

PLASTIC BONDED ELECTRODES FOR NICKEL-CADMIUM ACCUMULATORS

II. BASIC ELECTROCHEMICAL PARAMETERS OF THE NICKEL OXIDE ELECTRODE

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Summary

Plastic bonded nickel oxide electrodes, prepared at normal temperature by one-stage rolling onto a current collector of steel net or perforated sheet of a mixture of active mass used in pocket-type electrodes, a conducting component and a PTFE binder, were tested in alkaline electrolyte. At current loads of 3 - 100 mA/cm² the type of conducting admixture, the current collector and its surface treatment were found to have a pronounced influence on the current carrying capability of the electrode. The electrode performance, especially at 100 mA/cm², is dependent not only on the composition of the active layer but also on the quality (or nature) — but not degree of contact between it and the current collector.

Introduction

The reasons for the commencement of research and development work in Czechoslovakia on plastic bonded electrodes for alkaline nickel-cadmium accumulators were discussed in the preceding paper [1] on plastic bonded cadmium electrodes.

The plastic bonded nickel oxide electrode (subsequently referred to as: PB-Ni electrode) is prepared, in principle, by one-stage rolling of a mixture of nickel(II) hydroxide, a conducting component, and a binder, onto a metal collector at normal temperature [2, 3]. The problems which it presents are partly inherent in nickel oxide electrodes of all hitherto known types, *i.e.*, a complicated mechanism of the electrode process and thermodynamic instability of the anodic oxidation products, and, in addition, there are pronounced volume changes of the solid phase. It is the first type of nickel oxide electrode which has been prepared at a lower manufacturing cost than pocket or

sintered electrodes, and its electrochemical parameters are intermediate between these two types of nickel oxide electrode.

The complicated mechanism of the electrode process is due to the direct transfer of electrons between the reduced and oxidised form in the solid phase. In the course of the electrode process, one form changes into the other through a continuous phase change of nonstoichiometric oxides. From the point of view of maximum utilization of the oxidised form during discharge, the manufacturer of nickel oxide electrodes must overcome the problem of an abrupt change of solid phase electrical conductivity ranging from an order of $10^{-1} \text{ ohm}^{-1} \text{ cm}^{-1}$ in the oxidised state to $10^{-13} \text{ ohm}^{-1} \text{ cm}^{-1}$ in the reduced state [4].

The thermodynamic instability of the oxidised form leads to oxygen evolution, which proceeds even in the currentless state and is the cause of self discharge of nickel oxide electrodes.

The considerable volume changes of the active mass are due to the fact that we have to deal with hydrated nickel oxides, which behave as a hydrogel with a variable molar volume [5]. Thus, in parallel with the continuous phase change of the nonstoichiometric nickel oxides during the electrode process, the content of water of hydration, which is partly bound in the crystal lattice and partly physically adsorbed, undergoes a continuous change.

With commercial nickel oxide electrodes of all types, there is a complication in that the conductivity of the system, which behaves essentially as a semiconductor [6], must be improved by the addition of a conducting component, an expensive sintered plaque in the case of sintered plate cells or in the case of the economically preferable pocket electrode, whose conductivity, in comparison with sintered electrodes, is much worse, an admixture of powdered graphite.

The considerable volume changes occurring mainly during the first charge-discharge cycles are, in the case of pocket electrodes, utilized to improve their function. The active mass, in the dry state in the form of a briquette, is enclosed in the perforated steel pocket; the volume changes cause an internal pressure in the pocket which is favourable for the conductivity and thus for the electrochemical functioning of the electrode. With sintered electrodes, however, the volume changes are not so favourable and they limit the amount of active material which can be deposited in the porous support, especially when the mechanical strength of the latter is not very high.

