ELSEVIER

Journal of Alloys and Compounds 253–254 (1997) 635–641

Journal of
ALLOYS
AND COMPOUNDS

The state of research and development for applications of metal hydrides in Japan

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Abstract

This paper outlines Japan’s current activities in the research and development (R&D) and commercialization of metal hydrides for applications in hydride electrodes and batteries, hydrogen storage, hydrogen purification and effective utilization of thermal energy. Representative and notable activities and technologies promoted by private corporations and national projects are briefly covered.

Keywords: Metal hydride; Hydrogen storage alloy; Hydride battery; Hydride electrode; Hydrogen purification

1. Introduction

Metal hydrides (MHs) which reversibly absorb and desorb hydrogen at ambient temperature and pressure, so-called ‘‘hydrogen storage alloys’’, are regarded as important materials for solving energy and environmental issues. In Japan, the research and development (R&D) of metal hydrides and their applications has been extensively studied since the early 1970s. In recent years, R&D activities have intensified by virtue of the commercial success of nickel–metal hydride (Ni–MH) batteries and the promotion of various MH-related projects by the government and private corporations. For example, a non-profit association (The Research Association for the Development and Application of Metal–Hydrogen Systems, Japan: RADAMH), which presently consists of over fifty private members and sixty representing various corporations, has existed since 1991 for the purpose of promoting information exchange and social affairs among researchers and engineers in this field.

As listed in Table 1, applications of MHs extend in diverse technological fields [1–9]. Although good scientific and technical progress has been made through intensive R&D efforts, most of the applications until recently remained at the stage of ‘‘proof-of-principle’’ or at a prototype level due to various technical and/or economic difficulties. Only an aircraft fire detector using titanium hydride [3,5] and hydrogenation grinding methods for metal ingots such as titanium and zirconium were in practical use before the late 1980s. After 1990, however, commercial breakthroughs for MHs have been made in at least four fields: Ni–MH batteries, [4], a cathode for chlor-alkali electrolyzers [6], a hydrogen purifier for hydrogen-cooled power generators [7] and a hydrogenation-decomposition–desorption–recombination process for preparing Nd–Fe–B magnetic powders [8].

In this paper, recent advances in hydride technologies at the R&D and commercial levels in Japan are described

Table 1
Applications of metal hydrides

·Hydrogen storage and transport (stationary H_2 storage containers, H_2 carriers, H_2 tanks for H_2-fuelled vehicles)
·Hydrogen separation and purification (H_2 separators, H_2 recovery units, H_2 purification units, high-purity H_2 generators)
·Hydrogen fixation (H_2 getters, moderators and shielding materials in nuclear reactors)
·Separation of hydrogen isotopes
·Hydride electrodes (Ni–MH batteries, MH–air battery, hydrogen electrodes for electrolyzers and fuel cells)
·Effective utilization of thermal energy (heat storage, heat transport, heat pumps, refrigerators)
·Temperature and pressure sensors (fire detectors, thermometers)
·Thermal and mechanical energy conversion (H_2 compressors, H_2 liquefiers, actuators, hydride engines)
·Metallurgical processing (hydrogenation grinding, HDDR process for producing Nb–Fe–B magnet powder)
·Catalysts
·Switchable optical devices

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0925-8388/97/$17.00
PII S0925-8388(96)03090-3

with emphases on hydride electrodes, hydrogen storage, hydrogen purification and effective utilization of thermal energy.

2. Nickel–metal hydride (Ni–MH) batteries

2.1. Small-scale Ni–MH batteries

Sealed Ni–MH batteries of cylindrical and rectangular shape in various sizes are currently produced by several battery companies (Matsushita, Sanyo, Toshiba, Yuasa, GS-SAFT, Hitachi–Maxcell and Furukawa). Fig. 1 shows the recent trend in annual production of Ni–MH batteries in comparison with that of other primary and secondary types. Owing to the increasing demand for secondary batteries with high performance and environmental compatibility as a power source of portable electronic appliances, the production of sealed Ni–MH batteries has risen rapidly since commercialization in 1989. The yearly production in 1995 reached 310 million units or 95 billion yen in value [10]. For most of the batteries, $MmNi_5$ (Mm=mischmetal)-based alloys, with Ni partially substituted by Co, Al and Mn, are exclusively utilized. Their compositions are finely adjusted according to both the manufacturing processes and the desired performance characteristics of the batteries. The amount of alloys produced for the negative electrodes in 1995 was estimated to be about 3000 tons and to be valued at about 9 billion yen.

