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REVIEWS IN APPLIED ELECTROCHEMSTRY NUMBER 67

RVC as new carbon material for batteries

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Abstract Results of the last 10 years of our investigations on the use of reticulated vitreous carbon (RVC$^{®}$) in various types of cells show that this kind of porous conductive glassy carbon can be successfully used as a matrix for active mass and as a current collector in many primary and secondary batteries. This paper focuses mainly on the use of RVC$^{®}$ modified with metals like Pt, Rh, Pd, Ni, Pb and their oxides in construction of the following types of cells: (1) zinc-carbon primary and secondary batteries, (2) rechargeable cell systems with NiOOH cathode, (3) electrochemical capacitors with Pd and Pd–Rh alloys, and (4) lead acid battery. The most important improvement in performance has been achieved with lead-acid and zinc-carbon batteries. Especially for the modified zinc-carbon battery, which has been constructed in the popular AA format, manufactured on a small (laboratory) scale, and tested according to the IEC norms as a battery for commercial use, the capacity has been higher by ca. 20% compared with the batteries available on the market. Also, results obtained for thin layers of Pd and their alloys deposited on RVC$^{®}$ are promising in terms of the application of these kinds of electrodes in electrochemical capacitors. The pseudocapacitance of a Pd–Rh/RVC$^{®}$ composition may reach 540 F g^{-1}.

Keywords RVC$^{®}$ · Batteries · Current collector · Electrochemical power sources

1 Introduction

A porous, conductive glassy carbon, such as reticulated vitreous carbon (RVC$^{®}$), can be used as a matrix for active mass and a current collector in many “classical” primary and secondary batteries. As shown in Fig. 1 depicting a SEM image of RVC$^{®}$, this type of carbon is an open-pore material (20 ppi—pores per inch).

Table 1 collects the most important physico-chemical parameters of RVC [1]. Its low density, high void volume and low resistivity make it a desirable electron-conductive foam material for batteries.

Although RVC$^{®}$ was developed 30 years ago [2], it is currently not as wide-spread as expected, especially in electrochemical applications. In fact, only 200 papers on the subject have been published over the years, i.e. about six papers per year. Following its use as an electrode material in half-cell studies [3–5], RVC$^{®}$ has been tested in full cells [5]. In 1984, it was demonstrated [6] that RVC$^{®}$ can be covered electrochemically with a very thin layer of platinum, or platinum-rhodium alloy. Such an electrode exhibited the behaviour of a solid metal electrode.

Figure 2 shows typical cyclic voltammograms (CVs) of a pure platinum electrode (curve 1), platinum deposited on RVC$^{®}$ (curve 2) and a pure RVC$^{®}$ matrix (curve 3). The behavior of the Pt/RVC$^{®}$ electrode is very similar to the behaviour of the pure metal. Only the currents responsible for double layer charging/discharging are higher in comparison with pure platinum due to an additional charging-discharging current of the uncovered carbon support. Since the thickness of platinum deposited on RVC$^{®}$ is equal to a

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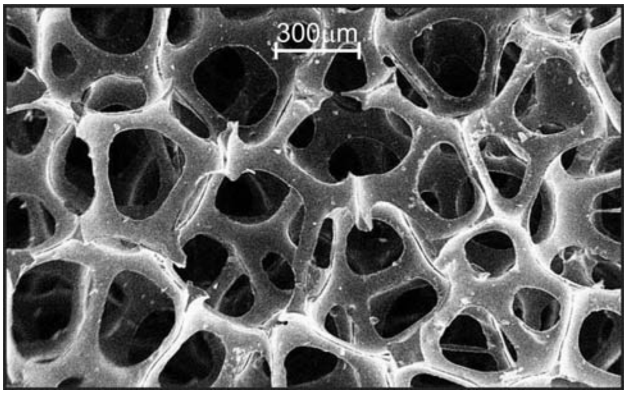

Fig. 1 SEM image of RVC® (20 ppi—pores per inch)

Table 1 Physico-chemical parameters of RVC® [1]

Physical properties	Values	
	RVC	GC
Density/kg m^{-3}	48	1650
Void volume/%	90–97%	0
Bulk resistivity/Ω cm	0.005	0.001
Thermal conductivity/cal cm °C w 10^{-4} tor	0.08–1200	0.01–0.02

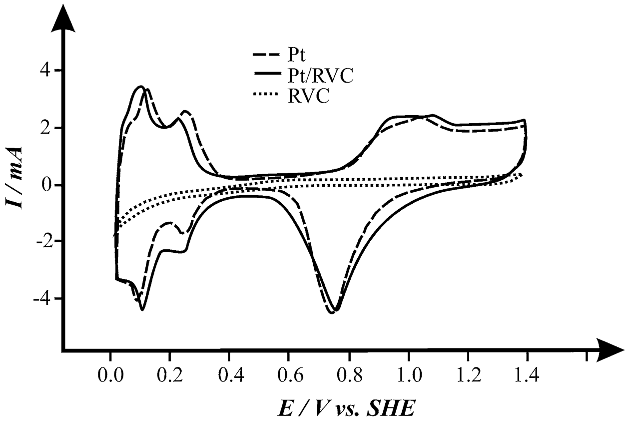

Fig. 2 Cyclic voltammograms for pure RVC®, pure Pt and Pt/RVC® electrodes in 0.5 M H_2SO_4 at sweep rate of 50 mV s^{-1}

few monolayers of Pt, this result indicates that platinum is deposited on the carbon surface irregularly as a few hundred nanometer particles as shown in the SEM image depicted in Fig. 3 [7].