At the present state of research on PB-Ni electrodes in Czechoslovakia, the powdered mixture used to prepare the briquette for pocket electrodes is blended with a plastic binder whose negative effect on the electrical conductivity is compensated for by a further addition of the conducting component. The use of powdered active material is advantageous for the manufacturer in that the changeover from pocket electrode technology to that of PB-Ni electrodes does not require extensive changes in the preparation of the active material, and the available technological experience can be used. The new technology must, however, overcome the difficult hygiene problems,

arising from the stringent standards applied to the use of toxic materials, which the manufacturer of both cadmium and nickel oxide pocket-type electrodes has to solve.

Thus, the conditions for the new method of manufacturing nickel oxide electrodes can be summarized as follows:

- (a) high conductivity between the electroactive particles;
- (b) satisfactory rolling properties of the electrode mix at normal temperatures;
- (c) minimum modification of the active mass currently used in pocket electrodes;
- (d) optimum hygienic conditions in the preparation and manipulation of the electrodes.

Condition (a), which means that the active particles are involved in the current forming process in spite of the considerable volume changes of the active layer, led to the development of an elastic and conducting bonding of the electroactive particles. A combination of a plastic material with a conducting component proved most satisfactory.

The rolling properties of the electrode mix (hydrated nickel oxide, conducting component, polymeric binder) can be controlled mainly by the type of conducting component and binder; its plasticity can be improved by the addition of a liquid rolling (milling) agent, which acts, to some extent, also as a pore forming agent [2]. Teflonised carbon black served both as binder and conducting component in a most satisfactory manner, although graphite mixed with powdered PTFE also gave promising results.

Condition (d) is fulfilled by using the plastic binder and the liquid rolling or milling agent, which almost completely eliminate the formation of toxic aerosols in the working atmosphere.

Certain manufacturing methods for the PB-Ni electrode have been described in the literature [7 - 12], but they are unacceptable either because they are too expensive (multistage rolling at elevated temperatures [10]) or because they do not fulfil condition (d) due to the use of too high working temperatures and large volumes of organic solvents [7, 8, 10]. Certain methods are suitable only for nickel oxide electrodes for Ni-Zn accumulators, where the demands on the electrochemical parameters of these electrodes are relatively very low [11, 12], especially in respect of life, which (because of the restricted life of the Zn electrode) attains only 100 - 400 cycles.

Experimental

All experimental electrodes were made from commercial active material of KBL type, manufactured in Czechoslovakia (Lachema, Bohumín) for nickel oxide pocket electrodes. It contains precipitated $\text{Ni}(\text{OH})_2$ blended with powdered graphite; the respective contents are 68 - 74 and 16 - 19% and the water content is 4 - 7%. The 74% $\text{Ni}(\text{OH})_2$ content corresponds to a theoretical capacity of $C_t = 0.214 \text{ A h/g}$, assuming conversion to NiOOH .

Teflonised carbon black was prepared by adsorption of a PTFE dispersion (GP-1 or GP-21, ICI, Great Britain) on two types of carbon black: carbon black 1 — acetylene black P-1042 (VEB Stickstoffwerke Piesteritz, G.D.R.) and carbon black 2 — furnace black Nigros (HAF) (Urxovy závody, Valašské Meziříčí, Č.S.S.R.). Graphite or carbonyl nickel powder was mixed with powdered PTFE (FLUON CD-1, ICI, Great Britain). Other binders such as polyethylene, polystyrene, polyamide, etc., proved unsatisfactory because of poor rolling properties of the electrode mix. The carbonyl nickel was the same as that used for manufacturing sintered nickel plaques for Ni-Cd accumulators (INCO 255), graphite of VA type was a product of Rudné doly, Týn n. Vlt., Č.S.S.R.; its sedimentation analysis showed a wide spectrum of particles in the range 10 - 200 μm of which about 55% passed a sieve of 0.16 mm mesh.

