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DEVELOPMENT OF HIGH-PERFORMANCE Na/NiCl₂ CELL

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ANL/CP--75875

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Abstract

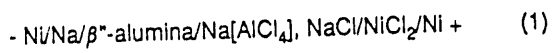
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These earlier studies also suggested that a porous electrode with a specially tailored morphology would improve performance by providing high surface area and sufficient pore cavities to support effective mass transport. Also, we found indications with these experiments that chemical additives would improve the properties of the electrochemically forming NiCl₂ layer.

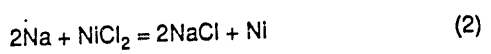
Based on the earlier work, we initiated systematic research to improve the Ni/NiCl₂ electrode. As a result, a breakthrough was achieved in 1991. The usable capacity was increased by five times and the area-specific impedance of the Ni/NiCl₂ electrode was reduced to one-third that of a baseline electrode [3]. Figure 1 shows the area-specific impedance (ASI_{15s}), measured by an interrupted galvanostatic method, as a function of discharged capacity per unit volume for several porous Ni/NiCl₂ electrodes. The baseline electrode represents our electrode fabricated without additives using an earlier sintering process characteristic for mid-1990. The usable capacity defined in this figure is the discharged capacity when the area-specific impedance (ASI_{15s}) reaches 4 ohm-cm² or the cell voltage drops to 1.9 V. The termination points of the curves at progressively higher utilized capacities indicate increased available energy density (Wh/cm³), and the lowered area-specific impedance of the Ni/NiCl₂ electrode (ohm-cm²) indicates higher power capability. The ASI of the Ni/NiCl₂ electrode was calculated by subtracting the ASI of the Na/ β -alumina electrode, which was measured in separate experiments, from the cell value.

Introduction

The cell diagram of the Na/NiCl₂ cell is



Sodium and Na[AlCl₄] are molten at the operational temperature (170 to 400°C) of the cell. Na[AlCl₄] is added to the porous Ni/NiCl₂ electrode to transport Na⁺ ions from the surface of the β -alumina electrolyte to the reaction sites at the interior of the positive electrode. The cell reaction is



These cells are under intensive development in England and Germany because of the attractive high voltage (2.58 V at 300 °C) and theoretical specific energy of the cell (790 Wh/kg). Also attractive are the mid-temperature range and the reliability of the cell construction owing to a failure mode that results in a short circuit within the cell. The specific power and energy of the present battery construction, however, are modest due to the performance-limiting positive electrode. To overcome this problem, we sought to improve the performance of this electrode and to better understand its charge and discharge processes.

Use of a single additive significantly improved electrode performance, as shown by the higher available specific energy and the much lower area-specific impedance in Fig. 1. The morphology of the sintered Ni/NiCl₂ electrode was modified by using a poreformer during fabrication to attain controlled pore-size distribution. This modified morphology along with an additive further improved performance. Recently, we found that a combination of the additives in the modified morphology electrode has a significant synergistic effect, producing even more energy per unit volume and even lower area-specific impedance. The chemical additives produce high nickel utilization and low electrode impedance, probably due to doping effects. By an optimal combination of additives, we were able to increase the utilization of the nickel matrix from 15% to about 45%.

Basic Studies

Our earlier potentiometric, coulometric, and cyclic-voltammetric investigations on nonporous Ni electrodes [1,2] suggested that a low-conductivity NiCl₂ layer formed on the electrode during charge. The increasingly higher resistance of the charge product stopped the further thickening of the layer and thereby limited the charge uptake and, consequently, the available capacity in the subsequent discharge. We termed this available capacity "area-capacity limit" (ACL). Depending on the conditions, e. g., temperature, the ACL is 0.4-0.8 C/cm² of the true surface area of the nonporous nickel electrode. During discharge, on the other hand, the NaCl formation (Eq. 2), accompanied by increasingly higher impedance, limited power.

We have studied the effects of pore-size distribution on electrode performance by measuring capacities of model electrodes of various porosities (nonporous Ni, Ni felt, Ni powder sintered with and without poreformer) under identical conditions. From the results, we determined the optimum pore-size distribution of the nickel electrode. Results also indicated that capacity density (mAh/cm³) improved with increased BET surface area [4]. These measurements provided firm theoretical basis for the improvement of the modified-morphology Ni/NiCl₂ electrodes shown in Fig. 1.

