

# Modification of $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ by applying a surface coating of MgO

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## Abstract

$\text{Mg}(\text{OH})_2$  is coated on the surface of semi-crystalline  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . By further heating the coated sample, the surface is modified. This lowers the initial discharge capacity and improves the cycle-reversibility of a test lithium-ion cell, which was made with surface-modified  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  as the cathode material. © 2000 Elsevier Science S.A. All rights reserved.

**Keywords:** Lithium ion battery; Cathode material; Coating; Crystallization; MgO;  $\text{LiNiO}_2$

## 1. Introduction

Significant research effort has been directed towards the development of reliable energy sources for portable electronic devices. The lithium secondary battery has been the prime applicable candidate to date [1–3]. Already, lithium-ion batteries have captured a large share of the market. There is an ongoing search for new, improved materials for such batteries. For the cathode material, however, no material has surpassed the lithiated oxides of cobalt, manganese, and nickel [4–7]. Thus, much effort has been focused on the optimization of those three materials.

Among the lithiated transition metal oxides,  $\text{LiNiO}_2$  should have the highest theoretical specific capacity [8,9]. Its synthesis as a product with acceptable performance is, however, difficult. Batteries made with  $\text{LiNiO}_2$  as cathode material are known to have safety problems. Introducing Co into  $\text{LiNiO}_2$  facilitates the synthesis, though it lowers the specific capacity of the battery [10].  $\text{LiCoO}_2$  and  $\text{LiNiO}_2$  have the same crystal structure, and form complete solid solutions over the entire range of compositions. The two-dimensional layer structure of  $\text{LiNiO}_2$  is largely stabilized by the introduction of a small amount of Co [11]. The solid solution of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is mostly synthesized by means of a conventional solid-state reaction that involves heating a mixture of metal nitrates, carbonates, oxides or

hydroxides. It can also be synthesized by unconventional methods, such as the sol–gel method, [12] the peroxide-precursor method [13], and the PVA-precursor method [14,15].

For  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , synthesized by the PVA-precursor method, it has been demonstrated [16] that the morphology and electrochemical properties of the product can be tailored to a limited extent by controlling the synthetic conditions. In this study, we report an alternative way to change the properties of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  by surface modification.

## 2. Experimental

Powder samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  were synthesized via a conventional solid-state reaction which used hydroxide precursors.  $\text{Ni}(\text{OH})_2$  and  $\text{Co}(\text{OH})_2$  (Ni to Co mole ratio of 0.85 to 0.15) were mixed thoroughly by grinding in a mortar for 30 min.  $\text{LiOH}$  [ $\text{Li}/(\text{Ni} + \text{Co})$  mole ratio of 1.05] was added to the mixture and mixed with further grinding. This hydroxide precursor was heated at  $500^\circ\text{C}$  to  $600^\circ\text{C}$  for 10 h in a flow of dry air to obtain a semi-crystalline product of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . By adjusting the heating temperature ( $500^\circ\text{C}$  or  $600^\circ\text{C}$ ), the crystallinity of the semi-crystalline product could be altered to a limited extent. In some cases, the crystallinity was controlled by adding a small amount of an appropriate organic solvent to the hydroxide precursor prior to heating.

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The surface of the semi-crystalline  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  was modified by coating it with magnesia xerogel. Several grams of the semi-crystalline product was transferred to a fritted filter tube, and dried briefly under vacuum. In a separate vessel,  $\text{Mg}(\text{OMe})_2$  was prepared by reaction between magnesium turnings and dried methanol. The methanolic solution of  $\text{Mg}(\text{OMe})_2$  was placed over the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  in the fritted filter tube, such that the powder was soaked with  $\text{Mg}(\text{OMe})_2$  solution. After the excess solution had drained down the filter, the wet solid was dried under vacuum and the powder changed colour from black to light gray. By exposing the gray powder to ambient humid air, the  $\text{Mg}(\text{OMe})_2$  was transformed to  $\text{Mg}(\text{OH})_2$  by a hydrolysis reaction. When heated at  $200^\circ\text{C}$  to  $300^\circ\text{C}$ , the  $\text{Mg}(\text{OH})_2$  was transformed to  $\text{MgO}$  by a condensation reaction.<sup>1</sup> The  $\text{Mg}(\text{OH})_2$ -coated  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  was heated further at  $750^\circ\text{C}$  for 12 h to give the final product of crystalline  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  with a modified surface. This synthetic route should provide powder samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  with a heterogeneous distribution of Mg with depth. By adding Mg reactant at the beginning of the synthesis, samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  that were homogeneously doped with Mg were also synthesized to allow a comparative study.

Powder X-ray diffraction (PXRD) patterns were obtained by means of a Philips X'pert diffractometer. As an internal standard for obtaining accurate diffraction angles, silicon powder (Aldrich) was added to the solid sample. Elemental analysis was carried out by ICP/MS. Several coin-type lithium-ion test cells were fabricated with  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  as the cathode material. The cathode of the cell was obtained by cutting out a tape made from a mixture of the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , PVDF binder, and conductive activated carbon. The electrolyte was  $\text{LiPF}_6$  dissolved in 1:1 EC/DMC, and the anode was metallic lithium. In order to compare the effect of surface modification on cell performance, all the fabrication conditions were kept the same except for the type of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  used in making the cathode. Continuous charge–discharge cycles were performed under a constant-current mode. The changes in cell voltage during cycling were measured by using a battery operation system purchased from Toyo, Japan. Cut-off potential was 2.8 to 4.1 V. The cycle started at a rate of 0.1 C, and was stepwise raised to 1 C. The main concern was the high rate of 1 C.