The above mentioned main components of the PB-Ni electrode were thoroughly mixed and then milled for a short time in an electric mill. The mixture was wetted with a rolling agent such as ethanol, which improved the rolling properties and also served to some extent as a pore forming substance. The slightly wetted mix was placed in the reservoir of a rolling machine and carried by the current collector between the rollers. By rolling the electrode mix on both sides of the current collector at normal temperature an electrode strip was formed which was dried, cut into 4.0×7.5 cm pieces and pressed at 120 MPa. The electrode thickness varied, according to the type of current collector and the composition of the electrode mix, from 2.0 to 2.4 mm. Steel screens electroplated with nickel were used as current collectors. Their characteristics (prior to nickel plating) are given in Table 2; their relative open area was 64% in all cases.

The theoretical capacity (C_t) of PB-Ni electrodes of 60 cm^2 working area was about 1.9 A h. The electrodes were tested in laboratory cells with two PB-Cd electrodes, each of about the same C_t value. The separator was either a layer of 42 g/m^2 , 0.1 mm thick, polyamide fabric "Upraka" (Hedva, Moravská Třebová, Č.S.S.R.), or an additional layer of 43 g/m^2 , 0.2 mm thick, nonwoven polyethylene fabric (Juta, Úpice n. Metují, Č.S.S.R.). The reference electrode was Hg/HgO in the same electrolyte as in the cells.

The electrode to be tested was first subjected to 3 - 4 formation cycles (charging at 3 mA/cm^2 , discharging at 5 mA/cm^2) in a solution of KOH of density 1.2 g/cm^3 . This was followed by charging at 3 mA/cm^2 and a high rate discharge test at 100 mA/cm^2 , and the capacities obtained to end potentials of 0, -200, and -500 mV were determined as a percentage of the theoretical capacity, C_t . Finally, the current-voltage ($i-E$) characteristic was measured after the electrode had been charged for 3 h at 12 mA/cm^2 : the end potentials on 5 s loadings at 17 - 200 mA/cm^2 applied at 30 s intervals were recorded. The results are given as the slope of the $i-E$ characteristic (ohm cm^2).

In another series of tests, the electrode life was determined. One type of life test consisted of an accelerated cycling regime in which the test electrode was charged for 2 h and then discharged for 1 h at 10 - 13 mA/cm^2 in

a solution of KOH (1.2 g/cm^3) containing 20 g of $\text{LiOH} \cdot \text{H}_2\text{O}$ per liter. Always, after about 200 accelerated cycles, three test cycles were made at 3 - 5 mA/cm^2 . These were followed by a discharge test at 100 mA/cm^2 and measurement of the i - E characteristics.

Another type of life test involved carrying out daily discharge cycles at 100 mA/cm^2 to -0.5 V , five times per week; the electrodes were always charged at 3 mA/cm^2 . The result of this test was the number of discharge cycles to a predetermined limit, according to which the electrode must give at least 27% C_t to an end potential of -300 mV .

Results and discussion

Influence of the type of conductive component

In consideration of the planned production of PB-Ni electrodes, the conductive component was selected according to the following criteria: (a) good electronic conductivity, (b) resistance against anodic oxidation (especially during overcharge), (c) good rolling properties of the electrode mix, and (d) low price and easy availability.

These conditions were best met with graphite, carbon black, and carbonyl nickel powder. Their influence was tested in an electrode mix of composition 75 - 86.6% KBL, 8.7 - 10% PTFE, and 3.3 - 16.3% conductive component. The current collector was a steel net of 4.5 mm mesh and 1.12 mm wire dia.

The discharge curves for the third or fourth formation cycle at the 5 mA/cm^2 rate for electrodes containing 82.5% KBL, 9.6% PTFE, and 7.9% conductive component are shown in Fig. 1. It is seen that apart from carbon black 2, the other three conductive components gave satisfactory discharge curves comparable with pocket electrodes. With these three components, the discharge curves had a constant form within the range of composition of the electrode mix given above. The capacity of electrodes containing carbon black 2 was lower than in the case of the other conductive components (Fig. 1) and the electrolyte turned markedly yellowish brown, evidence of anodic corrosion of this material. The oxide layer thus formed apparently causes some form of passivation, whereby a part of the active material ceases to take part in the electrochemical process. However, the oxidation products in the electrolyte have no adverse effect on the electrode process, as was proved by interchanging the electrolytes belonging to electrodes with carbon blacks 1 and 2.