Performance Measurements of Research Cells

For exact measurements of the energy density and area-specific impedance, we designed and operated small Na/NiCl₂ research cells of about 0.6-1.8 Ah capacity. The research cell

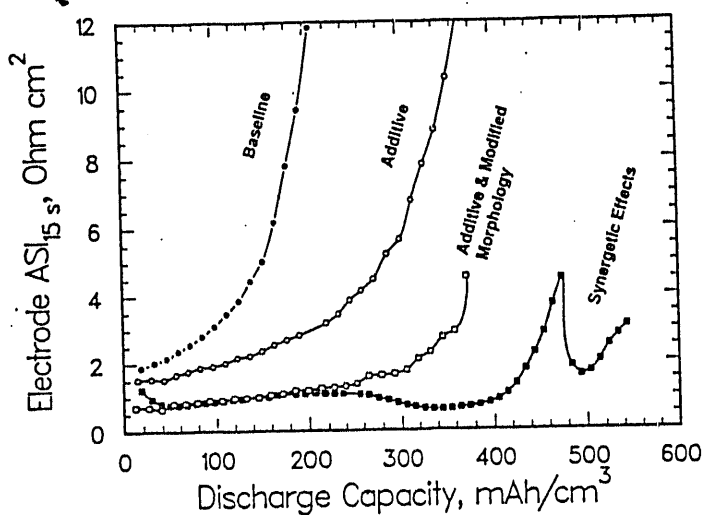


Figure 1. Performance characteristics of 0.5-cm thick Ni/NiCl₂ electrodes at 300 °C.

models the components and mimics the operational conditions of a full-size Na/NiCl₂ cell. By providing one-dimensional current density distribution in the Ni/NiCl₂ electrode, we can precisely measure the important electrochemical engineering parameters [5] necessary for the modeling and performance-projecting calculations that we use to evaluate designs of full-size batteries. The results obtained for some of the characteristic Ni/NiCl₂ electrode designs are explained and shown in Fig. 1.

Increased temperature improves energy density and lowers the impedance for the Na/NiCl₂ cell. The trend indicates more improvement at even higher temperatures, but, because of the higher solubility of the nickel species, this performance-improving approach has questionable value. Earlier Ni/NiCl₂ electrodes had to be operated at 300°C or higher temperatures to achieve acceptable performance (Fig. 2). Our latest Ni/NiCl₂ electrode composition (ANL92), however, can be operated with good performance in Na/NiCl₂ cells even at 170°C (Fig. 2). The exceptionally wide temperature range (170-340°C) within which the cell can be operated offers easy thermal management solutions. To date, several of our research cells achieved about 400 deep cycles at 300 °C without failure. In addition, the ANL92 electrode can be rapidly recharged; recharge times as short as 1 to 3 h have been achieved, as shown in Fig. 3.

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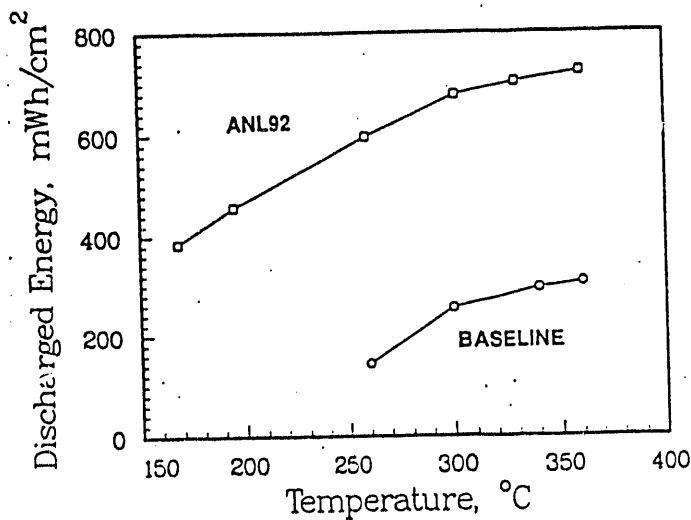


Figure 2. Effect of temperature on discharge capacity of a 0.5-cm thick ANL92 Ni/NiCl₂ electrode; 20 mA/cm² current density.

