



# Effect of local cell reaction at cathode on the performance of nickel metal-hydride battery



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## ABSTRACT

It has been widely known that nickel metal-hydride battery would degrade the capacity after shallow charge–discharge cycles. Whereas the formation of  $\gamma$ -NiOOH has been reported to degrade the capacity, its formation mechanism has not been clarified yet. We thought that local cell reaction between the cathode active material ( $\beta$ -NiOOH) and current collector (CC) results in the formation of  $\gamma$ -NiOOH to degrade the capacity. In the present study, we employed three types of CCs, i.e. gold, platinum and nickel meshes, to compare the phase change and capacity loss in terms of the local cell reaction. Only for nickel mesh CC, additional XRD peak around  $13^\circ$  in  $2\theta$  occurs accompanied by capacity degradation during charge–discharge cycles, while such phenomena do not for other CCs. This local cell reaction reduces  $\beta$ -NiOOH to transform into a  $\gamma$ -derivative which degrades the battery performance.

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## 1. Introduction

Nickel metal-hydride battery is taking a great attention as the most practical battery for the power sources of hybrid vehicles due to its relatively high discharge capacity and safety [1]. In addition, it is still important for the back-up power sources or portable devices. However, it has been well known that nickel metal-hydride batteries reduce the capacity after the shallow charge–discharge cycles [2–4], which is sometimes referred as memory effect [5–7]. Before the development of nickel metal-hydride battery, the widely used was nickel cadmium battery, in which the battery degradation due to shallow charge–discharge cycles has also been the critical issue [8–10]. Huggins ascribed the degradation of the nickel cadmium battery to the passivation of cadmium anode [11,12]. However, even after nickel metal-hydride battery with different anode (hydrogen absorbing alloy) had been developed, such a degradation has been still reported [7,13–15]. Consequently,  $\beta$ -NiOOH of the common cathode material of nickel cadmium battery and nickel metal-hydride battery has been thought to be the cause of the

degradation. Sato et al. ascribed the battery degradation to the formation of  $\gamma$ -NiOOH, which is an oxidant of  $\beta$ -NiOOH [16,17], at the interface between current collector (CC) and cathode active material ( $\beta$ -NiOOH) [5–7]. However, the detailed mechanism of the  $\gamma$ -NiOOH formation at the CC side instead of electrolyte side has not been revealed. We thought the local cell reaction would be the major factor of the transformation of  $\beta$ -NiOOH, since the reaction occurs between cathode active material and CC in the open circuit [18]. In the present study, we compared the phase formation and the cell performance by employing various metals as CC focusing on the local cell reaction between cathode active material and CC.

## 2. Experimental

### 2.1. Synthesis of cathode active material

$\beta$ -NiOOH of the cathode active material was synthesized by oxidizing nickel hydroxide (Aldrich Chemical Co., Inc) as the reported procedure [19]. 0.4 g of nickel hydroxide was mixed with 10 mL of distilled water to make a slurry, into which 10 mL of an aqueous solution of sodium hypochlorite was then added to oxidize nickel hydroxide. The solution was kept at  $20^\circ\text{C}$  for 1.5 h to proceed the reaction. The product material was thereafter separated by

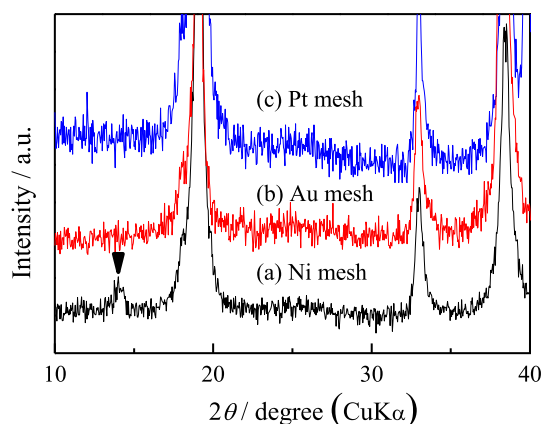
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vacuum filtration, washed by distilled water and dried for 1 day at 40 °C. After the processes above, black solid of  $\beta$ -NiOOH was obtained.

## 2.2. Charge-discharge cycles with rest time

The cathode was fabricated by mixing powder of the synthesized  $\beta$ -NiOOH as the active material, acetylene black as a conducting additive and PTFE as a binder at the ratio of 80:15:5 in weight. Platinum plate and Ag/AgCl electrodes were used as counter and reference electrodes, respectively. The electrolyte used was an 8 M potassium hydroxide. Three kinds of current collectors



**Fig. 1.** XRD patterns of cathode surface after the shallow discharge for Ex.1. (a), (b) and (c) represent the results for nickel mesh, gold mesh and platinum mesh CCs, respectively.