The capacities obtained in discharge tests both at the 5 mA/cm^2 and at the 100 mA/cm^2 rates are given in Table 1. Electrodes with carbon black 2 were not considered, as they obviously have no practical importance. The slopes of the current-voltage characteristics are also included. It is seen that, in all cases, especially in high discharge rate tests, the PB-Ni electrodes with carbon black 1 gave the most promising results. They were therefore considered for further work.

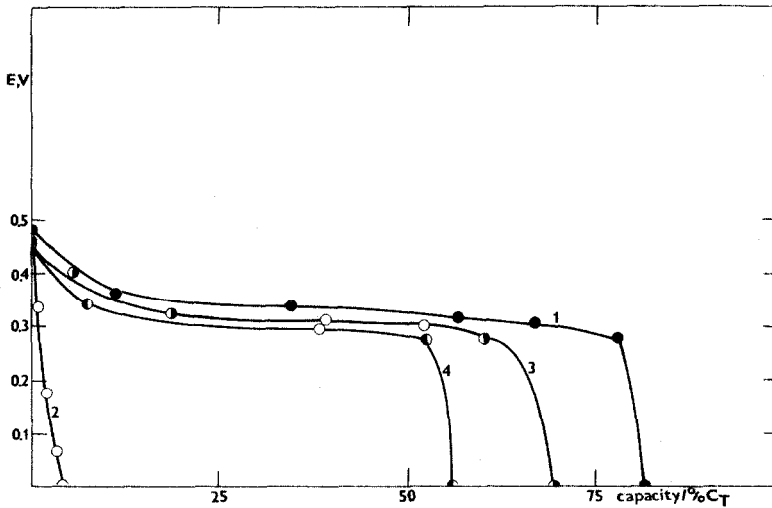


Fig. 1. Discharge characteristics at 5 mA/cm^2 rate of PB-Ni electrodes prepared from 82.5% KBL, 9.6% PTFE, and 7.9% conducting component. The conducting component was: 1, carbon black-1; 2, carbon black 2; 3, carbonyl nickel; 4, graphite.

TABLE 1

| KBL (%) | PTFE (%) | Conducting component type | component (%) | Electrode capacity (% C_t) | | | $E-i$ slope (ohm cm^2) | |
|---------|----------|---------------------------|---------------|-------------------------------|-------|--------|----------------------------------|--------|
| | | | | 5 mA/cm^2 | | | | |
| | | | | 100 mA/cm^2 | 0.0 V | -0.2 V | | -0.5 V |
| 75.0 | 8.7 | graphite c. black | 16.3 | 56.5 | 26.1 | 40.0 | 53.3 | 1.45 |
| | | | | 67.2 | 2.7 | 4.3 | 6.4 | 1.1 |
| | | | | 60.8 | 17.1 | 33.6 | 44.2 | 1.7 |
| 82.5 | 9.6 | graphite c. black | 7.9 | 83.1 | 45.3 | 56.5 | 70.4 | 1.7 |
| | | | | 61.3 | 9.6 | 16.0 | 38.9 | 2.5 |
| | | | | 83.1 | 16.0 | 40.5 | - | 2.2 |
| 86.7 | 10.0 | graphite c. black | 3.3 | | | | | |

The discharge characteristics of electrodes containing 82.5% KBL, 9.6% PTFE, and 7.9% carbon black 1 on a collector with 5.4 mm mesh and 1.12 mm wire diameter are given in Fig. 2. This admixture content corresponds to 17.5% S-55 (*i.e.*, carbon black 1 with 55% PTFE). These electrodes best meet the technological (rolling properties of electrode mix, mechanical strength, electrode thickness) as well as the electrochemical requirements (current carrying capability). A lower content of carbon black (S-75) causes some technological difficulties due to the high PTFE content, a higher content of the black (S-35) gives electrodes with a greater thickness and the rolling properties of the electrode mix are less satisfactory.