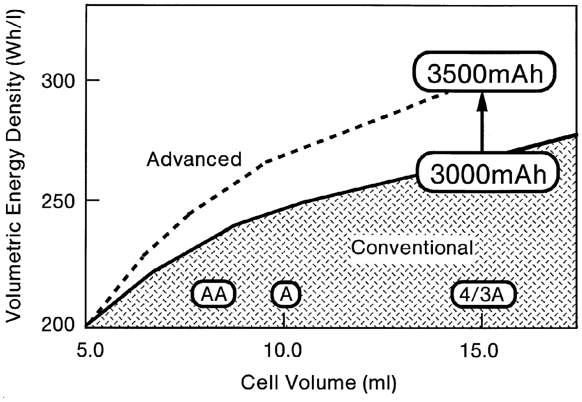

Fig. 2. Relation between volumetric energy density and cell volume for cylindrical Ni–MH batteries.

The main R&D aim for commercial Ni–MH batteries is directed toward higher energy density and lower price, and is highly competitive. Although standard Ni–MH batteries of AA size have a nominal capacity of 1100 mAh and specific energy densities of 180 Wh l^{-1} and 50 Wh kg^{-1}, improved types now reach 1300 mAh, 200 Wh l^{-1} and 60 Wh kg^{-1}, and are under commercial production. As shown in Fig. 2, a recent highlight is the development of highly advanced Ni–MH batteries with a 3500 mAh nominal capacity and energy densities as high as 300 Wh l^{-1} and 80 Wh kg^{-1} for the 4/3A (Long A) size by Matsushita [11], Sanyo [12] and Yuasa [13]. These advanced batteries, with a larger volumetric energy density than commercial lithium ion batteries, are being manufactured for use in personal computers of notebook types. It is claimed that this remarkable increase in energy density is due to several technical advances:

(i) Increase of both packing and utilization ratios of $Ni(OH)_2$ powders by effective use of conductive agents and by improvement of the porous electrode substrate in morphological design and mechanical strength.

(ii) Increase of the durability of $MmNi_5$-based alloys by control of their composition and metallurgical microstructures, and by introduction of mixing additives.

(iii) Development of a thin, corrosion-resistant and wettable separator by use of fine fibers and hydrophilisation treatment.

(iv) Improvement of battery-manufacturing techniques

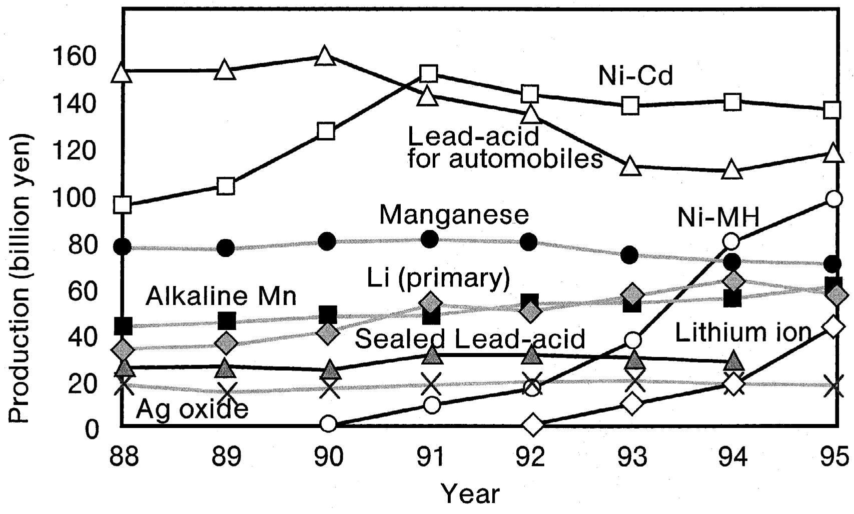

Fig. 1. Trend of battery production in Japan.

for the decrease of void space and the use of thinner and lighter components.

