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Reaction Mechanism of Layered Double Hydroxide as the Cathode Material of Nickel Secondary Battery

Noriyuki Sonoyama, Satoshi Yoshida, Takashi Inaba and Atsushi Nakayabu

(Department of Materials Science and Engineering, Nagoya Institute of Technology, Gokiso-cyo, Showa-ku, Nagoya 466-8555,Japan)

E-mail sonoyama@nitech.ac.jp

Introduction

Ni-hydrogen battery (NHB) has high energy density next to lithium ion battery. For this high energy density and safety, NHB is currently used for Hybrid electric vehicles and portable batteries. The weak point of this battery is limited theoretical capacity of its cathode electrode material: β-$Ni(OH)_2$. The reaction mechanism of cathode is expressed as follows.

$$\beta\text{-}Ni(OH)_2 + OH^- \Leftrightarrow \beta\text{-}NiOOH + H_2O + e^- \quad (1)$$

The theoretical capacity of reaction (1): 289 $mAhg^{-1}$ limits the energy density of NHB. To extend the energy density, much attention is payed to the following reaction that expand the theoretical capacity to 433 $mAhg^{-1}$.

$$\alpha\text{-}Ni(OH)_2 + OH^- \Leftrightarrow \gamma\text{-}NiOOH + H_2O + 1.5e^- \quad (2)$$

However, cycle stability of reaction (2) is low owing to instability of α-$Ni(OH)_2$. As the substitute of α-$Ni(OH)_2$, we focused on the layered double hydroxides (LDH) with the similar structure to α-$Ni(OH)_2$.

In the present study, we have examined the electrochemical property of Ni-Fe and Ni-V LDHs and its reaction mechanism was investigated using the ex-situ XRD and XAFS measurements.

Experimental methods

Ni-Al LDH was synthesized by a solvate thermal method [1]. Ni, Fe and V K-edge XAFS spectra were measured at BL-11S2 of Aichi synchrotron center.

Results and discussion

From the Ni K edge XANES measurement, it was confirmed that Ni ions in Ni-V LDHs and $Ni(OH)_2$ are oxidized to 3.5 valence state by charging to 0.5 V vs. Hg/HgO reference electrode. Ni-V LDH showed reversible capacity of 249 $mAhg^{-1}$ and its structure was sustained after 30 cycles of charge-discharge process, whereas the capacity of $Ni(OH)_2$ was 142.8 $mAhg^{-1}$and its structure degradated after 10 cycles.

From the EXAF spectra of these compounds, it became clear that the structural change of LDHs is reversible with small volume change, whereas that for $Ni(OH)_2$ indicates irreversible structural change from the first charge-discharge cycle with the 66% of volume change.

Conclusion

In the present study, it was demonstrated that LDHs show excellent performance as the cathode material of Ni rechargeable battery.

Reference

[1] Hai-Tao Xu, *et al*., *ACS Appl. Mater*., (2015) 20979-20986.