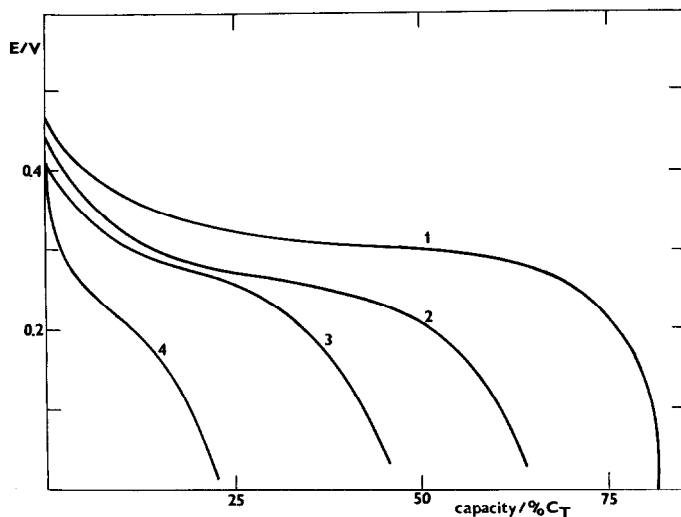


Fig. 2. Discharge characteristics of PB-Ni electrodes prepared from 82.5% KBL, 9.6% PTFE, and 7.9% carbon black 1 at various discharge rates: 1, 5 mA/cm²; 2, 20 mA/cm²; 3, 50 mA/cm²; 4, 100 mA/cm².

Based on tests of electrodes with different conductive component resistance against anodic oxidation and different bulk density (carbon black 1, 40 g/l; graphite VA, 540 g/l; carbonyl nickel, 770 g/l), it is obvious that the current carrying capability of the PB-Ni electrode and its life can be increased by improving the contact between the electroactive particles using a conductive component network. It can therefore be expected that the quality of the current collector will also play a role.

Influence of the current collector on electrode performance

The capacities obtained from electrodes with an active layer composition of 82.5% KBL + 17.5% S-55 at the 100 mA/cm² discharge rate to zero V after 3 - 4 formation cycles are given in Table 2, where the type of current collector and the number of discharge cycles at the 100 mA/cm² rate up to the given limit are also indicated. It is seen that with the use of a screen with 2.8 - 4.5 mm mesh and 0.7 - 1.1 mm wire diameter, higher capacities and cycle numbers were obtained than in the case of a screen with 5.0 mm mesh and 1.25 mm wire diameter.

Surface treatment of the nickel-plated screens by sand blasting followed by nickel spraying causes an increase in cycle life (at 100 mA/cm²) by a factor of 2 - 6. Pocket electrodes attained 120 - 180 cycles in comparable tests at the same rate. The slopes of the current-voltage characteristics after formation cycles were in the range 1.0 - 1.7 ohm cm² for PB-Ni electrodes (with no marked effect of the current collector and its surface finish), and 1.8 - 2.1 ohm cm² for pocket electrodes.

TABLE 2

| Collector type | Wire diam. (mm) | Mesh size (mm) | Mass (kg/m ²) | Capacity at 100 mA/cm ² (% C _d) | | No. of cycles to the limit | |
|----------------|--------------------|-------------------|------------------------------|---|-------------|----------------------------|---------|
| | | | | untreated | treated | untreated | treated |
| "Ferona" | 1.25 | 5.0 | 3.15 | 27.7 - 34.6 | - | 3 - 13 | - |
| "Metro" | 1.12 | 4.5 | 2.84 | 40.5 - 45.3 | 45.4 - 55.4 | 2 - 10 | 55 - 63 |
| "Ferona 0.8" | 0.80 | 3.15 | 2.00 | 36.8 - 46.4 | 46.4 - 52.8 | 20 - 40 | 60 - 69 |
| "Ferona 0.71" | 0.71 | 2.8 | 1.82 | 38.4 - 46.9 | 49.0 - 56.0 | 25 - 45 | 60 - 65 |

It is, hence, apparent that as a consequence of the screening effect of the perforated sheet on the active layer of the pocket electrode, the PB-Ni electrodes outdo pocket electrodes in current carrying capability after only 3 - 4 formation cycles. However, in continuing discharge cycles at the 100 mA/cm² rate, the PB-Ni electrode gradually loses its originally high current carrying capability, whereas the pocket electrode maintains this.