The surface of carbon can be covered with particles of diameters down to ca. 100–200 nm. The size of particles depends on the procedure and the parameters of the electrodeposition [7]. Similar results have been obtained for platinum–rhodium alloys [8].

A few years after the introduction of RVC®, this carbon material was used for the construction of microbiological cells [9–11]. These cells were classified as fuel cells, although an oxygen cathode was not used. In this invention, porous conducting carbon played the role of carrier of the electrode reagent and of current collector for electrons. During cell operation, in the pores of the carbon material, glucose or galactose were oxidized by microorganisms (*Proteus vulgaris, Escherichia coli*) with easy access of electrons to the surface of the carbon electrode through mediators (thionine). The cathode half-cell consisted of a solution of potassium ferricyanide in phosphate buffer and a platinum electrode [12, 13]. The efficiency of the reaction in this biological cell was between 30 and 60% depending on the microorganisms.

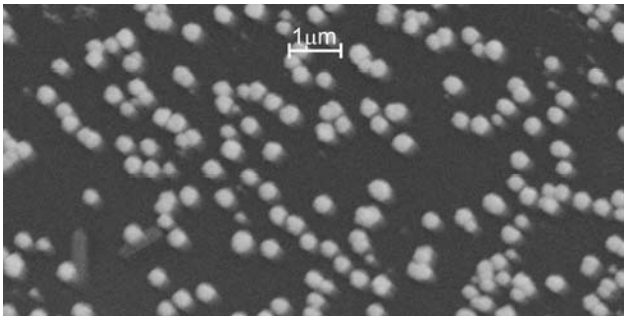

Fig. 3 SEM image of Pt particles deposited on RVC®

RVC® with different porosity has also been used in another type of battery in which bromine in the form of a polymeric salt—poly(diallydimethylammonium) bromine, was used as the electron acceptor [14]. The RVC® was principally a carrier for the cathodic mass. In one version of these batteries, RVC® has been also used as a zinc carrier in the anode [15]. The ease of handling of bromine relative to chlorine and the absence of cooling requirements make these cells more attractive than Zn–Cl_2 cells.

For modification of the RVC® surface [16], polyaniline (PANI) has been used. Such electrodes were employed as cathodes in model systems of non-aqueous lithium rechargeable batteries e.g., Li/$LiClO_4$-propylene carbonate/PANI-poly(p-styrene sulfonic acid)-RVC® [17, 18]. The durability of this cell was characterized by over 30 stable charging-discharging cycles. The system Zn/$ZnCl_2$, NH_4Cl/PANI-RVC®, with an open circuit voltage (OCV) equal to 1.2 V, has been used as an aqueous secondary battery. The specific capacity of this battery was over 100 mAh g^{-1} [19]. Also, the Pd/PANI/RVC® system has been studied as a material for hydrogen storage [20].

This paper focuses mainly on the use of RVC® modified with metals like Pt, Rh, Pd, Ni, Pb and their oxides in our laboratories, for construction of various types of cells.

2 Results

In our studies we have found that RVC® modified with metals like Pt, Rh, Pd, Ni, and Pb and their oxides can be used as a matrix for an active mass and a current collector in many primary and secondary batteries. Some details of our results were described earlier [5]. In our laboratories, RVC® has been used in the construction of the following cells: zinc–carbon primary and rechargeable batteries, rechargeable cell systems with the NiOOH cathode, electrochemical capacitors with Pd as well as Pd–Rh alloys, and lead acid batteries.

2.1 Zinc–carbon primary and secondary batteries

The discharge mechanism of the zinc–carbon (manganese dioxide) cell is complex and there are products of the electrode reactions, which are insoluble. The insolubility of these compounds causes the resistance of the cell to increase with time of discharging. This is one of the main reasons of the voltage drop in zinc–carbon cells.

In our new construction of the zinc–carbon cell [21–25], the carbon rod (cathodic current collector) is replaced with RVC®. Construction details of standard and of our new zinc–carbon cells are shown in Fig. 4 The RVC® fulfills functions as the current collector and, simultaneously, as the carrier for the cathodic active mass i.e., the mixture of MnO_2, acetylene black, and electrolyte. In the modified cell, each pore of the RVC® acts as a separate half-cell and the average amount of half-cells calculated for a typical AA-size battery is about 1,000. If an unwanted reaction takes place, e.g. precipitation of insoluble compounds, or self discharging, only a limited number of pores of the cathode volume is affected. For this reason, the proposed modification of cathode construction leads to improved cell operation.

