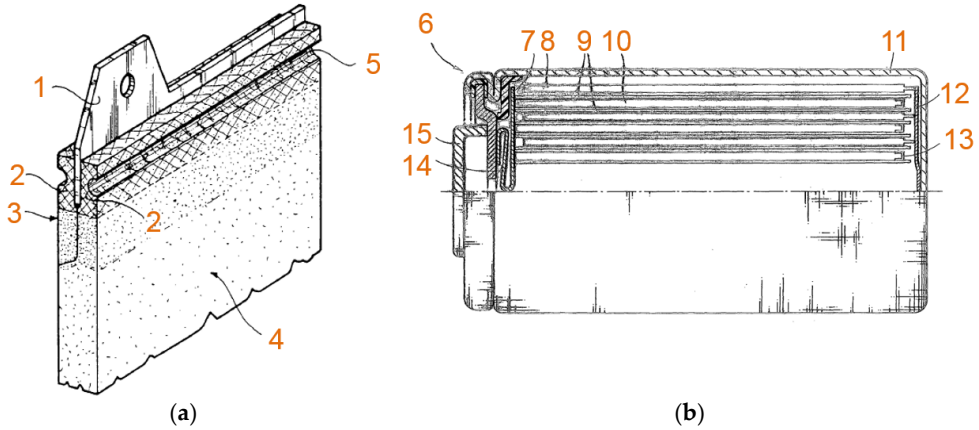
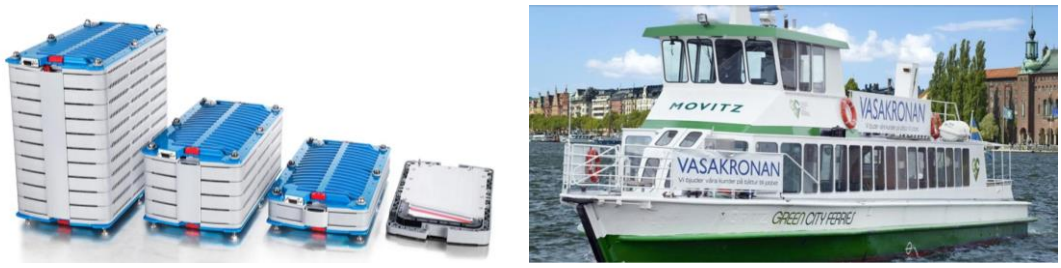


Figure 7. Schematics of (**a**) A metal-foam connection and (**b**) A cylindrical cell design from Alcatel. The parts are 1: metal strip, 2: foam without $Ni(OH)_2$, 3: auxiliary foam, 4: main foam, 5: slot formed by compression [71]; 6: folded part of can, 7: metal tab, 8: negative electrode, 9: separator, 10: positive electrode, 11: can, 12: a piece of nickel plated steel disc, 13: adhesive tape, 14: lug, and 15: cover [72].

2.4. Nilar

Nilar was founded in Sweden in 2011. Before moving to Sweden, Nilar used to operate its research facility in Aurora, CO, USA. Nilar's main product is a prismatic, bi-polar Ni/MH battery used in transportation applications (Figure 8). Nilar owns 14 USA Patents covering components, casing, assembly, module construction of a bi-polar design Ni/MH battery. Nilar filed 13 EPAs between 2002 and 2012, which included assembly of bipolar plates design [80] and manufacture [81], design [82] and manufacture [83–86] of a current collector, a case [87], a stacking arrangement [88], a pressure sensor [89], a powder reduction unit [90], a gasket design [91], and an energy managing system [92] for bipolar Ni/MH batteries. These bi-polar Ni/MH batteries are very important for large-scale transportation applications, which require both large capacity and high-rate drain capability. Recently, Prof. Dag Noréus filed an international Patent Cooperation Treaty (PCT) with Nilar regarding the addition of H_2, O_2, and/or H_2O_2 in sealed Ni/MH batteries to extend cycle life [93].



(**a**) (**b**)

Figure 8. (**a**) Nilar's bipolar prismatic Ni/MH batteries and (**b**) A ferry powered by them [94].

2.5. Hoppecke

Hoppecke was founded in 1927 to produce industrial batteries. It supplied lead-acid, Ni/MH (Figure 9), and Li-ion batteries to railway applications. Hoppecke filed two EPAs in 2011 disclosing a positive electrode manufacturing process [95] and a flooded design for a Ni/MH battery [96]. The latter is unusual and opens the way for stationary storage associated with alternative energies, such as solar and wind power.

Figure 9. Prismatic flooded Ni/MH batteries from Hoppecke [97].