### 3. Results and discussion

The synthesis of polycrystalline powders of surface-modified  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is shown schematically in Fig. 1. Metal hydroxides were used as reactants. LiOH was

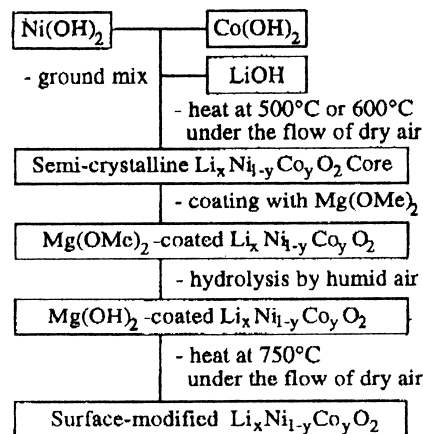


Fig. 1. Synthesis of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , with a surface modified by applying a coating of  $\text{MgO}$ .

added separately to a thoroughly grained mixture of hydroxides of nickel and cobalt. If added at the beginning, LiOH caused segregation of the mixture. By heating the hydroxide precursor in a stream of dry air above  $500^\circ\text{C}$ , a semi-crystalline core of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  was obtained as a single-phase product.

The sol–gel method was used to coat  $\text{Mg}(\text{OH})_2$  on the surface of the semi-crystalline  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . Because  $\text{Mg}(\text{OMe})_2$  in methanol readily reacts with water to form precipitates of  $\text{Mg}(\text{OH})_2$ , application of the coating was carried out under dried argon in a Schlenk line. If methanolic  $\text{Mg}(\text{OMe})_2$  had become turbid from  $\text{Mg}(\text{OH})_2$  precipitating out in solution, it was hard to keep a uniform application of  $\text{Mg}(\text{OH})_2$  over the surface of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . A second heat treatment was carried out at  $750^\circ\text{C}$  to ensure full crystallinity of its  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  phase.

The series of XRD patterns in Fig. 2 show the development of the crystalline phase of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , as the hydroxide precursor is heated at the designated temperature for 5 min. Below  $500^\circ\text{C}$ , it is evident that the reaction among metal hydroxides is incomplete in the given period, since the diffraction peaks for LiOH are still present, and those for  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  have not appeared. The peaks for  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  started to emerge around  $500^\circ\text{C}$ , and became sharper as the temperature was raised. Therefore, the single phase product of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is obtainable by heating above  $500^\circ\text{C}$ . From this observation, it is suggested that heating near  $500^\circ\text{C}$  to  $600^\circ\text{C}$  should cause a subtle difference in crystallinity of the single phase product. Such a difference is presumed to influence significantly the characteristics of  $\text{MgO}$ -coated materials, as described below.

The XRD patterns of semi-crystalline powders of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , which were heated at 500 or  $600^\circ\text{C}$  for 5 h, are compared in Fig. 3. The patterns contain all the major diffraction peaks for  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . No other phase is present. Compared with the sample heated at  $600^\circ\text{C}$  (Fig. 3a), the one heated at  $500^\circ\text{C}$  (Fig. 3b) has much broader peaks. Scanning electron microscopic obser-

<sup>1</sup> Verified by taking powder X-ray diffraction patterns from  $\text{Mg}(\text{OH})_2$  xerogel heated at the designated temperature.

