Research Article

Synthesis and Characterization of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) Cathode Materials for Lithium-Ion Batteries Prepared by a Sol-Gel Method

Hailang Zhang

College of Chemistry and Material Engineering, Jiangnan University, Wuxi 214122, China

Correspondence should be addressed to Hailang Zhang; zhl8868@vip.163.com

Received 29 May 2013; Accepted 13 November 2013; Published 17 February 2014

Academic Editor: Wen-Hua Sun

Copyright © 2014 Hailang Zhang. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Prospective cathode materials $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) for a lithium-ion secondary battery were synthesized using a sol-gel method. The structural and electrochemical properties were examined by means of X-ray diffraction (XRD), scanning electron microscopy (SEM), cyclic voltammetry (CV), and charge-discharge tests. The results show that the $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ maintains the $\alpha\text{-NaFeO}_2$ type layered structure regardless of the magnesium content in the range $x \leq 0.1$. On the other hand, Mg-doping improves the capacity retention well. Besides, the Mg-doping promotes the diffusion of Li^+ in $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$. Moreover, Mg-doping suppresses the phase transitions that usually occur in LiNiO_2 during cycling and improves the charge-discharge reversibility of $\text{Li}/\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$. High temperature cycling performance of the cathode at 55.5°C is also improved by Mg-doping, which is possibly attributed to the total stronger metal-oxygen bonding and the enhanced structure stability of those delithiated Mg-doped cathodes during cycling.

1. Introduction

Research in cathode materials for rechargeable lithium-ion batteries has a great deal of interest on the layered LiNiO_2 and LiCoO_2 materials in recent years [1–3]. Doped LiNiO_2 may possess better electrochemical properties than LiNiO_2 . Lithium nickel cobalt oxide [4–6] is becoming one of the most promising positive electrode materials for lithium-ion batteries. Lithium nickel cobalt oxide (LiNiCoO_2) for practical lithium-ion polymer battery production has become commercially available from several Japanese companies [7]. Recently, much attention has been focused on the development of $\text{LiNi}_{1-y}\text{Co}_y\text{O}_2$ ($0 \leq y \leq 1$) systems, which have strong potential for commercial application [8]. Of the various y values, $y = 0.2$ to 0.3 has produced the most interesting electrochemical properties [9]. $\text{LiNi}_{0.74}\text{Co}_{0.26}\text{O}_2$ has been synthesized via a solution route with excellent electrochemical properties [10] and Mg-doped $\text{LiNi}_{0.74}\text{Co}_{0.26}\text{O}_2$ with excellent electrochemical properties has been synthesized by a particular sol-gel method [11]. The reversible capacity of $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ tends to decrease during cycling and the

thermal stability needs to be improved. Lattices doped by Al [12, 13], Mg [14], Mn [15, 16], Mg/Ti [17], and so on were investigated to solve the problems. In fact, Mg^{2+} has no 3d state, yet the 3d state of Mn^{4+} is $3d^3(t_{2g}^3e_g^0)$, different from $\text{Cr}^{6+}3d^0(t_{2g}^0e_g^0)$.

It was reported that Fe and Mg substitute for Ni in $\text{LiNi}_{0.7}\text{Co}_{0.2}\text{Ti}_{0.05}\text{Mg}_{0.05}\text{O}_2$ [18] and $\text{LiNi}_{0.6-x}\text{Mg}_x\text{Co}_{0.25}\text{Mn}_{0.15}\text{O}_2$ [19] could reduce the cation mixing, improving structural integrity and cycle stability. Obviously, substitute for Ni in the Ni-based oxide appears to be a good method to modify the structural and electrochemical performance of these materials. In this study, we employed magnesium (Mg^{2+}) as a dopant due to the more negative Gibbs free energies of the MgO [$\Delta fG_{\text{MgO}}^\theta = -610 \text{ kJmol}^{-1}$] compared to those of NiO [$\Delta fG_{\text{NiO}}^\theta = -211.7 \text{ kJmol}^{-1}$]. Therefore, by substituting a part of Ni with Mg in layered $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ the total metal-oxygen bonding of the doped material is stronger than that of undoped one. Hence, I have synthesized a novel layered material of

$\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ by substituting a small amount of Ni with Mg. The structures and electrochemical properties of the compounds have been investigated in this paper.

