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Effects of electrode thickness on power capability of a sintered-type nickel electrode

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Abstract

The performance capability of sintered-type nickel electrodes with thicknesses of up to 4.6 mm was studied in an Ni/H_2 cell. The utilization of active material at high rate discharge diminishes as the thickness of electrode increases. Full utilization at C rate discharge was obtained with electrodes with a thickness of 2.5 mm. A 1 mm thick electrode showed a well-defined discharge curve up to $20C$ rate discharge. Electrodes of up to 3.5 mm thickness showed sufficiently high rate capability for a geosynchronous earth orbit (GEO) application, while the maximum acceptable thickness for a low earth orbit (LEO) application is limited to a value below 2.5 mm. It appears that there is a limited amount of usable capacity at a given current density because of electrode polarization. The value of the current density appears to be independent of electrode thickness.

Keywords: Electrodes; Nickel; Thickness

1. Introduction

The use of few thicker electrodes instead of many thin electrodes is often advantageous for a battery cell. Reducing the number of electrodes in a cell improves the specific energy of the cell and reduces the cell production cost. The main disadvantage of using thick electrodes is that charge and discharge rate capabilities of the cell diminishes as the thickness increases. The maximum practical thickness can also be limited by the rate capability of the counter electrode. Such a limitation of the counter electrode might be encountered in anodes of many alkaline secondary cells such as Ni/Cd, Ni/Zn and Ni/Fe cells. In these cells, the rate-limiting electrode that determines the maximum practical thickness is likely to be the anode rather than the nickel cathode.

In an Ni/H_2 cell, however, the rate-limiting electrode is usually the nickel electrode. Therefore, a thick nickel electrode can be used if it has high sufficient rate capability for the intended application. In the Ni/H_2 cell, use of few thick nickel electrodes has a substantial weight benefit [1]. The cell capacity of an Ni/H_2 cell is independent of the number of hydrogen electrodes since its active material is in gaseous state. Therefore, a reduced number of hydrogen electrodes and other auxiliary components such as separators and gas distribution screens is needed for a thick nickel electrode cell, thereby reducing the cell weight and its cost. To take advantage of these benefits of using thick nickel electrodes, it is important to know quantitatively the rate capability of the electrode as a function of electrode thickness. Present report describes the results of a study on the rate capability of a sintered-type nickel electrode of various thicknesses up to 4.6 mm.

2. Experimental

Nickel sinter plaques were prepared using a dry sinter technique in various thicknesses ranging from 1 to 4.6 mm. These plaques were made of carbonyl nickel powder (INCO type 287) and a 20×20 mesh nickel screen (fiber diameter: 0.008 inch). Test plaques were prepared by sintering in hydrogen atmosphere at 950 °C for 12 min. Impregnation of the active material on these plaques (about 9 cm×9 cm in size) was carried out at 70 °C in 50% aqueous alcoholic solution of 1.8 M $Ni(NO_3)_2$ and 0.18 M $Co(NO_3)_2$ using multiple current steps, except for plaque type A. Plaque thickness, porosity, and active material loading level are summarized in Table 1.

Boilerplate Ni/H_2 test cells having a stack configuration, see Fig. 1, were prepared using the nickel electrodes which were cut to a size of 3.7 cm×6.3 cm. These electrodes had an active area of 21.7 cm^2 excluding tab area. The H_2 elec-

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Table 1
Test plaque thickness, porosity, and active material loading levels

Plaque type (mm)	Thickness (mm)	Porosity (%)	Loading level (g/cm^3 void)
1.0	0.975	82.1	1.69
1.6	1.588	83.2	1.62
2.5	2.455	82.8	1.62
3.5	3.520	82.7	1.66
4.6	4.591	82.4	1.67

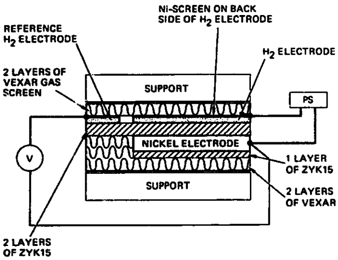

Fig. 1. A schematic setup of the Ni/H_2 test cell stack.

trode and separators (ZYK-15) were the same as those used in a standard Hughes/USAF Ni/H_2 cell [2], except for their dimensions. An aqueous 31% KOH solution was used as the electrolyte. In order to measure the nickel electrode potential free from polarization of the hydrogen electrode, a reference H_2 electrode was included in the cell stack.