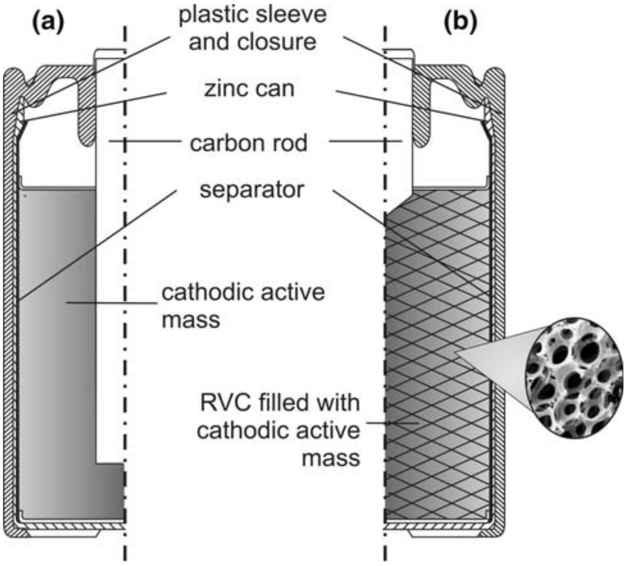

Fig. 4 Cross-section of standard (**a**) and modified (**b**) zinc-carbon batteries

Figure 5 shows discharging curves at $R = 43\ \Omega$ load for the standard zinc–carbon cell (dashed line) and the modified cell with the new cathode design (RVC®/MnO_2, solid line). The modified batteries were assembled using commercially available elements employed in the zinc–carbon battery industry. Both the standard and the modified cells are shown in the inset to Fig. 5. The results obtained in this test showed that discharging times of the new zinc–manganese dioxide cell (using NH_4Cl electrolyte) can be significantly longer, i.e. about 18%, than those for commercially available higher class batteries using $ZnCl_2$ electrolyte.

Since RVC® is a conductive material, in our battery construction it substitutes not only the carbon rod (the current collector) but also, partially, the conductive additives to MnO_2 (carbon black). It was found that the best results of operation were obtained when the RVC®/MnO_2 cathode system contained only 3% of carbon black in the mixture with MnO_2 vs. a typically used 10% carbon content in the active paste. The use of an open-pore material for current collector and the decrease in the amount of carbon black allows for an increase in the amount of cathodic MnO_2 active mass, responsible for charge storage capacity of the battery. Additional benefits of this new construction are:

- increased storage capacity,
- higher discharging current,
- improved voltage stability during discharging.

To demonstrate that the RVC® modification of the zinc–carbon cell is beneficial for battery performance, the batteries constructed in our laboratory were tested at a

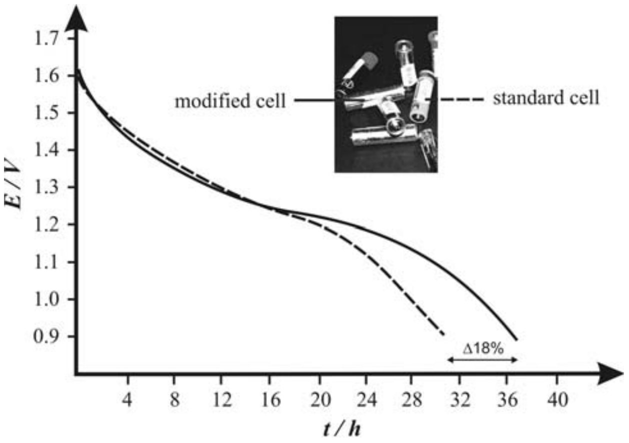

Fig. 5 Continuous discharging curves (43-Ω load) of standard and modified zinc carbon cells (AA size)

certified battery testing laboratory—the Laboratory of Cells and Batteries in Poznan (Poland). The results of these tests confirmed that the discharging times of the new zinc–manganese dioxide cell were ca. 20% longer than those typically observed for commercially available batteries. Figs. 6a–d present the intermittent discharging results simulating the work of various devices such as: portable radio receivers (Fig. 6a), portable tape recorders (Fig. 6b), electric toys (Fig. 6c) and flashlights (Fig. 6d).

In the insets to Figs. 6a–d, the average discharging times typical for our RVC®-modified batteries, the IEC norm and the commercial (heavy duty) HD batteries are shown in a bar diagram. For each application scenario the discharging times were always higher than the IEC norm and were either comparable or higher than those obtained for the higher-class commercial (HD) batteries.

In our work, RVC® was also applied as an anode and cathode mass carrier and current collector in a new rechargeable Zn/RVC®–(MnO_2,C)/RVC® battery with an acidic $ZnSO_4$ electrolyte [26]. The results with this new battery are promising and we expect that its durability measured in terms of charging/discharging cycles will be much higher in comparison with alkaline rechargeable Zn–MnO_2 batteries. Figure 7a shows a scheme of the new construction of a zinc–carbon rechargeable cell based on RVC® as the active mass carrier and electric collector. Preliminary results of the charging/discharging of this battery are shown in Fig. 7b.

2.2 Rechargeable cell systems with NiOOH cathode

The electrochemical behavior of nickel deposited on RVC® has also been investigated in terms of its applicability as a cathode material in nickel–cadmium and nickel–metal hydride batteries [27–30]. It was found that the Ni/RVC® system behaves as solid nickel, sintered nickel or Ni-foam, which are widely used as carriers for nickel hydroxides—the material of positive electrodes in Ni–Cd and Ni–MH rechargeable batteries. Figure 8a–c present cyclic voltammograms of Ni foam, sintered Ni, and Ni deposited on RVC®. The SEM images of Ni deposited on RVC®, as well as sintered Ni and Ni foam, which are commonly used in batteries, are also shown in these figures.