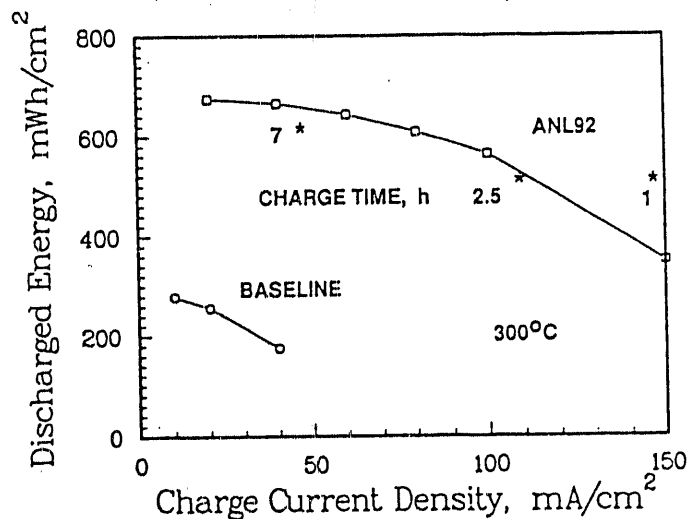


Figure 3. Effect of current density and recharge time on discharge energy. *Charge time in hours.

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Modeling Performance projections for EV Batteries

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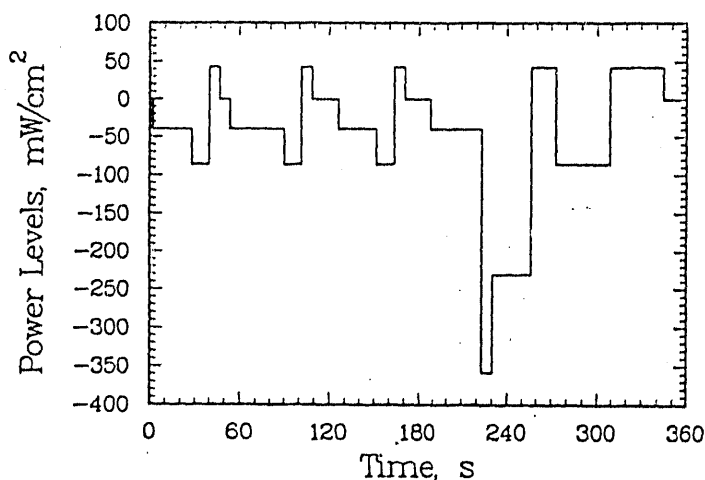


Figure 4. Area-specific power levels of SFUDS cycles used to investigate dynamic performance of Na/NiCl₂ research cells.

Acknowledgment

The authors are grateful to Dr. Kenneth Heitner and Mr. Edward Dowgiallo of the Office of Transportation Systems - Electric and Hybrid Propulsion Division, U. S. Department of Energy. This work was supported by the Department of Energy under contract No. W-31-109-Eng. 38.

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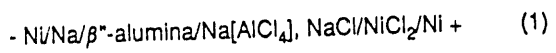
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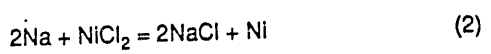
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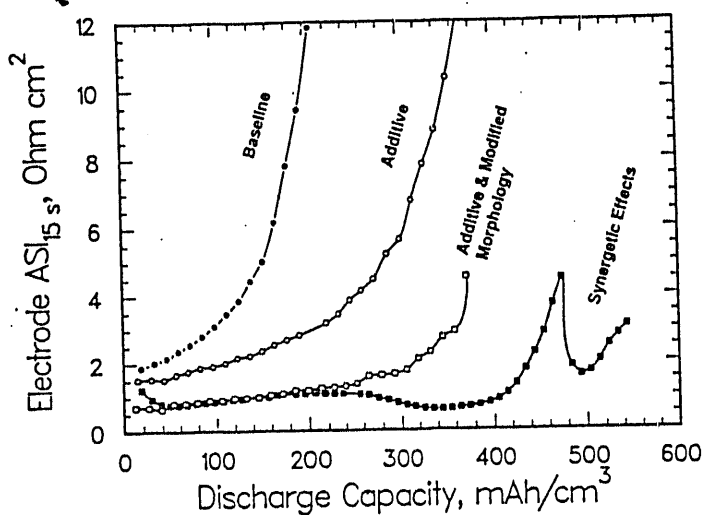