3. MH Alloy Suppliers

GfE (Nuremberg, Germany) and Treibacher (Althofen, Austria) were previously the only two European companies producing MH materials used as active materials in the negative electrode of Ni/MH battery. Nowadays, all the Ni/MH battery manufacturers have switched to vendors from Japan and China.

3.1. GfE

GfE was founded in 1911 to produce metal products with electric furnaces. Today, it still offers an AB_2-based MH alloy, mainly for the use of solid hydrogen storage [98]. GfE filed one EPS in 1999 regarding addition of a metal oxide (Cr_2O_3, for example) to hydrogen storage material (Mg, for example) as a catalyst [99].

3.2. Treibacher

Treibacher Industries AG was founded in 1898 to produce ferroalloy for the steel industry. Today, it still offers AB_2 and AB_5 MH alloys for hydrogen storage and battery applications, respectively [100]. Treibacher filed an EPA in 2001 to cover a Cr-containing AB_5 MH alloy [101]. It also filed an international PCT on a two-chamber hydrogen storage canister [102].

4. Separator Suppliers

Scimat (Swindon, UK) and Freudenberg (Weinheim, Germany) were once two major separator companies making non-woven grafted polyethylene/polypropylene separators for Ni/MH battery producers worldwide. In 2006, Scimat was acquired by Freudenberg and, together, they still supply separators for high-performance Ni/MH batteries, especially in the HEV area. In 2015, Freudenberg also became the largest (75%) stockholder for Nippon Vilene—the top nonwoven producer in Japan.

4.1. Scimat

Scimat was founded in 1987, focusing on the production and development of automated controlled separator production process. It was acquired by its main competitor, Freudenberg, in 2006. Between 1985 and 1998, Scimat filed 7 EPSs on a grafting method preparing microporous particles [103], a halopolymer film [104,105], a double-layer (one porous, the other one partially blocking the pores) membrane [106], a grafted sheet composed of copolymer and polyolefin [107], an ultraviolet radiation process to increase the surface hydrophilicity of the separator [108], and a laminate composed of fiber and vinyl monomer on the surface [109]. The Scimat separator is very important in the HEV application.

4.2. Freudenberg

Freudenberg was founded in 1849 to make leather products. Combining the intellectual property and production capacity of past rivals Scimat and Nippon Vilene, Freudenberg became the only separator producer outside of China. Freudenberg filed 8 EPAs from 1993 to 2010, covering the subjects of a non-woven hydrophilic separator material [110], a grafting process making separator material from polymers [111], an addition of alkaline hydroxide or carbonate in the separator before use [112], a separator material containing a titrimetrically determined ammonia binding property [113], a chemically active separator material with an alkaline electrolyte [114], a bi-layer separator with different fiber diameters [115], a core-shell duel-component fiber as a separator [116], and a separator material with coating to enhance its puncturing resistance [117]. Together with Eveready Battery Company (St. Louis, MO, USA), Freudenberg filed an EPA on a cell design using a separator less than 0.15 mm thick in the dry state [118] (Figure 10).

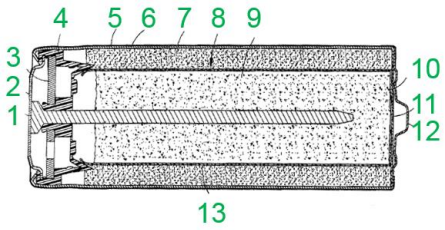


Figure 10. Schematic of a cylindrical cell design from Eveready/Freudenberg. The parts are 1: negative current collector, 2: negative terminal, 3: metal plate, 4: polymer seal, 5: metalized plastic film, 6: steel can, 7: tubular positive electrode, 8: separator, 9: negative electrode, 10: bottom of negative electrode, 11: bottom of folding cylinder, 12: positive terminal, and 13: folding can [118].

5. Other Companies

In addition to the companies mentioned above, which are (or were) involved directly with the manufacturing of Ni/MH batteries, there are other companies and universities in Europe that occasionally applied for EPAs in the Ni/MH battery field. In battery design, Deutsche Automobilgesellschaft (now a subsidiary of Daimler AG, Stuttgart, Germany) filed 3 EPAs on a prismatic cell [119] with the negative electrode covered by polytetrafluoroethylene (PTFE) threads [120], and a bi-polar design of electrode stacking [121].

In the $Ni(OH)_2$ area, the University of Southampton (Southampton, UK) filed one EPA [122] and two PCTs [123,124] on the subject of a porous positive electrode active material with regularly spaced pores. In the MH alloy area, Höganäs AB (Höganäs, Sweden) filed two EPAs on an organically modified silicic acid coating for MH alloys [125], and MH alloys prepared by gas atomization with a unique dendritic microstructure [126]. Fraunhofer-Gesellschaft (Munich, Germany) also filed an EPA on an organically modified silicic acid polycondensated coating for MH alloys [127].