The shapes of the cyclic voltammograms shown in Fig. 8a–c obtained on Ni/RVC®, sintered nickel and on

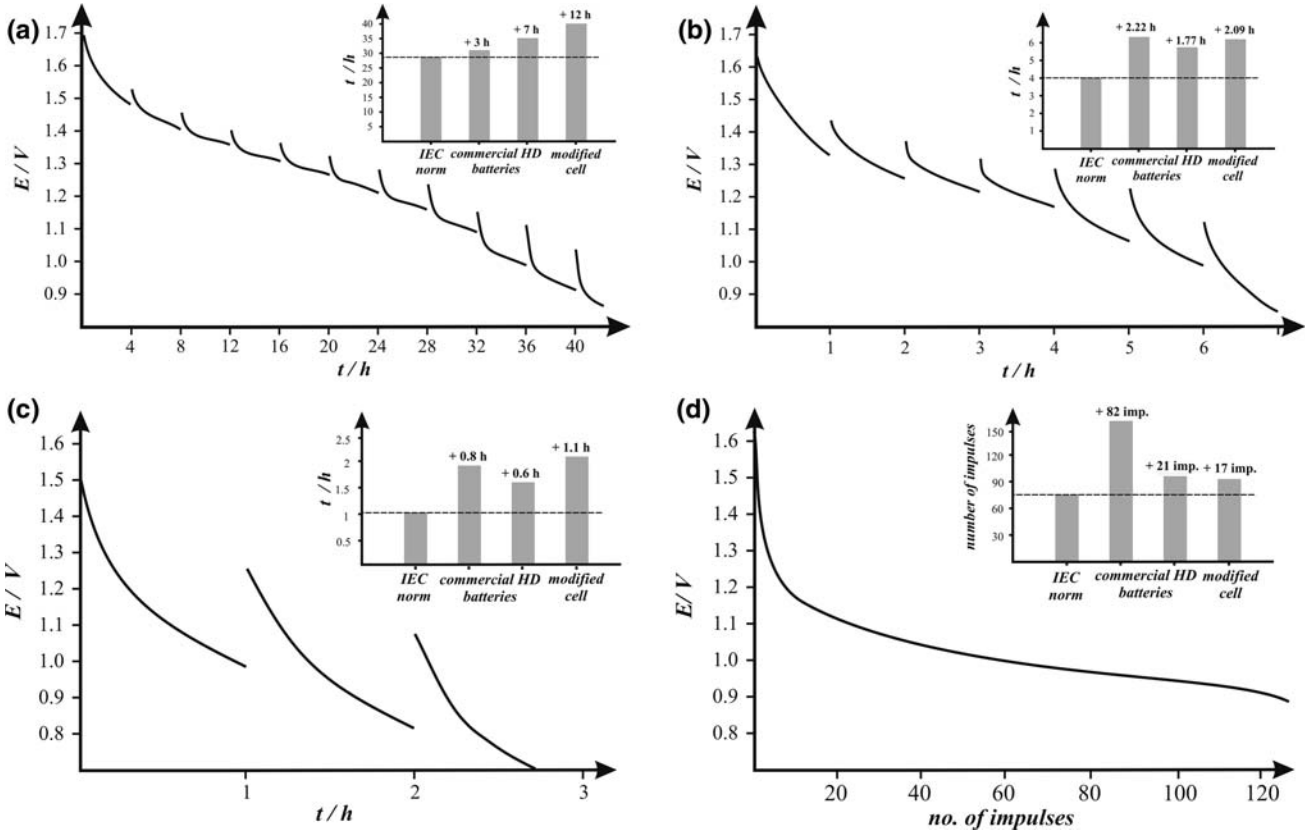

Fig. 6 Intermittent discharge curves of modified batteries: **a** 43 Ω load, 4 h daily (radio receiver mode), **b** 10 Ω load, 1 h daily (walkman mode), **c** 3.9 Ω load, 1 h daily (electric toy mode), and **d** number of discharges impulses at 1.8 Ω load (common photoflash mode). Insets: average discharging results obtained for new and commercial cells