In order to study the charge and discharge rate capabilities of these electrodes, capacities of the test cells were measured at various charge and discharge rates. For the charge rate effect, the capacity was measured at a constant discharge rate of $C/2$ to 0.5 V, after charging at various rates, e.g., at $C/10$ rate for 18 h, $C/2$ rate for 160 min, C rate for 80 min, or $2C$ rate for 40 min. For the discharge rate effect, the cell capacity was measured after a constant rate charging at $C/2$ for 160 min, by discharging them at various rates ranging from $C/5$ to $20C$.

3. Results and discussion

Chemical (or theoretical), rated, and measured capacities of electrodes of different thicknesses and their active material utilization are shown in Table 2 in order to demonstrate the effect of the electrode thickness on the utilization. The chemical capacity was calculated from the total amount of active material loading assuming one electron transfer reaction for both nickel and cobalt hydroxides. The rated capacities were based on capacity values measured just after activation of the cell. The capacities in Table 2 are the average of two values measured after a few conditioning cycles by charging cells at $C/2$ rate for 160 min and then discharging at the same rate. The utilization is the ratio of these measured capacities over the chemical capacity. The utilization values indicated that the active material can be fully utilized up to 2.5 mm possibly to 3.5 mm thickness at a discharge rate up to $C/2$.

Discharge voltage curves of test cells containing the 0.98 mm thick electrode at various discharge rates are shown in Fig. 2. The discharge voltage curves showed a characteristic shape of a well-behaved electrode with a single plateau and a sharp discharge knee, even at an extremely high rate of $20C$. With thicker electrodes, however, a second voltage plateau at about 0.8 V is observed at low discharge rates (C rate or lower). The voltage of this plateau was more or less independent of the discharge rate. Size of this second plateau increases as the electrode thickness increases. This second plateau is reminiscent of a similar plateau developed after long cycling of a thin electrode in a conventional Ni/H_2 cell [2]. At high discharge rates (roughly above $C/2$), an additional plateau [3] developed with thick electrodes at a voltage between the main plateau and the second one as shown in Fig. 3. When the discharge rate was increased to a $2C$ rate or higher, the third plateau voltage shifted below the second plateau voltage such that the second plateau disappeared under the third. This new third plateau occupied as much as about one half of the discharge capacity as shown in Fig. 4 for thick (2.5 mm or thicker) electrodes. The magnitude of this plateau increased as the electrode thickness and the discharge rate increased. The source of this new plateau is not well understood.

In order to estimate electrode polarization in terms of thickness, a half-discharge voltage was measured at various discharge rates. This half-discharge voltage at various discharge rates was defined as the cell voltage when the cell was discharged to one half of the measured value at $C/2$ rate to 0.5 V (see values in Table 2). Plots of the half-discharge voltage of individual cells against the discharge current densities gave

Table 2
Test cell capacities and electrochemical utilization of active material

Plaque type (mm)	Chemical capacity (Ah)	Rated capacity (Ah)	Measured capacity to 0.5 V		Measured capacity to 1 V	
			Cap. (Ah)	Util. (%)	Cap. (Ah)	Util. (%)
1.0	0.847	0.76	0.81	95.5	0.79	92.8
1.6	1.346	1.16	1.37	101.5	1.33	98.4
2.5	2.068	1.84	2.17	105.1	2.02	97.8
3.5	2.732	2.34	2.60	95.2	2.45	89.6
4.6	3.611	2.80	2.88	79.7	2.62	72.5

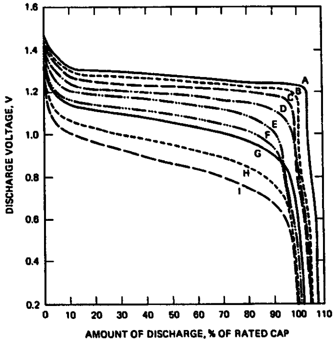

Fig. 2. Discharge voltage vs. H_2 electrode of a 0.98 mm thick nickel electrode at various discharge rates ($C/2$ to $20C$): (A) 0.5C; (B) 1C; (C) 2C; (D) 4C; (E) 6C; (F) 8C; (G) 10C; (H) 15C, and (I) 20C.