2. Experimental

LiNO_3 (AR), $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (AR), $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$ (AR), and $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (AR) were used as starting materials of lithium, nickel, cobalt, and magnesium in $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$), respectively. LiNO_3 was initially dissolved in citric acid solution. The amount of citric acid is equal to the total molar amount of Co, Ni, and Mg. Then, $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ were added to the mixture. The whole mixture was heated by water bath at 80°C . During the heating process, a clear, pink solution without any precipitation formed. At last, the clear solution was slowly dried and turned into gel. The xerogel was dried, ground, and then heat-treated in an oven at 120°C for 12 h. The gel precursor was calcined at 500°C in air for 6 h, and cooled to room temperature in a tube-furnace. The heat-treated products were ground in an agate mortar to obtain powders. And then the powder was calcined at 800°C for 12 h.

The phase identity and crystal structure of the materials were investigated by measuring X-ray diffraction (XRD) using a D-8 X-ray diffractometer (Bruker, Germany). The surface morphology was observed by means of a scanning electron microscopy (SEM, SIRION, FEI) with Cu $K\alpha$ radiation at 40 kV, 70 mA.

For fabrication of cathodes, the prepared products were first mixed with acetylene black and polyvinylidene fluoride (80:8:12 in weight) in *N*-methylpyrrolidone (NMP). The slurry obtained was then coated onto Al foil and dried at 80°C for 18 h for further roll pressing. The electrochemical tests were done using CR2032 coin-type cells, which consist of the cathode, lithium foil as the anode, cegard 2325 as the separator, and 1 mol dm^{-3} LiPF_6 in ethylene carbonate (EC)-diethyl carbonate (DEC) (1:1 in volume, LB302 from Guotai Huarong, China). The active material of cathode is 10.5 mg for each cell. The cells were charged and discharged at room temperature or 55°C at 0.1 C in the voltage range of 2.0–4.4 or 4.6 V (versus Li/Li^+).

Cyclic voltammograms of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) were obtained by IM6 in the voltage range 2.0–4.8 V at a scanning rate of 0.1 mVs^{-1} . The reference and counter electrodes were fabricated from lithium metal.

3. Result and Discussion

Figure 1 shows the XRD spectra of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) powders. All peaks can be indexed based on a hexagonal $\alpha\text{-NaFeO}_2$ structure with a space group of $R\bar{3}m$. No impurity-related peaks are observed from the XRD patterns with Mg-doping. The peaks are sharp and well defined, suggesting that the compounds are well crystallized. The clear splitting of the peaks 006/012 and 108/110 indicates an ordered distribution of cations in the layered structure [1, 20]. The lattice constants, a and c , c/a , and $I(003)/I(104)$ of the $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) are shown in Table 1. From Table 1, one can see that with the Mg content

FIGURE 1: XRD patterns of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$: (a) $x = 0$; (b) $x = 0.05$; (c) $x = 0.1$.

increasing, the lattice constants a increase from 2.8547 to 2.8601 and c increases and then decreases. Larger ionic size of Mg^{2+} (0.660 Å) than Ni^{3+} (0.560 Å) might cause the increase in lattice parameters. The unit cell volume expansion caused by the increase of lattice parameters may assist the intercalation and deintercalation of Li ions during electrochemical processes. Meanwhile, c/a ratio is an indication of the cation ordering in the layered cathode materials [21]. For the synthesized $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$), all the c/a ratios are found to be higher than 4.9, indicating the higher cation ordering in all samples [21]. Besides, the ratio of $I(003)/I(104)$ is also used as an indicator for cation mixing in the structure of layered cathode materials [17]. The Mg-doped materials also present a little higher $I(003)/I(104)$ values than the undoped material. Similar to the change of c/a ratio, the increase for $I(003)/I(104)$ is not linear with the increase of Mg content and $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ shows the highest $I(003)/I(104)$ ratio. These results indicate that Mg-doping improves the characteristics of layer structure for $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ and $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ has the best layered characteristic.