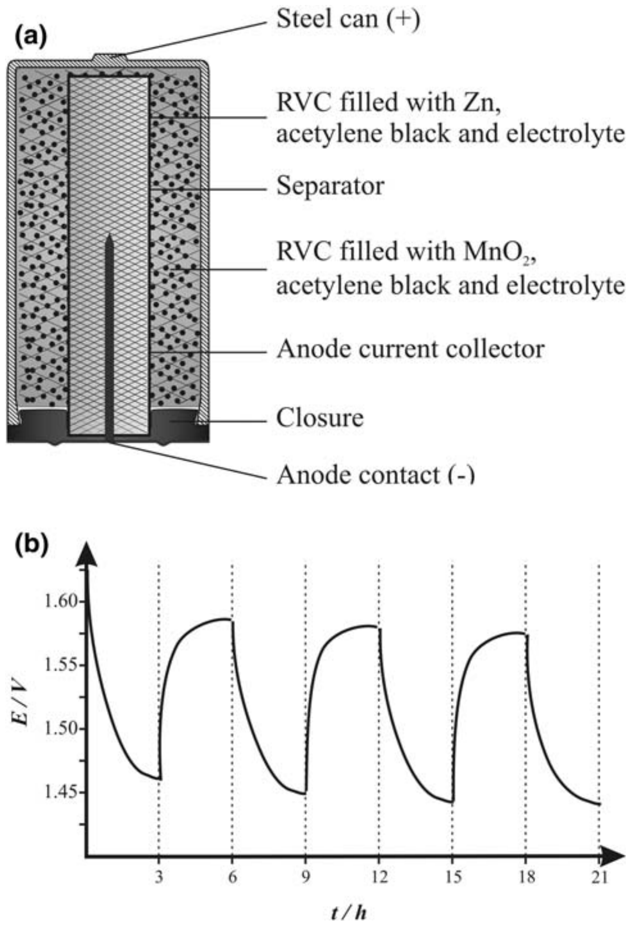

Fig. 7 Scheme of rechargeable zinc-carbon cell with new anode and cathode (**a**). Discharging and charging curves under 30 mA current draw (**b**)

nickel foam electrodes are almost the same as in the case of pure solid Ni electrodes described in the literature [29, 31, 32]. In our experiments, the Ni/RVC® electrode has been used as a carrier for $Ni(OH)_2$ cathodic active material and as a current collector.

The plots for the charging/discharging processes (Fig. 9) for Ni/RVC®-$Ni(OH)_2$/NiOOH and sintered Ni–$Ni(OH)_2$/NiOOH systems are similar to each other and the discharging times are even higher for the modified cell than for the sintered Ni–$Ni(OH)_2$/NiOOH standard one. Therefore, the Ni/RVC® can be considered a promising material for Ni-cathode batteries.

The specific capacity and capacity density calculated for the new cathode is compared with the sintered nickel and the nickel foam systems in Table 2. It can concluded that the batteries provided with Ni/RVC®–NiOOH electrodes should have an approximately two times higher specific capacity (Ah g^{-1}) than the sintered Ni–NiOOH batteries, but worse than the nickel foam electrode. The "foam" nickel has the best specific capacity and capacity density (twice) as a NiOOH on RVC®carrier.

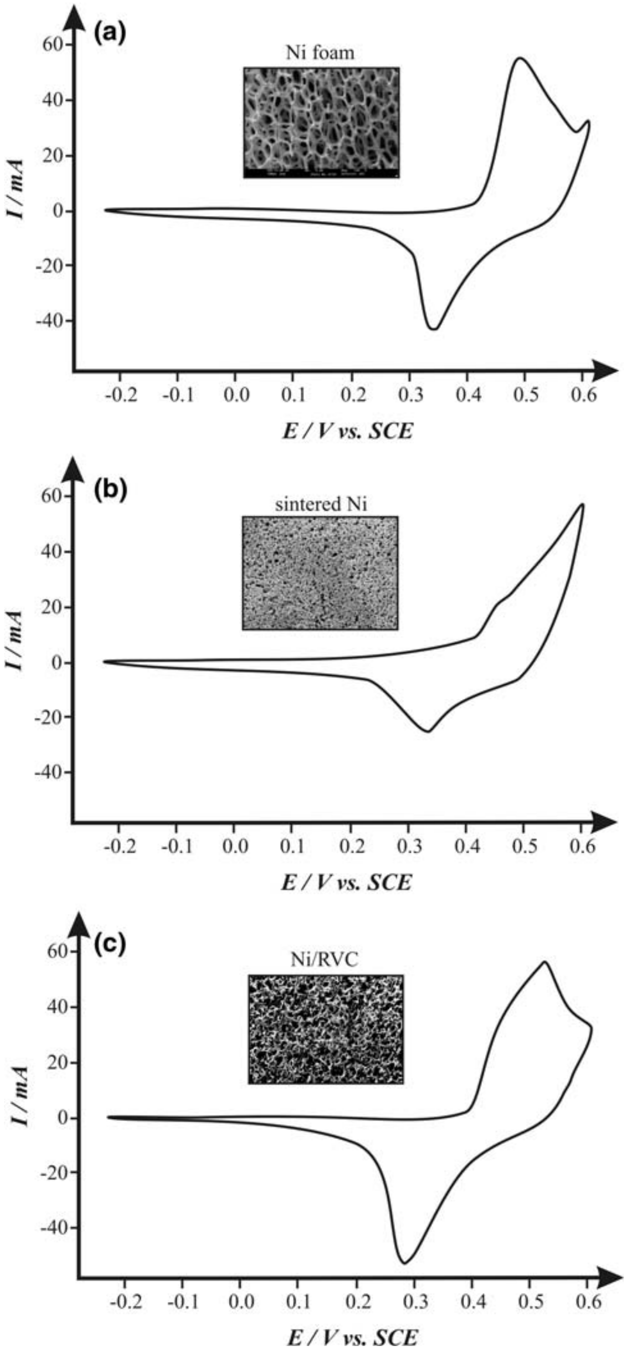

Fig. 8 Typical cyclic voltammogram obtained in 6 M KOH at sweep rate of 100 mV s^{-1} for Ni foam (**a**), sintered Ni (**b**), and Ni deposited on RVC® (**c**). Insets: SEM images (of respective Ni deposits), magnification 100×

In general, the Ni/RVC® carrier is very interesting from the ecological and the economic point of view. It would be useful even in the design of stationary rechargeable batteries, where the weight of accumulators is not so important, e.g. batteries for energy storage working with wind or solar power sources. This is because the cost of this material is lower than the cost of nickel foam or sintered