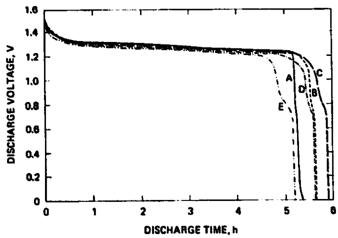

Fig. 3. Discharge voltage vs. H_2 electrode of nickel electrodes of various thickness (0.98 to 4.59 mm) at $C/2$ discharge rate: (A) 0.98 mm; (B) 1.59 mm; (C) 2.46 mm; (D) 3.52 mm, and (E) 4.59 mm.

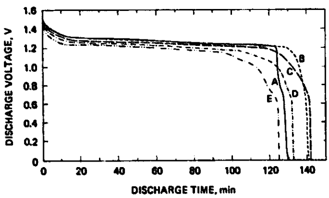

Fig. 4. Discharge voltage vs. H_2 electrode of nickel electrodes of various thickness (0.98 to 4.59 mm) at 4C discharge rate: (A) 0.98 mm; (B) 1.59 mm; (C) 2.46 mm; (D) 3.52 mm, and (E) 4.59 mm.

a straight line as shown in Fig. 5. The slope of this straight line, which represents the polarization of the nickel electrode, increased as the thickness increased. Information on the polarization values would be useful for future designing of a battery cell for high power application.

In order to evaluate the effects of charge rates on the electrode capacity, capacity values at $C/2$ rate discharge after charging at various rates are summarized in Table 3. Thin

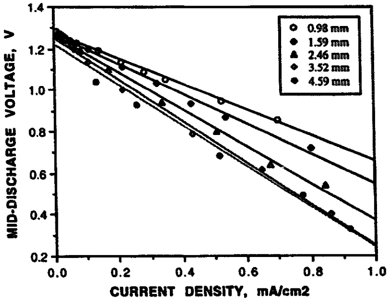

Fig. 5. Mid-discharge voltage vs. H_2 electrode of nickel electrodes of various thickness (0.98 to 4.59 mm) at various current densities.

Table 3
Effects of charge rates on electrode capacities. All capacities were measured at a $C/2$ discharge rate after charging the cells at various charge rates

Electrode thickness (mm)		Charge rates			
		0.1C	0.5C	1C	2C
1.0	a	0.73	0.79	0.84	0.81
1.6	a	1.22	1.32	1.31	1.25
2.5	a	1.99	2.02	1.81	1.57
3.5	a	2.24	2.45	1.9	1.51
4.6	a	2.67	2.62	2.36	1.63
1.0	b	0.76	0.81	0.85	0.83
1.6	b	1.31	1.37	1.36	1.32
2.5	b	2.16	2.17	1.94	1.73
3.5	b	2.46	2.6	2.05	1.7
4.6	b	3.18	2.9	2.72	1.98

a: discharged to 1.0 V.
b: discharged to 0.5 V.

electrodes (up to 1.6 mm) capacities were not affected significantly by the charge rate as observed earlier [4]. However, the capacities of thick electrodes (2.5 to 4.6 mm) decreased as the charge rate increased. The degree of the rate dependence increased as the thickness increased.

Capacity values at various discharge rates after charging at $C/2$ rates are summarized in Tables 4 and 5. The capacity values decreased as the discharge rate increased, as observed earlier, with the presently used thin electrodes [4,5]. This rate dependence increased rapidly as the thickness increased over 1.6 mm, particularly when the capacity was measured by a voltage cutoff value of 1.0 V instead of 0.5 V because the cell polarization increased rapidly as the electrode thickness increased. For example, electrodes thicker than 2.5 mm appear to have unacceptably large polarization for a low earth orbit (LEO) discharge rate. A nickel electrode with thickness up to 1.6 mm appears to be acceptable for application in an LEO satellite where the operation discharge rate is expected to be 1.37C or lower. Thick electrodes may be acceptable for low rate applications such as for a geosynchronous earth orbit (GEO) satellite.