Concerning cost reduction, the average price per unit decreased from 350 yen in 1993 to 300 yen in 1995. This is primarily due to higher production levels and the cost lowering of constituent materials.

2.2. Large-scale Ni–MH batteries

2.2.1. Applications to photovoltaic power generation systems

The R&D of large Ni–MH batteries was initiated in 1989 for their use as an auxiliary power source for photovoltaic (PV) power generation systems in the Solar Energy R&D program of Japan's Sunshine Project [14]. The primary aim of this four-year project promoted by NEDO (New Energy and Industrial Technology Development Organization) was to develop two kinds of Ni–MH batteries of a 12 V–100 Ah class for reduction of electric peak demand and absorption of output variations of photovoltaic cells in utility-connected PV systems, and for a back-up power source in stand-alone types.

One module battery named ''Long-cycle'' type with target parameters of a 6000 cycle life and charge–discharge currents of 1 C rate at the depth of discharge (DOD) of 80% and the other battery named ''High-rate'' type with a 3000 cycle life and a 2 C rate were fabricated by Matsushita and Sanyo, respectively, in 1992. ONRI (Osaka National Research Institute) supported the project in analyzing and evaluating the performance of various $MmNi_5$-based alloys, their electrodes and batteries. According to a durability test recently finished at the Central Research Institute of Electric Power Industry, the high-rate battery retained 80% of its initial capacity after 3000 cycles. The test for the long-cycle battery is still underway.

The second phase of this project (1993–1996) involves ONRI, NEDO and Matsushita. The main R&D goals are suppression of temperature rise of the battery during high-rate charge and discharge operations, improvement of performance of the alloys and battery at higher operating temperatures ($T>40$ °C), and lowering of manufacturing cost of the battery.

2.2.2. Applications to electric vehicles

The use of Ni–MH batteries for electric vehicles (EVs) has grown rapidly since the early exploration at ONRI [15]. As far as the authors know, several prototype EVs powered by Ni–MH batteries have been developed, and four battery makers (Matsushita [16,17], Yuasa [18,19], GS [20], Furukawa [21]) and one automobile maker (Toyota Automatic Loom Work [18,19]) are currently working on batteries using $MmNi_5$-based alloys. This is part of a general response to legislation against air pollution by exhaust gases from automobiles and to suitable performance characteristics of Ni–MH battery for EVs, as comparatively shown in Table 2. For EV applications, the primary requirements are a large energy density, high-temperature performance in terms of durability and charging efficiency and a high rate capability. It was recently reported that two 12 V-module batteries have been developed with energy densities of over 65 Wh kg^{-1} and 155 Wh l^{-1}, good charge–discharge characteristics after 500 cycles, and a specific power of 200 W kg^{-1} [16,18].

In the Scandinavian Electric Car Rally held in August of 1995, a Toyota RAV4 EV equipped with a stacked Ni–MH battery of the 30 kWh class (24 units of the 12 V–100 Ah module) produced by Matsushita won the first prize. According to newspapers, Toyota has commercialized an EV (RAV4L) powered by a Ni–MH battery, with a driving range of 215 km (10–15 mode) per charge at the price of 4.95 million yen in Japan in autumn 1996 (Fig. 3) [22]. It has also been announced that Honda plans to market a similar vehicle, with a range of over 200 km per charge, in California by early spring 1997 [23].