Scanning electron micrograph was performed to characterize the approximative grain sizes and surface morphologies of the $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$). As shown in Figure 2, the undoped material appears to be homogeneous size distribution with submicron particle size. In fact, the particle feature of this material synthesized by sol-gel method is favourable for the intercalation-deintercalation reaction of electrode during the charge-discharge processes, and it is expected to deliver a larger capacity. A similar size distribution (about 240 nm) and morphology is observed for Mg-doped materials regardless of the Mg contents. This suggests that the Mg is well permeated into the bare $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ to form a solid solution, indicating that Mg ions are homogeneously dissolved into the colloidal precursor in the solution prior to following heat treatments for calcination. Besides, the smaller particle size of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ tends to improve the electrochemical performance of the electrode by

TABLE 1: Results of lattice parameters by XRD analysis of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$).

Sample	a (Å)	c (Å)	c/a	$I(003)/I(104)$	V (Å ³)
$\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$	2.8547	14.1360	4.952	1.313	99.76
$\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$	2.8593	14.1724	4.957	1.541	100.35
$\text{LiNi}_{0.6}\text{Mg}_{0.1}\text{Co}_{0.3}\text{O}_2$	2.8601	14.1594	4.951	1.486	100.31

reducing the ion diffusion pathway during Li^+ intercalation and deintercalation processes [11].

The electrochemical performances of the studied compounds have been evaluated at room temperature and high temperature, respectively. The charge-discharge curves of the studied phase appear to be quite monotonous indicating that no major structural transitions occur during the lithium extraction/insertion reactions.

Figure 3 presents the voltage versus capacity profiles of all doped and undoped $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ materials in the first charge/discharge cycle. During the first cycle, the cell efficiency was 91.3% for $x = 0$. The first coulombic efficiencies decrease with gradual substitution of Mg with Ni up to certain limit ($x = 0.1$).

Charge-discharge characteristics of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) were investigated by performing cycle tests in the range of 2.0–4.4 V. The initial discharge capacities for $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) are 178.9 ($x = 0.05$), 171.7 ($x = 0$), and 147.5 mAh g^{-1} ($x = 0.1$), respectively. The increase in specific capacity for $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ could be attributed to the enhancement of layered characteristics with Mg-doping. For LiNiO_2 and $\text{LiNi}_{1-x}\text{Co}_x\text{O}_2$ cathodes, a capacity loss always occurs at the initial charge-discharge cycle due to the cation mixing [22]. The sample $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ has less cation mixing so it has the less capacity loss. Therefore, it is easy to understand that $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ has the smallest capacity loss.

Figure 4 shows the charge-discharge curves versus cycle number during the first and the 20th cycles for $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ at C/10 rate in the 2.0–4.4 V range. The reversible capacity obtained for the first cycle is rather good ($>150 \text{ mAh/g}$) with an irreversible capacity loss of 2.1 mAh/g . As shown in the derivative curve, the Li extraction occurs at 3.7 V which is equal to that obtained in the case of LiNiO_2 [23]. This result confirms that previously obtained result by Han et al. [24] that Ni^{3+} is preferentially oxidized to Ni^{4+} compared to Co^{3+} .

To understand the effect of Mg-doping on the cycling stability of $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, the cycling performance of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) in the voltage 2.0–4.6 V at the 0.1C charge-discharge rate was investigated. The curves of discharge capacity versus cycle number for $\text{Li/LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) cell are shown in Figure 5. From Figure 5, it can be seen that the discharge capacity of bare $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ decreases to 158.1 mAh g^{-1} after 20 cycles with capacity retention of 87%. The cycling performance of Mg-doped materials are improved. For $x = 0.05$, the capacity retention after 20 cycles is 98.4%. Evidently, the cycling reversibility of $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ cathode is improved significantly by Mg-doping. The better capacity retention of Mg-doped $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ during cycling tests could be attributed to that the doped Mg assists maintenance