Table 4
Effects of discharge rates on electrode capacities. All capacities were measured by discharging cells to 1.0V at various rates after charging the cells at a $C/2$ rate for 160 min

Discharge rate (C)	Capacity to 1.0 V (Ah)				
	1.0 mm	1.6 mm	2.5 mm	3.5 mm	4.6 mm
0.2	0.788	1.325	2.095	2.525	2.655
0.5	0.786	1.324	2.022	2.449	2.616
1.0	0.761	1.277	1.902	2.016	2.061
2.0	0.743	1.226	1.492	1.283	0.926
4.0	0.734	1.072	0.737	0.350	0.186
6.0	0.691	0.791	0.288	0.093	0.035
8.0	0.624	0.397	0.090	0.016	0.014
10.0	0.542	0.220	0.023	0.010	0.008
15.0	0.264	0.040	0.016		
20.0	0.084	0.020			

Table 5
Effects of discharge rates on electrode capacities. All capacities were measured by discharging cells to 0.5 V at various rates after charging the cells at a $C/2$ rate for 160 min

Discharge rate (C)	Capacity to 0.5 V (Ah)				
	1.0 mm	1.6 mm	2.5 mm	3.5 mm	4.6 mm
0.2	0.795	1.369	2.188	2.616	2.844
0.5	0.809	1.366	2.174	2.600	2.898
1.0	0.791	1.352	2.158	2.572	2.851
2.0	0.773	1.326	2.120	2.498	2.708
4.0	0.773	1.314	2.035	2.105	2.059
6.0	0.759	1.280	1.791	1.593	1.375
8.0	0.743	1.182	1.366	0.064	0.391
10.0	0.755	1.193	1.121	0.042	0.038
15.0	0.752	1.076	1.054		
20.0	0.738	0.978			

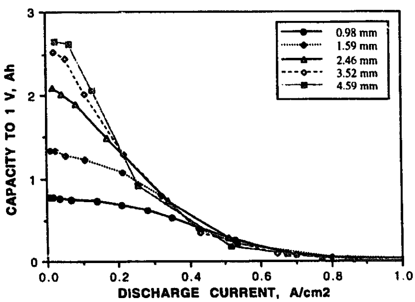

Fig. 6. Discharge current density vs. capacity (discharge to 1.0 V) of nickel electrode of various thicknesses.

The capacities of electrodes of various thicknesses versus current density at the geometrical surface of the electrode are plotted in Fig. 6; they appeared to form an asymptotic curve. This asymptotic curve shows a limitation in the maximum capacity that can be achieved at a given current density regardless of the electrode thickness. For example, the maximum capacity to 1.0 V measured at 0.3 A/cm^2 was about 0.8 Ah regardless of the thickness as shown in Fig. 6. The reason for this limitation may be that either the polarization of the electrolyte dominates or only the active material within a certain depth from the electrode surface can be discharged with the observed polarization. For example, this limiting capacity is 1.3 Ah for a 21.7 cm^2 electrode at 0.2 A/cm^2 and 0.9 Ah at a 0.3 A/cm^2 one regardless of the electrode thickness. This limiting value appears to indicate that the active material within 1 mm from the electrode surface is utilized, for example, at 0.3 A/cm^2.

4. Summary

In summary, the present results describe the rate (power) capabilities of nickel electrodes as a function of their thickness. The results provide a guideline for the maximum acceptable electrode thickness for a given power level. For example, a 1 mm thick electrode showed a well-defined discharge curve up to 20C rate discharge indicating that the maximum usable discharge rate may be higher than 20C. Electrodes up to 3.5 mm thickness showed sufficiently high rate capability for a GEO application, while the maximum practical thickness for an LEO application appears to be limited to about 2.5 mm. However, for a long cycle life application of the thick electrodes the stability of the rate capability with cycling needs to be studied since the rate capability of a nickel electrode is reported to be deteriorated with cycling [5].

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