2.3. Hydrogen storage alloys for battery applications

$MmNi_5$-based alloys are still under development to improve their electrode performances further and to reduce the content of Co. Their electrochemical characteristics can

Table 2
Typical performance of various stacked batteries for electric vehicles

Battery type	Cell voltage (V)	Energy density		Power density (W kg^{-1})	Cycle life (cycle)	Self discharge[a] (%)	Operating temperature (°C)	Reliability safety[b]
		(Wh dm^{-3})	(Wh kg^{-1})					
Ni/MH	1.2	150	70	250	>1000	15	−20~40	○
Li ion	3.6	160	100	300	>1000	10	−20~40	?
Ni/Cd	1.2	120	60	250	>1000	15	−20~45	●
Pb/PbO_2	2.0	80	30	150	300~500	5	−10~40	●
Zn/Br_2	1.7	50	60	100	500	7	0~60	?
Na/S	1.8	120	80	90	500	0	>330	?
Ni/Zn	1.5	130	75	130	250	15	−20~40	?
Li/FeS	1.5	100	100	100	300	0	450	?

[a] Percent per month at 20 °C.
[b] ●: good, ○: satisfactory.

Fig. 3. The electric vehicle (TOYOTA RAV4L) powered by a Ni–MH battery, which is scheduled for commercialization in the autumn of 1996 [22]. Curb weight: 1410 kg, Maximum speed: 125 km h^{-1}, running distance per charge: 215 km (10–15 mode).

be improved by controlling better their compositions and metallurgical microstructures and by surface treatment with alkaline solutions and additives. The reduction of Co content has usually resulted in a short charge–discharge cycle and remains a main unsolved problem. High temperature testing is becoming mandatory in relation to MHs for large Ni–MH batteries, because of their greater thermal inertia relative to small batteries, especially at high charge and discharge rates. It is expected that effects of partially substituted elements on electrode performance, especially on durability, become more clear through this type of testing.

Other hydrogen storage alloys with a large hydrogen capacity, e.g., Zr–Ni–Mn–V [24,25], Ti–V–Ni [26] and Mg–Ni [27] systems, are also under investigation metallurgically and electrochemically at both the fundamental and applied levels.

2.4. Standardization of testing methods of MHs for use in batteries

It is not always easy to obtain reproducible and reliable data of electrochemical properties of MHs, especially for MHs which easily passivate. Discharge capacities and durabilities of MH electrodes depend not only on properties of MHs themselves but also on electrode preparation. For these reasons, research to establish and standardize testing methods for a wide range of MHs has been underway at NMC (New Material Center) since 1992 [28]. Comparative tests for the influence of preparation and measuring methods on discharge capacity are ongoing on $LaNi_5$, Ti–Ni and Zr–Ni-based alloys via inter-laboratory sharing of alloys, electrode substrates, conductive materials and electrodes. Analytical investigations on factors which cause variations in the measured capacities are also assessed, including testing conditions and methods, storage conditions of alloys, surface treatments, metallurgical microstructures, surfaces states, etc.

2.5. Recycling of Ni–MH batteries

Rare metals such as Ni, Co, Mm, Ti, Zr and V are the main constituents of Ni–MH batteries. Therefore, it is anticipated that recycling of spent Ni–MH batteries becomes inevitable not only for environmental protection but also for effective utilization of rare metal resources and cost lowering of raw materials. In RADAMH, a feasibility study for the recycling of both small and large Ni–MH batteries has been conducted from technical and economical points of view. The material and methods under investigation consist of collection, separation and dismantling of spent Ni–MH batteries, and of mechanical and metallurgical processes for selective separation of constituent metals (especially for that of Ni, Co and Mm) and related compounds which are re-usable as raw materials. According to preliminary findings, small Ni–MH batteries using $MmNi_5$-based alloys can be recycled at a loss, whereas the large ones yield profit [29]. The loss is mostly due to the expense for collection of spent batteries and the relatively small scale of the operation.