of the original layered crystal structure during intercalation-deintercalation of Li ions. In other words, the magnesium ions, with a size close to that of lithium, remain in the divalent state during cell charge-discharge. Therefore, their presence in the interslab space does not strongly affect lithium reintercalation because no shrinkage of the structure occurs upon cycling. This result explains how the magnesium-substituted phases have good cycling properties with enhanced capacity. The presence of an optimum concentration of magnesium ions in the lithium site prevents any local collapse of the interslab space during the deintercalation process. At a higher dosage of Mg^{2+} ions in the cathode material, the Ni^{2+} preferentially occupies the interslab spacing during the electrochemical process, which causes the low capacity delivery. On the other hand, the presence of Mg^{2+} ions in the lithium sites at the optimum concentration significantly reduces the usual changes observed in the cell parameters and accounts for good capacity retention [25].

The cycling discharge capacities for $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ in the different voltage regions are presented in Figure 6. The excellent cycling stabilities in both voltage regions show that small particle size and good surface morphology could be considered as good features to obtain this excellent electrochemical performances.

Figure 7 shows the discharge capacity curves for $\text{Li/LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) cell during the first 20 cycles at C/10 rate and at 55°C in the 2.0–4.4 V range. Thermal stability of the charged cathodes at 55°C is also improved by Mg-doping, which is possibly attributed to the the total stronger metal-oxygen bonding and the enhanced structure stability of those delithiated Mg-doped cathodes during cycling.

In order to investigate the effect of extra Ni^{2+} ions in the lithium site on the rate capability of this material, the cells were cycled at different current rates 0.1C, 0.2C, 0.3C, 1C, and 2C between 2.0 and 4.6 V. As shown in Figure 8, it appears clearly that this $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ material exhibits a good reversibility at different rates. Although the capacity decreases with increasing the rate, $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ could keep more than 96% of its capacity during 40 cycles for all the tested rates, even if for 1C or 2C higher rates. This confirms that lithium ion's diffusion in this material becomes easier.

Cyclic voltammetry (CV) was used to investigate the electrochemistry of the cathode materials $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$, as a function of Mg contents. The shape of the CV curve, peak potentials, and peak currents represents the electrochemical properties of the electrode and discloses the phase transitions that occur during charge-discharge experiments, which strongly affect the capacity fading during cycle [26]. In general, when a cathode experiences phase transformation,

FIGURE 2: Scanning electron micrograph obtained for $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$): (a) $x = 0.00$; (b) $x = 0.05$; (c) $x = 0.1$.

FIGURE 3: Initial charge-discharge curves of $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) at 0.1 C in 2.0–4.4 V: (a) $x = 0.05$; (b) $x = 0$; (c) $x = 0.1$.

a peak occurs in the CV curve due to the coexistence of two phases. During charge-discharge experiments, LiNiO_2 shows four different phases (one monoclinic phase, M, and three hexagonal phases, H1, H2, and H3) with three peaks in the CV curve. The three peaks in LiNiO_2 correspond to the coexistence of H1 and M, M and H2, and H2 and H3, respectively. LiCoO_2 also shows four phases (one monoclinic phase, M, and three hexagonal phases, H1, H2, and H3) in the CV curve [27, 28]. The three peaks in LiCoO_2 correspond to

FIGURE 4: Charge-discharge curves versus cycle number (1st and 20th) of $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ at C/10 rate in the 2.0–4.4 V range.

the coexistence of the phases H1 and H2, H2 and M, and M and H3.

In this research, cyclic voltammograms of bare $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ and Mg-doped $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ are compared in order to deduce the cause of the better capacity retention of $\text{LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$. The CV measured CV curves are presented in Figure 9. The curve clearly shows

FIGURE 5: Discharge capacity versus cycle number for $\text{Li/LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) cell at 0.1 C in 2.0–4.6 V: (a) $x = 0$; (b) $x = 0.05$; (c) $x = 0.1$.