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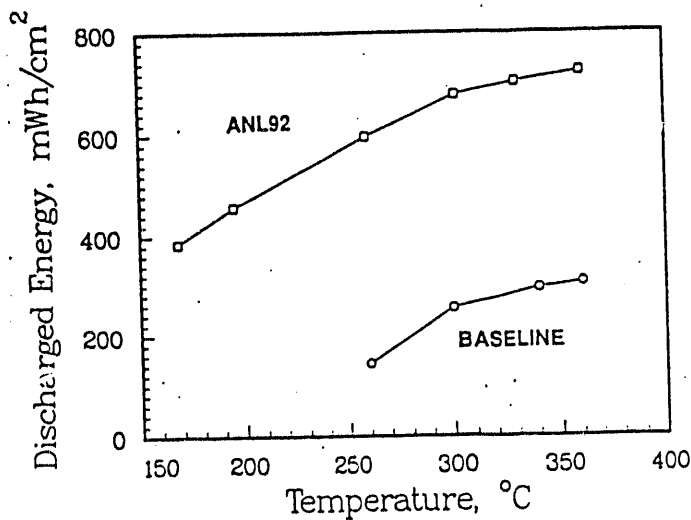


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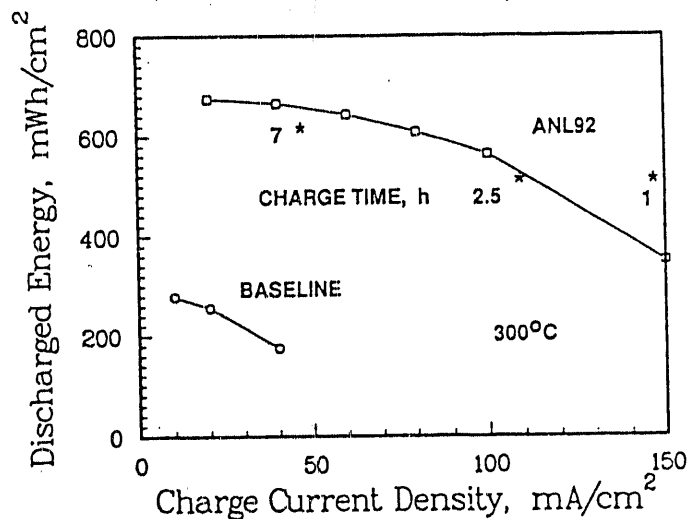


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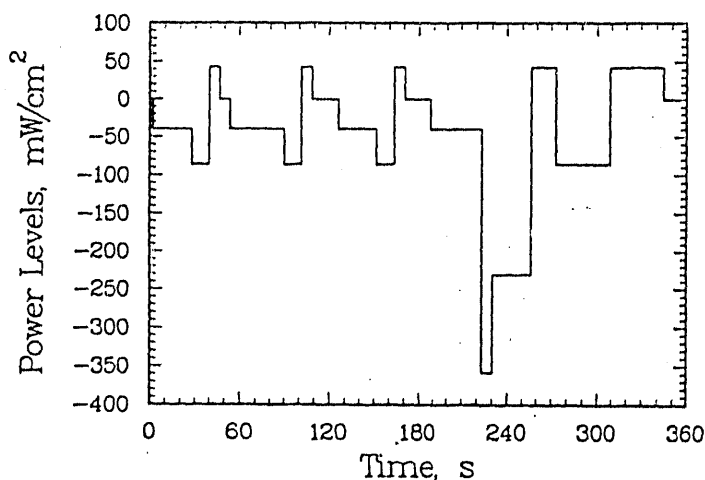


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