3. A highly durable cathode containing MH for chlor-alkali electrolyzers

A cathode coated with Raney-nickel catalyst and MH which operates at a low hydrogen overvoltage and is very durable has been developed for chlor-alkali electrolyzers by Asahi Glass Co. Ltd. [6,30], and is in industrial use. The cathode, with a porous composite-coating layer of Raney-nickel alloy and MH powders on a Ni matrix, is prepared by a special plating method. As seen in Fig. 4, incorporation of the MH powder helps to maintain the cathode near the reversible potential of the hydrogen electrode during power interruption shutdowns and under short-circuit conditions, and avoids oxidative deterioration

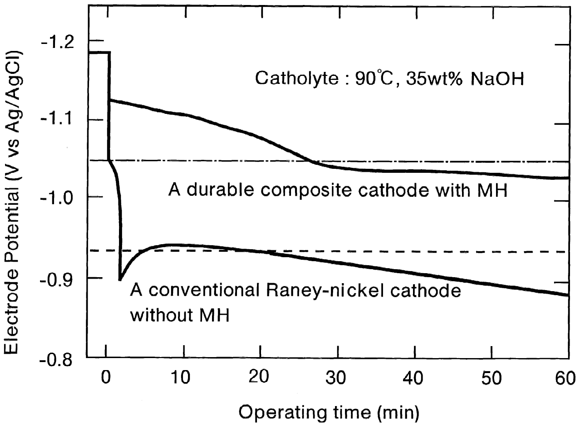

Fig. 4. Potential stability of the cathodes with and without metal hydride (MH) powder kept at short-circuit conditions [30]. – · –: Hydrogen electrode potential; — — — Oxidation potential of Ni to $Ni(OH)_2$.

of the catalyst for electrolyzers using ion-exchange membranes.

4. Storage and transport of hydrogen in the form of MHs

4.1. National projects

The following projects are promoted in Japan's New Sunshine Project. Osaka National Research Institute (ONRI) and National Institute of Materials and Chemical Research (NIMC) have been conducting the R&D of storage and transport of hydrogen since 1974 [1,31]. Their recent activities are directed toward the search for new MHs and their characterization in order to advance this technology further [31]. Both national laboratories also support the promotion of the following national projects in a form of collaboration.

Under rising concerns about global energy and environment problems, a large-scale and long term R&D project on international clean energy systems utilizing hydrogen was initiated in 1993 [32,33]. Known as the WE-NET (World Energy Network) project, this includes R&D on storage and transport of hydrogen in the form of MHs. After assessing the present state of R&D in this field and conducting feasibility studies on the applicability of MHs to hydrogen energy systems during 1993–1996 [33], the participants will continue research into new MHs suitable for stationary hydrogen storage systems and tanks for hydrogen fuelled cars starting in 1997. The targeted parameters are as follows;

1. Effective hydrogen storage capacity of more than 3 wt.%.
2. Hydrogen desorption temperature below 100 °C.
3. Operating hydrogen pressure below 1 MPa.
4. Maintenance of over 90% of the initial capacity after 5000 cycles.

In addition, R&D of hydrogen storage using MHs is performed as a part of the study on a system to recycle CO_2 (i.e. combustion of fossil fuels) to useful chemical substances by Research Institute of Innovative Technologies for the Earth (RITE) [34,35]. The main theme is development of MHs durable to moisture and oxygen contaminants in hydrogen produced by water electrolysis.

4.2. R&D in the private sector

In recent years, R&D of storage and transport of hydrogen by MHs has been conducted by private corporations such as Materials & Energy Research Institute Tokyo, and others. For example, a portable H_2 supply based on $Mm_{0.82}Y_{0.18}Ni_{4.95}Mn_{0.05}$ alloys, storing gaseous hydrogen of 0.4 N m_3 and weighing about 4.5 kg, has been developed for a fuel cell system by Sanyo Electric Co. Ltd. [36] and is commercially available.

After successive development of two hydrogen-fuelled model cars equipped with a rotary engine and an MH tank, Mazda have just developed a similar car of a practical type with an improved MH tank [37]. The tank is made of $MmNi_5$-based alloy (350 kg) and allows the car to drive about 200 km per hydrogen charge. The car is undergoing its running tests on public roads.