FIGURE 6: Discharge capacities for a $\text{Li/LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ cell during the first 40 cycles at $C/10$ rate in the different voltage ranges.

the presence of three peaks at different voltage positions for $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, which arise from multiple phase transitions during charge-discharge cycling. The second peak in the CV curve for $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, while diminishing in intensity, merges with the first peak with increasing Mg content. The oxidation peak at 3.65 V almost does not change. Mg-doping suppresses the phase transitions that usually occur in LiNiO_2 during cycling and improves the charge-discharge reversibility of $\text{Li/LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$. The simplified CV curves and the peak shift are associated with the suppression of phase transitions due to superior maintenance of the layered structure after Mg addition.

FIGURE 7: Discharge capacity versus cycle number for $\text{Li/LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) cell at 0.1 C and 55.5°C in 2.0–4.4 V: (a) $x = 0$; (b) $x = 0.1$.

FIGURE 8: The specific discharge capacities versus cycle number of $\text{Li/LiNi}_{0.65}\text{Mg}_{0.05}\text{Co}_{0.3}\text{O}_2$ cells at different rate 0.1 C, 0.2 C, 0.3 C, 1 C, and 2 C in the voltage range 2.0–4.6 V ((a) = 0.1 C; (b) = 0.2 C; (c) = 0.3 C; (d) = 1 C; (e) = 2 C).

4. Conclusion

$\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ cathodes ($x = 0, 0.05, 0.1$) with sub-micron particles have been successfully synthesized by a sol-gel method calcinated at 800°C for 12 h. The structural and electrochemical properties have been systemically investigated to examine the effects of Mg-doping on initial discharge capacity and capacity retention. The results show that all the prepared $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ materials maintain the α - NaFeO_2 type layered structure regardless of the magnesium

FIGURE 9: Cyclic voltammograms for $\text{LiNi}_{0.7-x}\text{Mg}_x\text{Co}_{0.3}\text{O}_2$ ($0 \leq x \leq 0.1$) powders for $x = 0.00$ (a), 0.05 (b), and 0.10 (c).

content in the range $x \leq 0.1$. Mg-doping improves the capacity retention significantly. Besides, the Mg-doping promotes the diffusion of Li^+ in $\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$. Moreover, Mg-doping suppresses the phase transitions that usually occur in LiNiO_2 during cycling and improves the charge-discharge reversibility of $\text{Li}/\text{LiNi}_{0.7}\text{Co}_{0.3}\text{O}_2$. Thermal stability of the charged cathodes is also improved by Mg-doping, which is possibly attributed to the lowered oxidation ability and the enhanced structure stability of those delithiated Mg-doped cathodes during cycling. Furthermore, charge-discharge cycling at different rates reveals that Mg-doped material has rather excellent capacity retention and rate capability.

Conflict of Interests

The author declares that there is no conflict of interests regarding the publication of this paper.