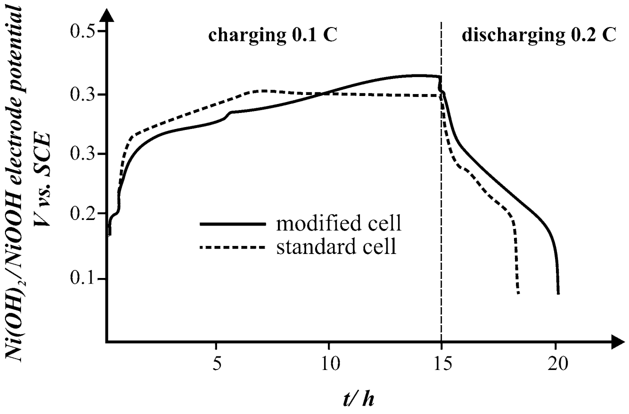

Fig. 9 Charging and discharging curves of $Ni(OH)_2/NiOOH$ electrodes deposited on RVC® in 6 M KOH. Charging at 0.1 C (9 mA g^{-1}) for 14 h. Discharging at 0.2 C (18 mA g^{-1})

Table 2 Specific capacities and capacity densities of Ni/RVC®–$Ni(OH)_2/NiOOH$, "foam" Ni–$Ni(OH)_2/NiOOH$ and sintered Ni–Ni$(OH)_2/NiOOH$—positive electrodes in secondary alkaline batteries

Electrode	Ni/RVC–NiOOH	Sintered Ni–NiOOH	Foam nickel–NiOOH
Specific capacity/mAh g^{-1}	88.70	38.47	160.0
Density capacity/mAh cm^{-3}	99.3	426.0	237.6

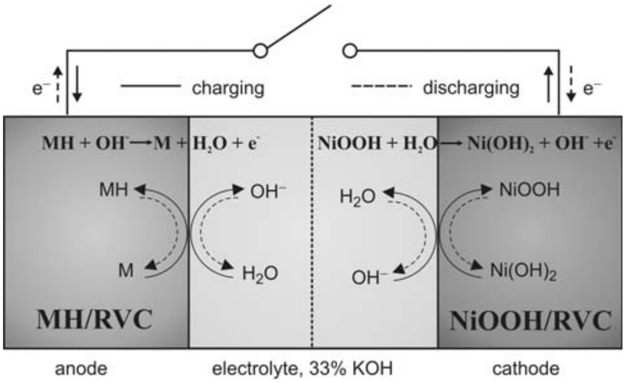

Fig. 10 Electrode reactions in rechargeable Ni–MH cell with RVC® as carrier of cathodic and anodic active masses and as current collector

nickel. Also, the toxicity of carbon covered with nickel is lower and its neutralization is much easier than in the case of "foam" or sintered nickel. The Ni/RVC® carrier can be taken into consideration particularly in applications where low weight matters. In that respect it should be pointed out that when low battery weight and low price play an important role, Ni/RVC® can be used as a matrix not only for the $Ni(OH)_2/NiOOH$ electrode, but also for the anode, e.g., as a carrier of the hydrogen absorbing alloy in NiMH systems. Summarizing, the Ni/RVC® system may be a good matrix for both electrodes in rechargeable battery systems such as Ni–Cd, Ni–Fe and Ni–MH. In the coming era when hydrogen is expected to become the main energy carrier [33] the Ni/RVC® matrix may find application in large-size hydrogen storage systems. The scheme of a Ni–MH cell based on RVC® as the active mass carrier and the current collector is shown in Fig. 10.

2.3 Electrochemical capacitors with Pd and Pd–Rh alloys

Since it has been found that hydrogen can be electrochemically inserted/extracted into/from Pd-based thin layer electrodes, such materials can be treated as phase charging-discharging systems. To characterize the material in the aspect of energy storage one can convert the charge consumed in hydrogen sorption/desorption reactions [34–38] into pseudocapacitance. These Pd-based layers can be treated as phase charging-discharging systems. Figure 11 presents an SEM image of a Pd–Rh alloy (3% Rh) electrochemically deposited on RVC®.

Pseudocapacitance (C) can be calculated for the process of electrooxidation of hydrogen absorbed in the form of the β-phase (absorption potential in the range from −0.2 to 0 V vs. NHE) according to the equation:

$$C = Q_H^{ox} \Delta E^{-1} m^{-1}$$

where Q_H^{ox} is the charge of hydrogen oxidation, ΔE is the potential region in which all absorbed hydrogen is electrochemically removed, and m is the mass of the material. The calculations were done separately by taking the mass of the electroactive material only (i.e. Pd or Pd–Rh layer) as well as taking the sum of the deposit mass and the substrate mass, i.e., the total mass [34]. These values are included in Table 3.