5. Hydrogen purification

In the field of applications of MHs to separation and purification of hydrogen, a good result is obtained with respect to a hydrogen purifier for hydrogen-cooled power generators [7]. The R&D of the purification unit, together with two MH tanks, a dehumidifier and a deoxydation unit, was done collaboratively by three companies, viz. Japan Steel Works (JSW), Kansai Electric Power and Mitsubishi Electric, with the aim to decrease windage loss of the generator due to the contamination of the hydrogen coolant. As shown in Fig. 5, a purification unit using $Ca_{0.85}Mm_{0.15}Ni_{4.85}Al_{0.15}$ alloy of 120 kg and having a processing capacity of 24 N m^3 h^{-1} established a hydrogen purity of more than 99.9% and a durability of 10 000 cycle in H_2 charge and discharge in its verification test for a 60 MW-class generator. Commercial operation is imminent.

6. Effective utilization of thermal energy

The R&D of heat storage units, heat pumps, refrigerators and heat transport systems using MHs were actively performed in the 1980s, but commercial success for these technologies has not been achieved yet. It is considered that the most likely application of MHs in the area of effective utilization of thermal energy is refrigeration systems. Recently, Sanyo has developed a new

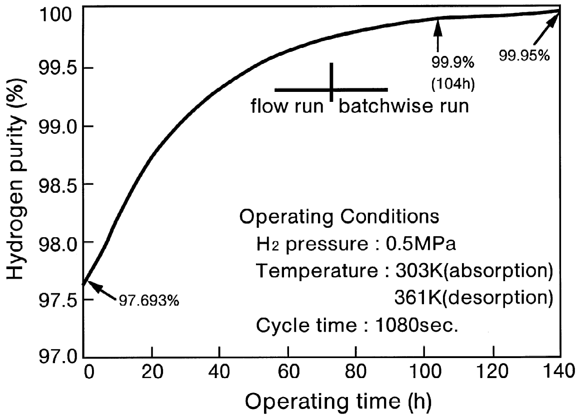

Fig. 5. Variation of hydrogen purity in a power generator with operating time [7].

refrigeration system, to generate refrigeration heat below −20 °C, with the support of NEDO as a part of Japan's New Sunshine Project [38]. The generator uses a $LaNi_5$-based alloy and has a coefficient of performance greater than 0.4 by use of driving heat of about 150 °C. Refrigeration systems, which use $TiCr_2$-based alloys and are capable of reaching temperature levels between −20 and −40 °C, are now undergoing performance tests, and are planned to be commercialized cooperatively by JSW and refrigerator makers [39].

A national R&D project named Eco-Energy City Project was started in 1993 under Japan's New Sunshine Project, aiming at developing technologies to make effective use of waste heat in industrial areas [40,41]. In the project, the R&D of MHs and their applications relating to transport and upgrading of waste heat has been re-intensified. At present, the applications of MHs to effective utilization of thermal energy are not likely to be in practical use. Further technical advances in this field may result from this project.

7. Conclusion

The R&D for applications of MHs is under intensive exploration in Japan. The areas of hydride batteries, hydrogen storage and effective heat utilization are prominent. The annual production quantity of small-size Ni–MH batteries has rapidly increased, reaching 310 million units in 1995, and the commercialization of large-scale Ni–MH batteries for use in electric vehicles is just around the corner. In addition, commercial success has been achieved in their applications as a cathode for chlor-alkali electrolyzers, a hydrogen purification unit for hydrogen-cooled power generators, and an HDDR process for preparing Nd–Fe–B-based magnet powders. However, traditional $LaNi_5$-based alloys are mainly exploited in commercial systems and those at a demonstration stage. It is sure that small Ni–MH batteries, which is the main success to date for applications of MHs, will receive strong competition from lithium ion batteries with superior weight energy densities. Further advances in the applications of MHs will depend on the development of new alloys consisting of cheaper elements and having a larger hydrogen capacity. A greater diversity of applications, including new fields, will be important for the future market prospects of MH-related systems.

Acknowledgments

The authors would like to thank Japanese researchers and engineers who have served to provide much of the material and information presented in this review. We are also grateful to Dr. J. Highfield for kindly proof-reading the manuscript.

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