References

- [1] J. Yamaki, M. Makidera, T. Kawamura, M. Egashira, and S. Okada, "Voltage prediction of nano-sized LiNiO_2 cathode for use in Li-ion cells," *Journal of Power Sources*, vol. 153, no. 2, pp. 245–250, 2006.
- [2] S. N. Kwon, J. Song, and D. R. Mumm, "Effects of cathode fabrication conditions and cycling on the electrochemical performance of LiNiO_2 synthesized by combustion and calcination," *Ceramics International*, vol. 37, no. 5, pp. 1543–1548, 2011.
- [3] L. Li, R. Chen, F. Sun, F. Wu, and J. Liu, "Preparation of LiCoO_2 films from spent lithium-ion batteries by a combined recycling process," *Hydrometallurgy*, vol. 108, no. 3-4, pp. 220–225, 2011.
- [4] D. Li, Z. Peng, H. Ren, W. Guo, and Y. Zhou, "Synthesis and characterization of $\text{LiNi}_{1-x}\text{Co}_x\text{O}_2$ for lithium batteries by a novel method," *Materials Chemistry and Physics*, vol. 107, no. 1, pp. 171–176, 2008.
- [5] K. Mukai, J. Sugiyama, and Y. Aoki, "Structural, magnetic, and electrochemical studies on lithium insertion materials $\text{LiNi}_{1-x}\text{Co}_x\text{O}_2$ with $0 \leq x \leq 0.25$," *Journal of Solid State Chemistry*, vol. 183, no. 7, pp. 1726–1732, 2010.
- [6] G. T.-K. Fey, R. F. Shiu, T. P. Kumar, and C. L. Chen, "Preparation and characterization of lithium nickel cobalt oxide powders via a wet chemistry processing," *Materials Science and Engineering B*, vol. 100, no. 3, pp. 234–243, 2003.
- [7] <http://www.seimichemical.co.jp>, <http://www.smm.co.jp>.
- [8] D. Li, Z. Peng, W. Guo, and Y. Zhou, "Synthesis and characterization of $\text{LiNi}_{0.9}\text{Co}_{0.1}\text{O}_2$ for lithium batteries by a novel method," *Journal of Alloys and Compounds*, vol. 457, no. 1-2, pp. L1–L5, 2008.
- [9] C.-C. Chang, J. Y. Kim, and P. N. Kumta, "Divalent cation incorporated $\text{Li}_{(1+x)}\text{MMg}_x\text{O}_{2(1+x)}$ ($M = \text{Ni}_{0.75}\text{Co}_{0.25}$): viable cathode materials for rechargeable lithium-ion batteries," *Journal of Power Sources*, vol. 89, no. 1, pp. 56–63, 2000.
- [10] J. Cho and B. Park, "Preparation and electrochemical/thermal properties of $\text{LiNi}_{0.74}\text{Co}_{0.26}\text{O}_2$ cathode material," *Journal of Power Sources*, vol. 92, no. 1-2, pp. 35–39, 2001.
- [11] J. Cho, "Li $\text{Ni}_{0.74}\text{Co}_{0.26-x}\text{Mg}_x\text{O}_2$ cathode material for a Li-in cell," *Chemistry of Materials*, vol. 12, pp. 3089–3094, 2000.
- [12] P. H. Duvinneaud and T. Segato, "Synthesis and characterisation of $\text{LiNi}_{1-x-y}\text{Co}_x\text{Al}_y\text{O}_2$ cathodes for lithium-ion batteries by the PVA precursor method," *Journal of the European Ceramic Society*, vol. 24, no. 6, pp. 1375–1380, 2004.
- [13] C. J. Han, J. H. Yoon, W. I. Cho, and H. Jang, "Electrochemical properties of $\text{LiNi}_{0.8}\text{Co}_{0.2-x}\text{Al}_x\text{O}_2$ prepared by a sol-gel method," *Journal of Power Sources*, vol. 136, no. 1, pp. 132–138, 2004.
- [14] C. Pouillier, F. Pertion, P. Biensan, J. P. Pèrès, M. Broussely, and C. Delmas, "Effect of magnesium substitution on the cycling behavior of lithium nickel cobalt oxide," *Journal of Power Sources*, vol. 96, no. 2, pp. 293–302, 2001.
- [15] D. D. MacNeil, Z. Lu, and J. R. Dahna, "Structure and electrochemistry of $\text{Li}[\text{Ni}_x\text{Co}_{1-2x}\text{Mn}_x]\text{O}_2$ ($0 \leq x \leq 1/2$)," *Journal of the Electrochemical Society*, vol. 149, no. 10, pp. A1332–A1336, 2002.
- [16] Y. Sun, C. Ouyang, Z. Wang, X. Huang, and L. Chen, "Effect of Co content on rate performance of $\text{LiMn}_{0.5-x}\text{Co}_{2x}\text{Ni}_{0.5-x}\text{O}_2$ cathode materials for lithium-ion batteries," *Journal of the Electrochemical Society*, vol. 151, no. 4, pp. A504–A508, 2004.
- [17] Y. Gao, M. V. Yakovleva, and W. B. Ebner, "Novel $\text{LiNi}_{1-x}\text{Ti}_{x/2}\text{Mg}_{x/2}\text{O}_2$ compounds as cathode materials for safer lithium-ion batteries," *Electrochemical and Solid-State Letters*, vol. 1, no. 3, pp. 117–119, 1998.
- [18] V. Subramanian and G. T.-K. Fey, "Preparation and characterization of $\text{LiNi}_{0.7}\text{Co}_{0.2}\text{Ti}_{0.05}\text{M}_{0.05}\text{O}_2$ ($M = \text{Mg}, \text{Al}$ and Zn) systems as cathode materials for lithium batteries," *Solid State Ionics*, vol. 148, no. 3-4, pp. 351–358, 2002.
- [19] P.-Y. Liao, J.-G. Duh, and H.-S. Sheu, "Structural and thermal properties of $\text{LiNi}_{0.6-x}\text{Mg}_x\text{Co}_{0.25}\text{Mn}_{0.15}\text{O}_2$ cathode materials," *Journal of Power Sources*, vol. 183, no. 2, pp. 766–770, 2008.
- [20] Y. J. Gao, Y. B. Chen, H. Q. Liu et al., "Structural characterization of layered $\text{LiNi}_{0.85-x}\text{Mn}_x\text{Co}_{0.15}\text{O}_2$ with $x = 0, 0.1, 0.2$ and 0.4 oxide electrodes for Li batteries," *Journal of Alloys and Compounds*, vol. 509, pp. 7915–7921, 2011.
- [21] T. Ohzuku, K. Nakura, and T. Aoki, "Comparative study of solid-state redox reactions of $\text{LiCo}_{1/4}\text{Ni}_{3/4}\text{O}_2$ and $\text{LiAl}_{1/4}\text{Ni}_{3/4}\text{O}_2$ for lithium-ion batteries," *Electrochimica Acta*, vol. 45, no. 1-2, pp. 151–160, 1999.