In the charging algorithm and devices, Peugeot (Paris, France) presented a method relating to inner pressure to the state-of-charge (SOC) [128]; Friwo Geräetebau (Ostbevern, Germany) patented a temperature sensor equipment charging device [129]; SGS Thomson Microelectronics (Geneva, Switzerland) disclosed a $-\Delta V$ cut-off method [130,131]; and Solarc Innovative Solarprodukte (Berlin, Germany) disclosed a solar-cell compatible battery charging unit [132].

EU countries are very concerned about battery recycling activities [133]. A special program, COLABATS, was initiated in Europe in 2013 to conduct the development of recycling technology for Ni/MH and Li-ion batteries [134]. In terms of battery recycling, Umicore (Brussels, Belgium) patented a smelting process to reclaim Ni and Co [135]; Montanuniversität Leoben (Leoben, Austria) disclosed a method of reclaiming rare earth metals by using a non-oxidizing acid [136]; Akkuser Ltd. (Nivala, Finland) has a Patent issued on the separation of spent batteries by chopping or crushing followed by a refining or smelting process [137]; CT Umwelttechnik AG (Winterthur, Switzerland) patented a recycling process for the mixture of various kind of batteries [138]; Titalyse S.A. (Croix-de-Rozon, Switzerland) patented an apparatus to sort spent batteries with different size and shape by an optical camera with imaging software [139]; Enviro EC AG (Zug, Switzerland) published a method of reclaiming MH from spent batteries by magnetic separation [140]; SNAM (Saint-Quentin-Fallavier, France) also reclaimed MH by spraying surfactant and later sedimentation [141]; and Eco Recycling SRL (Rome, Italy) reported a physical/chemical combinational method to reclaim Cu and plastic materials [142]. Additionally, French Alternative Energies and Atomic Energy Commission (Gif-sur-Yvette, France) filed a PCT on a process to reclaim rare earth elements, specifically Ni, Co, Mn, and Fe, by selective salt precipitation from a mixture of acid solution for spent Ni/MH battery [143]. Finally, only one EPA was found for the application of Ni/MH batteries, a low self-discharge battery used in soap dispensers filed by Vivian Blick [144].

6. Comparisons to Other Countries

The development of MH alloy and associated Ni/MH battery started from European companies, such as Battelle, Philips, Benz (Stuttgart, Germany) and followed by the successful commercialization by American and Japanese companies [145]. Now the Ni/MH batteries are mainly fabricated in China and Japan. Throughout the years, many important patents were filed in these four areas/countries, namely EU, USA, Japan, and China. The nature of these patent applications is compared in Table 2. Compared to other area/countries, European companies filed the fewest patents (application) in the Ni/MH field. However, their patents in areas such as button cells (owned by Varta Micronbattery), bi-polar design (owned by Nilar), and separators (owned by SciMat and now Freudenberg) are all very important. USA has the largest consumer market in the world and most of its patents related to Ni/MH batteries were filed by Japanese and European companies.

Table 2. Comparison of numbers of patents (applications) filed in EU, USA, Japan, and China and number of associated companies (institutes). * The number of companies used in USA only counts for American companies (institutes).

Country	EU	USA	Japan	China
Number of patents (application) related to Ni/MH	126	371	275	692
Number of patents in negative electrode	22	58	128	136
Number of patents in positive electrode	18	54	27	83
Number of patents in manufacturing technology	10	33	28	104
Number of companies (institutes) filed patents	29	18 *	46	99

7. Conclusions

A total of 126 patent applications related to Ni/MH batteries and filed by European companies to the European Patent Office were reviewed. The scope covers metal hydride formulations and processing, nickel hydroxide preparation, electrode fabrication, cell design, charging method, and recycling. Together with reviews of patents (applications) in the Ni/MH field filed in the USA, Japan, and China, this work completes our studies on the intellectual properties in this field. Compared to companies (institutes) in other countries, European companies started the Ni/MH battery, but were not as active in the development of the production technology. They still made significant contributions in the separator and prismatic cell areas.

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

Abbreviations

The following abbreviations are used in this manuscript:

Ni/MH	Nickel/metal hydride
HEV	Hybrid electric vehicle
MH	Metal hydride
WIPO	World Intellectual Property Organization
EPA	European Patent Application
EV	Electric vehicle
PCT	Patent Cooperation Treaty
PTFE	Polytetrafluoroethylene
SOC	State-of-charge

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