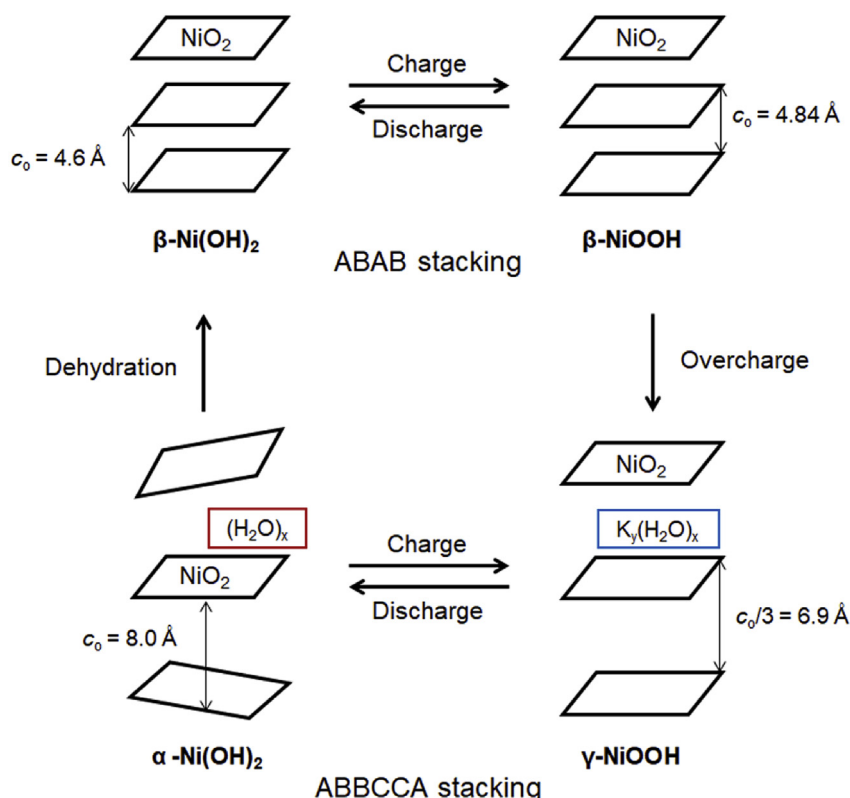
(CC) of nickel, gold and platinum meshes were employed to compare the local cell reaction with the active material. Focusing on the local cell reaction between the pure  $\beta$ -NiOOH and CC, we did not add zinc or cobalt hydride which is used for the commercial battery to improve the battery performance. For each CC, six kinds of charge-discharge cycles below (Ex. 1, Ex. 2 (A), (B), (C), and Ex. 3 (A), (B)) were conducted.

For Ex. 1, the cell was at first discharged to the cutoff voltage of 0.19 V (vs. SHE, Standard Hydrogen Electrode), followed by charging for 6 h to achieve the full charged state. The cell was then discharged for 50 mAhg<sup>-1</sup> of NiOOH, which is about a half of the cell capacity. After that, the circuit was opened for 1 day for nickel mesh and 3 days for gold and platinum meshes. We refer this open circuit period as rest time.

For Ex. 2, the cell was initially discharged to the cutoff voltage of 0.19 V (vs. SHE) and charged for 6 h, the cycle of which has been repeated for 3 times to stabilize the cell reaction. After the cycles of stabilization process, the charge-discharge cycle tests have been carried out. We selected three types of potential regions as (A) 20–40% of the state of charge (SOC), (B) 40–60% SOC and (C) 80–100% SOC. In these charge-discharge processes, 2 h of rest time were inserted every end of charge and discharge processes to investigate the local cell reactions occurring at the open cell circuit. Additionally, we conducted the charge-discharge cycle tests without rest time for 40–60% SOC region in Ex. 2(D) for the comparison with Ex. 2(B).

After the experiments of Ex. 2 (A) and (B), Ex. 3 was conducted for the degraded cells with nickel mesh CC. Full charge (6 h of charging) and discharge (down to cut-off voltage of 0.19 V vs. SHE) were carried out for 10 times to observe the recovery of the battery.

All of the charge-discharge cycles were conducted at a current density of 30 mA g<sup>-1</sup>.



**Fig. 2.** Bode's diagram for the structure relationship between  $\alpha$ -Ni(OH)<sub>2</sub>,  $\beta$ -Ni(OH)<sub>2</sub>,  $\beta$ -NiOOH and  $\gamma$ -NiOOH [17]. Diagram is schematically drawn based on the literatures [36,37].

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