- [22] C. H. Chen, J. Liu, and K. Amine, "Symmetric cell approach and impedance spectroscopy of high power lithium-ion batteries," *Journal of Power Sources*, vol. 96, no. 2, pp. 321–328, 2001.
- [23] R. K. B. Gover, R. Kanno, B. J. Mitchell, A. Hirano, and Y. Kawamoto, "The role of nickel content on the structure and electrochemical properties of $\text{Li}_x(\text{Ni}_y\text{Co}_{1-y})\text{O}_2$," *Journal of Power Sources*, vol. 97–98, pp. 316–320, 2001.
- [24] C. J. Han, W. S. Eom, S. M. Lee, W. I. Cho, and H. Jang, "Study of the electrochemical properties of Ga-doped $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ synthesized by a sol-gel method," *Journal of Power Sources*, vol. 144, no. 1, pp. 214–219, 2005.
- [25] R. Sathiyamoorthi, P. Shakkthivel, S. Ramalakshmi, and Y.-G. Shul, "Influence of Mg doping on the performance of LiNiO_2 matrix ceramic nanoparticles in high-voltage lithium-ion cells," *Journal of Power Sources*, vol. 171, no. 2, pp. 922–927, 2007.
- [26] K. Dokko, M. Nishizawa, S. Horikoshi, T. Itoh, M. Mohamedi, and I. Uchida, "In situ observation of LiNiO_2 single-particle fracture during Li-Ion extraction and insertion," *Electrochemical and Solid-State Letters*, vol. 3, no. 3, pp. 125–127, 2000.
- [27] J. N. Reimers and J. R. Dahn, "Electrochemical and in situ x-ray diffraction studies of lithium intercalation in Li_xCoO_2 ," *Journal of the Electrochemical Society*, vol. 139, no. 8, pp. 2091–2097, 1992.
- [28] T. Ohzuku and A. Ueda, "Solid-state redox reactions of LiCoO_2 (R3m) for 4 volt secondary lithium cells," *Journal of the Electrochemical Society*, vol. 141, no. 11, pp. 2972–2977, 1994.