Our results suggest that one of the main reasons why PB-Ni electrodes lose their performance is a gradual loss of contact between the active layer and the metal screen, probably owing to volume changes of the active layer during cycling [13]. The surface treatment of the screen by nickel spraying is effective in this respect since it ensures a better adhesion of the active layer to the screen. With pocket electrodes, the active layer is mechanically pressed into the perforated steel pocket and the contact resistance between them decreases in the course of formation cycles.

The effect of surface treatment of the current collector is also favourable in life tests of the accelerated cycling type. The electrodes used had again the composition 82.5% KBL + 17.5% S-55. The collectors were of three types: "Feron 0.71" with and without surface treatment, and "Metro" with a nickel-sprayed surface. The changes in the capacity at the 3 mA/cm² rate, the slope of the *i*-*E* characteristics, and the capacity at the 100 mA/cm² rate with the number of accelerated cycles are shown in Fig. 3(a) - (c). The favourable effect of the surface treatment of the current collector is obvious; with test cycles at the 3 mA/cm² rate the collector with thinner wire and smaller mesh size also has a positive influence.

For commercial purposes, only applications at the 20 mA/cm² rate are practical at the present state of development of the PB-Ni electrode. Further development work must aim at maintaining the current carrying capability (measured, for example, at the 100 mA/cm² rate and from current-voltage characteristics) during long-term accelerated cycling.

The quality of the active layer-current collector interface also manifests itself in another type of life test, namely, daily discharge cycles at the 100 mA/cm² rate to -0.5 V. In these tests, expressed as the number of cycles up to a predetermined limit, four groups of current collectors were used: nickel-plated steel screens (different plating methods), similar screens with additional surface modification by nickel spraying, steel screens of different form, and finally, metal sheet perforated from both sides with, and without, surface treatment. The electrode mass was 82.5% KBL and 17.5% S-55 and the results obtained are given in Table 3.

With the first group of collectors (different galvanic plating), we found a favourable effect of dull against bright plating and also a favourable effect of thinner wire and smaller mesh size of the steel screen. This shows the importance of the adhesion of the active layer to the collector and of the contact surface area between both phases for the current carrying capability.

The effect of better adhesion is further confirmed by the second group of collectors (nickel-sprayed steel screen), which shows less scattered, and markedly higher numbers of discharge cycles at the 100 mA/cm² rate.