The value of specific pseudocapacitance for Pd and Pd–Rh deposits on RVC® is comparable with the data reported for supercapacitors based on reversible insertion of

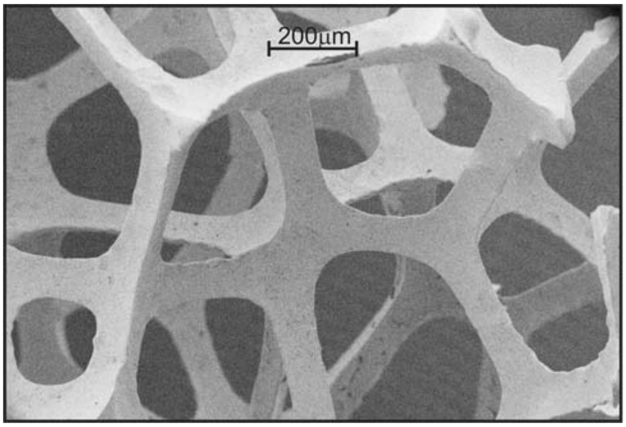

Fig. 11 SEM image of Pd–Rh alloy (3% Rh) electrochemically deposited on RVC® (magnification 50×)

Table 3 Specific pseudocapacitance of Pd and Pd–Rh hydrogen absorbers deposited on various substrates [35, 38]

Electrode material	Specific pseudocapacitance (Fg^{-1})	
	For electroactive mass (i.e. only Pd or Pd–Rh alloy)	For total mass (deposited metals + carrier)
Pd/RVC	1000–2000	240–430
Pd/Au/RVC	1000–2000	130–200
Pd–Rh/RVC	1100–2200	250–540

species (e.g. protons) into the structure of bulk solid electrodes, e.g. polymers or RuO_2 [38–44]. Capacitances obtained for Pd/RVC®, and Pd–Rh/RVC® systems are higher in comparison with the values for typical double layer electrochemical capacitors [39–42] where high-surface-area carbon is used. It should be added that under the conditions of cyclic voltammetry, pseudocapacitance values calculated from Eq. 2 for a particular sample were higher when a lower scan rate was applied. This effect originates from the fact that scan rate affects the shape and width of the hydrogen oxidation peak, and, as a result, ΔE. At a low scan rate, all absorbed hydrogen can be oxidized in a relatively narrow potential range, which results in pseudocapacitance increase. The values of specific pseudocapacitance are in the range 130–540 F g^{-1}, i.e., comparable with those for supercapacitors using various insertion reactions. The best results were obtained for the Pd–Rh/RVC® system. The values of specific pseudocapacitance were calculated for the electrooxidation of hydrogen absorbed as the β-phase ($H/Me \approx 0.7$). The calculations were done for the mass of the electroactive material (i.e., Pd or Pd–Rh layer) with substrate—carrier (RVC®).

2.4 Lead acid battery

During the past 150 years, the construction of the lead-acid battery has changed significantly, but the main disadvantage of the classic battery continues to be the low specific energy density, mainly due to the high mass of lead-based carriers of the electrochemically active mass. The specific energy of lead acid batteries is much lower in comparison with modern types of battery, i.e., Ni–MH and Li–ion, but the cost per energy unit, its robustness and the uncomplicated production technology make this type of battery still attractive. One of the goals pursued by designers of lead–acid batteries is to increase the ratio between capacity and weight.

Pavlov [45] introduced a parameter γ, reflecting the ratio of cathodic active mass to surface area of carrier (grid) of the mass. For typical lead-acid batteries this parameter ranges between 1.6 and 1.8 for tubular and between 2 and 2.5 for book-mould SLI battery plates. To achieve lower γ factors, lead current collectors used in these batteries should be replaced with other lightweight electro-conducting materials. According to our studies [46–52], the use of carbon current collectors is a promising way of improving lead-acid battery performance.

The idea of a lead acid-battery construction with RVC®, depicted in Fig. 12 was first described by Czerwinski in 1995 [46] in a patent. Since than a few patents and papers on the subject have originated from our group [47–53]. The idea of RVC® application as a lightweight carrier and current collector in lead–acid batteries was also presented at the Labat'96 Conference [49]. In 2003, Gyenge et al. [54, 55] have adopted this invention for construction of a lead-acid battery, in which RVC® covered with a Pb alloy was used as the active mass carrier. Recently, we have constructed a model lead-acid battery, in which porous glassy carbon was modified with a deposited thin layer of pure lead [56]. The Pb/RVC® electrode electrochemical behaviour is almost identical to that of pure metallic lead. Similar results were obtained with a Pb/RVC® electrode electrochemically (anodically) covered with PbO_2. The electrochemical behaviour of both electrodes proved that there was no influence of the RVC® matrix on the deposited lead and lead–lead dioxide.

These materials, Pb/RVC® (Fig. 13a) and $Pb–PbO_2$/RVC® (Fig. 13b), have been used as carriers and current collectors of the negative and positive electrodes, respectively. Pure lead or lead oxide(IV), were deposited electrochemically on previously prepared Pb/RVC®. The cell constructed from the electrodes obtained in such a simple way, had a specific capacity of ca. 14 Ah kg^{-1} [53]. Much better results have been obtained for a construction in which RVC® covered with lead was mechanically filled with active mass mixtures of lead powder and lead oxides with additives, i.e., expanders, normally used for classic lead-acid battery construction.

Before electrochemical tests, the negative and positive electrodes were treated with a standard seasoning procedure (Laboratory of Piastów-Sznajder Batteries S.A. and Jenox Battery S.A.) This work was part of a new project

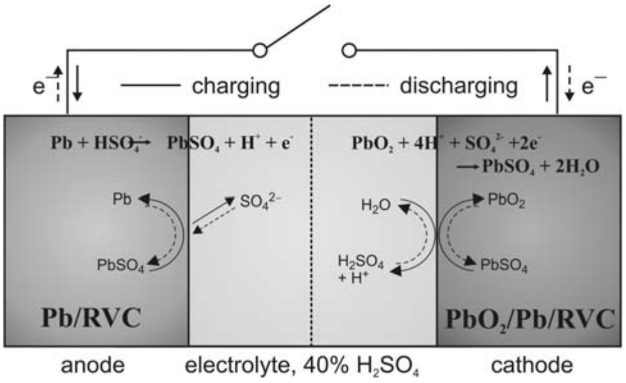

Fig. 12 Electrode reactions in prospective lead acid battery with modified RVC® grids

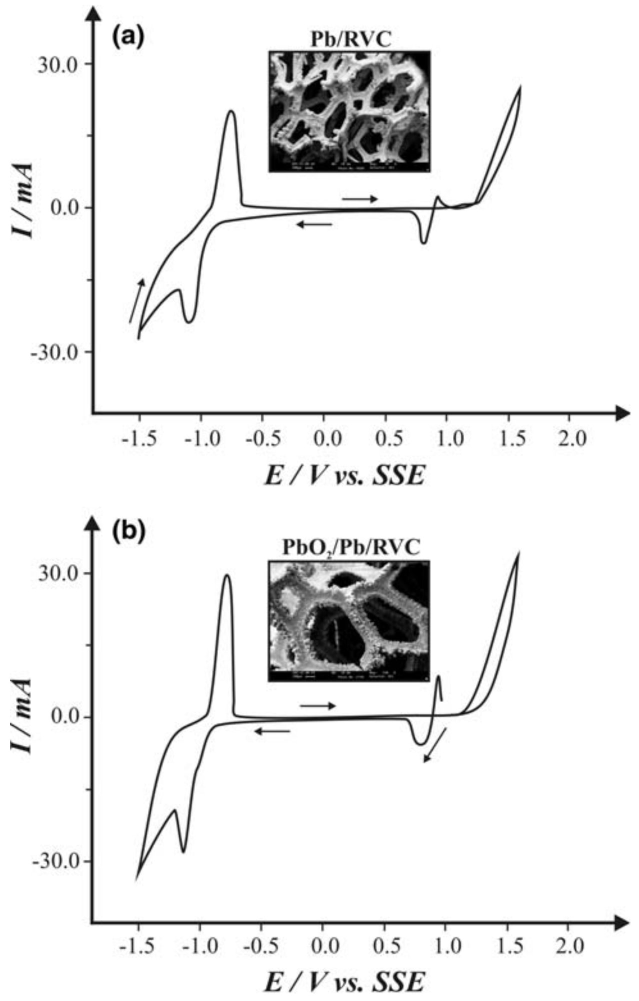

Fig. 13 Cyclic voltammograms of: Pb/RVC® (**a**) and PbO_2/Pb/RVC® (**b**) in 0.5 M H_2SO_4. Temperature 298 K, scan rate 1 mV s^{-1}. Insets: SEM images of respective electrodes, magnification 100×

aimed at constructing a new type of small-sized lead acid rechargeable battery [56]. The Pb and PbO_2 electrodes, obtained in this way, were tested for durability by discharging in electrolytes with different sulfuric acid concentrations. Furthermore, the positive and the negative plates were constructed with RVC® covered with pure lead without antimony or other alloy components. The lead-alloy component, i.e. antimony, facilitates unwanted reactions, which results in a shorter life (i.e. grid corrosion, self-discharging reactions). Preliminary results show that the specific capacity of this RVC® based battery is comparable (or better) than the capacity of SLI batteries [56].

3 Summary

Using the example of reticulated vitreous carbon (RVC®), it has been shown that porous conductive carbon itself and/or modified with deposited metals and their oxides can be used as an attractive electrode material in batteries. This material can be used as an active mass carrier and a current collector. It has to be noted that, often, many types of classical battery have reached their theoretical limit of capacity (capacity of carried active mass) and further improvement of these batteries calls for modification of all elements of the cell. For example, this is the case for zinc–carbon batteries. The application of RVC® as the active mass carrier and the current collector allows for an increase, by ca. 20%, in the amount of stored MnO_2—the cathodic reagent in these batteries. This is due to the fact that the RVC® modified with metals used as an electrode material shows the characteristics of the previously used solid elements, but has a smaller volume. This leads to increased energy density (kW kg^{-1}), which makes the batteries competitive with other types of zinc–carbon power sources. A similar trend has been demonstrated with lead–acid batteries, where the heavy carriers and collectors made from lead alloys have been substituted with RVC® covered by lead and/or lead dioxide. Also, RVC® covered with nickel was successfully applied as current collector and carrier of $Ni(OH)_2$—the active cathodic mass used in alkaline rechargeable batteries, i.e. Ni–Cd and Ni–MH. Modified RVC® can also be applied in a new type of electrochemical power source—electrochemical supercapacitors.

All these results indicate that porous carbon materials like RVC® are promising materials for many kinds of new electrochemical power sources functioning as an electrode material and/or current collector and simultaneously acting as active mass carrier.

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