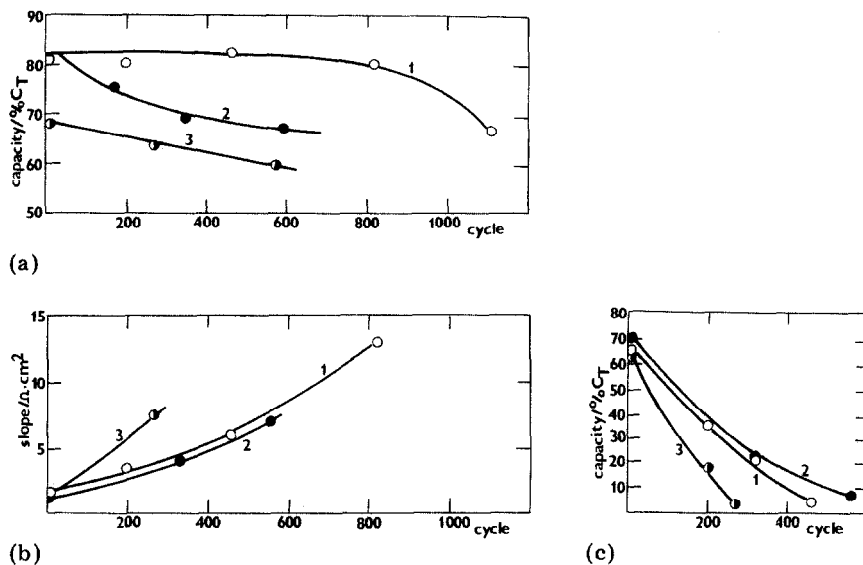


Fig. 3. Electrochemical parameters measured in the course of accelerated cycling of PB-Ni electrodes prepared from 82.5% KBL, 9.6% PTFE, and 7.9% carbon black-1 with different current collectors: 1, Feron 0.71 (metallized); 2, Feron 0.71 (mat nickel plated); 3, Metro (metallized). 3(a). Cycle life time characteristics at 3 mA/cm² discharge rate; 3(b) slopes of current-voltage characteristics; 3(c) cycle life time characteristics at 100 mA/cm² discharge rate.

TABLE 3

| Collector type | No. of cycles to the limit |
|---|----------------------------|
| "Feron" (mat nickel plated) | 3 - 13 |
| "Metro" (mat nickel plated) | 2 - 10 |
| "Feron 0.8" (bright Ni plated) | 19 - 20 |
| "Feron 0.8" (mat nickel plated) | 20 - 40 |
| "Feron 0.71" (mat nickel plated) | 25 - 45 |
| "Metro" (metallized) | 60 - 69 |
| "Feron 0.8" (metallized) | 60 - 65 |
| "Feron 0.71" (metallized) | 55 - 63 |
| "Feron" (with nickel gauze) | 27 - 54 |
| "Metro" (with nickel gauze) | 56 - 70 |
| "Feron 0.4" (corrugated) | 55 - 60 |
| Perforated steel sheet (Ni plated) | 2 - 5 |
| Perforated steel sheet (sand blasted, Ni plated) | 25 - 30 |
| Perforated steel sheet (sand blasted, metallized) | 48 - 50 |

With the third group of collectors, we attempted to increase the contact surface area by combining the dull, nickel-plated collectors with a fine nickel gauze (1.25 mm mesh and 0.16 mm wire diameter), rather than by nickel spraying (which is not economical). The fine nickel gauze was either cut into narrow strips, drawn through the coarse meshes of the basic collector and, at some points, electrically welded to it, or the fine gauze was additionally pressed from both sides into the electrode and then electrically welded to the central collector. For comparison, a wavy steel screen with a wire half as thick and an appreciably smaller mesh size against the common collectors was used. The purpose was to ensure a maximum contact surface area. However, electrodes with this collector type had low mechanical strength and, hence, are not suitable for practical use. This third group of collectors did not differ much from the second in respect of the number of cycles attained, hence nickel spraying of the current collectors can be replaced by a mechanical modification whereby the contact surface area between the collector and the active layer is enlarged.

A current collector from a perforated metal sheet of 0.1 - 0.2 mm thickness would be preferable from the manufacturing point of view. The corresponding results are given in Table 3, from which the use of such a collector appears practical; the number of cycles attained increased in the order: glossy surface of the collector — surface treated by sand blasting — surface treated by nickel spraying.

Conclusions

A mechanically rigid nickel oxide electrode capable of long-term cycling at up to the 20 mA/cm^2 discharge rate can be prepared by a rolling technique with the use of PTFE. Its current carrying capability depends on the conducting component, which compensates for the effect of the nonconducting plastic. Carbon black proved most suitable because of its low bulk density (high dispersivity) and resistance against anodic oxidation, the good rolling properties of the electrode mix, and for economic reasons. The cycle life of the electrode described, determined either by a series of cycles at the 100 mA/cm^2 rate or by discharge at the same rate in the course of accelerated cycling, depends on the quality of the active layer-collector interface. The current carrying capability is proportional to this contact surface area which can be enlarged either by metallization of the collector or by a combination of coarse and fine metal screens.

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