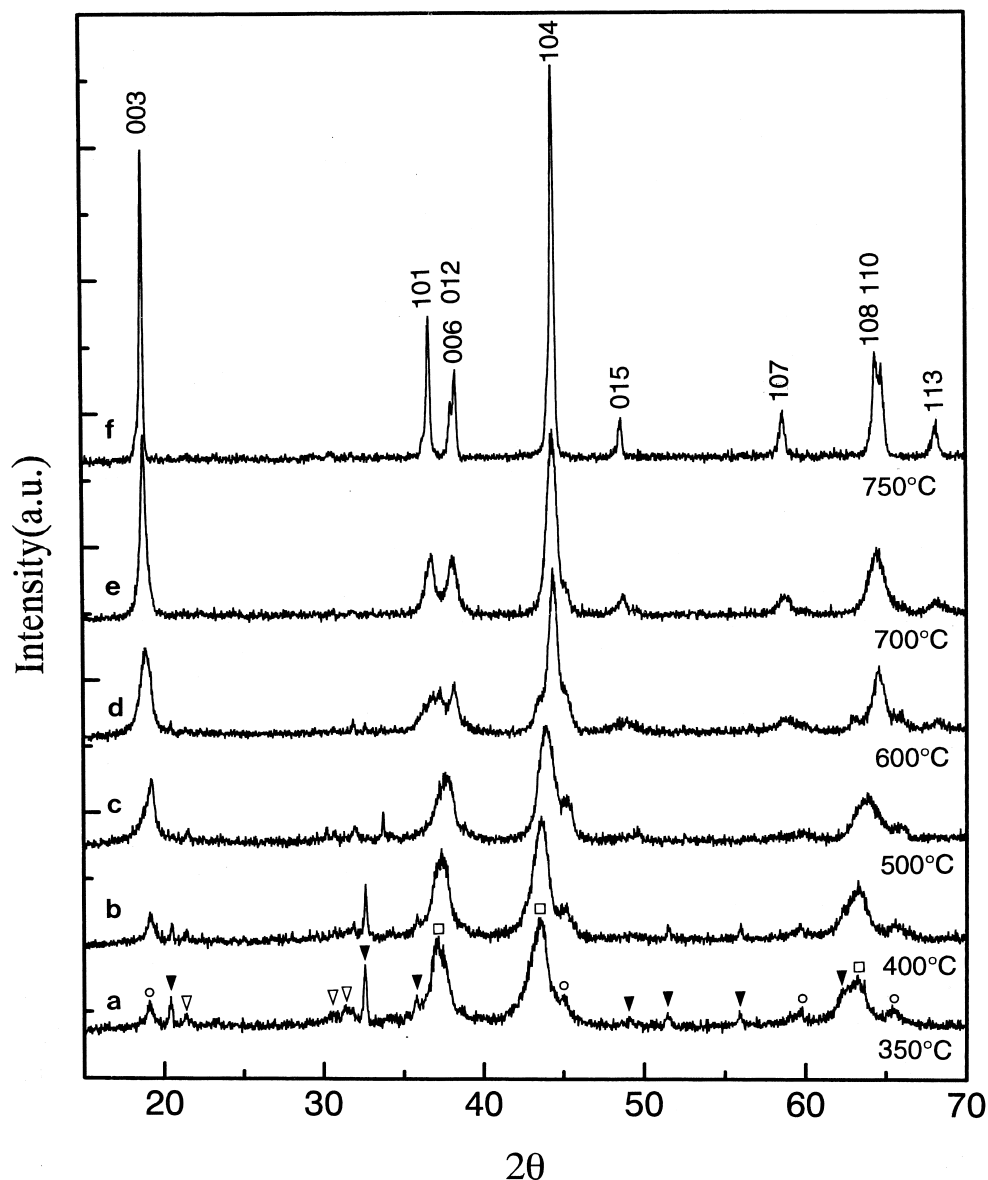


Fig. 2. XRD patterns in series for samples obtained by heating hydroxide precursor (at 3°C/min) at designated temperature for 5 min. □ = NiO, ○ = Co<sub>3</sub>O<sub>4</sub>, ▽ = Li<sub>2</sub>CO<sub>3</sub>, ▼ = LiOH.

variations record no difference in particle size. Therefore, the difference between the two patterns indicates a subtle difference in grain size (namely, crystallinity). The XRD patterns for Li<sub>x</sub>Ni<sub>1-y</sub>Co<sub>y</sub>O<sub>2</sub> with a surface modified by overlaying MgO are presented in Fig. 3. When 500°C-core is coated, small diffraction peaks develop upon heating at 750°C. These peaks are not those for Li<sub>x</sub>Ni<sub>1-y</sub>Co<sub>y</sub>O<sub>2</sub> (marked by closed triangles in Fig. 3e). The peaks are at  $2\theta = 37^\circ$ ,  $43^\circ$  and  $63^\circ$ . The position ( $2\theta = 37^\circ$ ,  $43^\circ$  and  $63^\circ$ ) and relative intensity of the diffraction peaks are nearly identical to those for MgO, MgNiO<sub>2</sub> and Mg<sub>0.4</sub>-Ni<sub>0.6</sub>O [17]. Therefore, it is difficult to assign the peaks to a particular phase. The mixed metal oxides, Mg<sub>1-x</sub>Ni<sub>x</sub>O, would be generated by a diffusion reaction between the

MgO coating and the underlying Li<sub>x</sub>Ni<sub>1-y</sub>Co<sub>y</sub>O<sub>2</sub>. The diffusive reaction would become more facile if one of the reactants was less crystalline. In order to get circumstantial evidence on the identity of the minor phase, Mg(OH)<sub>2</sub> was coated on a sample of Li<sub>x</sub>Ni<sub>1-y</sub>Co<sub>y</sub>O<sub>2</sub> whose crystallinity was very poor (compare Fig. 3b and c).<sup>2</sup> The broadness of the diffraction peaks in Fig. 3c indicates poor crystallinity of the core. Careful comparison between Fig. 3e and f suggested that those diffraction peaks at  $2\theta = 37^\circ$ ,  $43^\circ$  and  $63^\circ$  grew bigger when the core of the Li<sub>x</sub>Ni<sub>1-y</sub>Co<sub>y</sub>O<sub>2</sub> had

<sup>2</sup> Addition of acetone to the hydroxide precursor further lowered the crystallinity of the semi-crystalline powder obtained at 500°C.

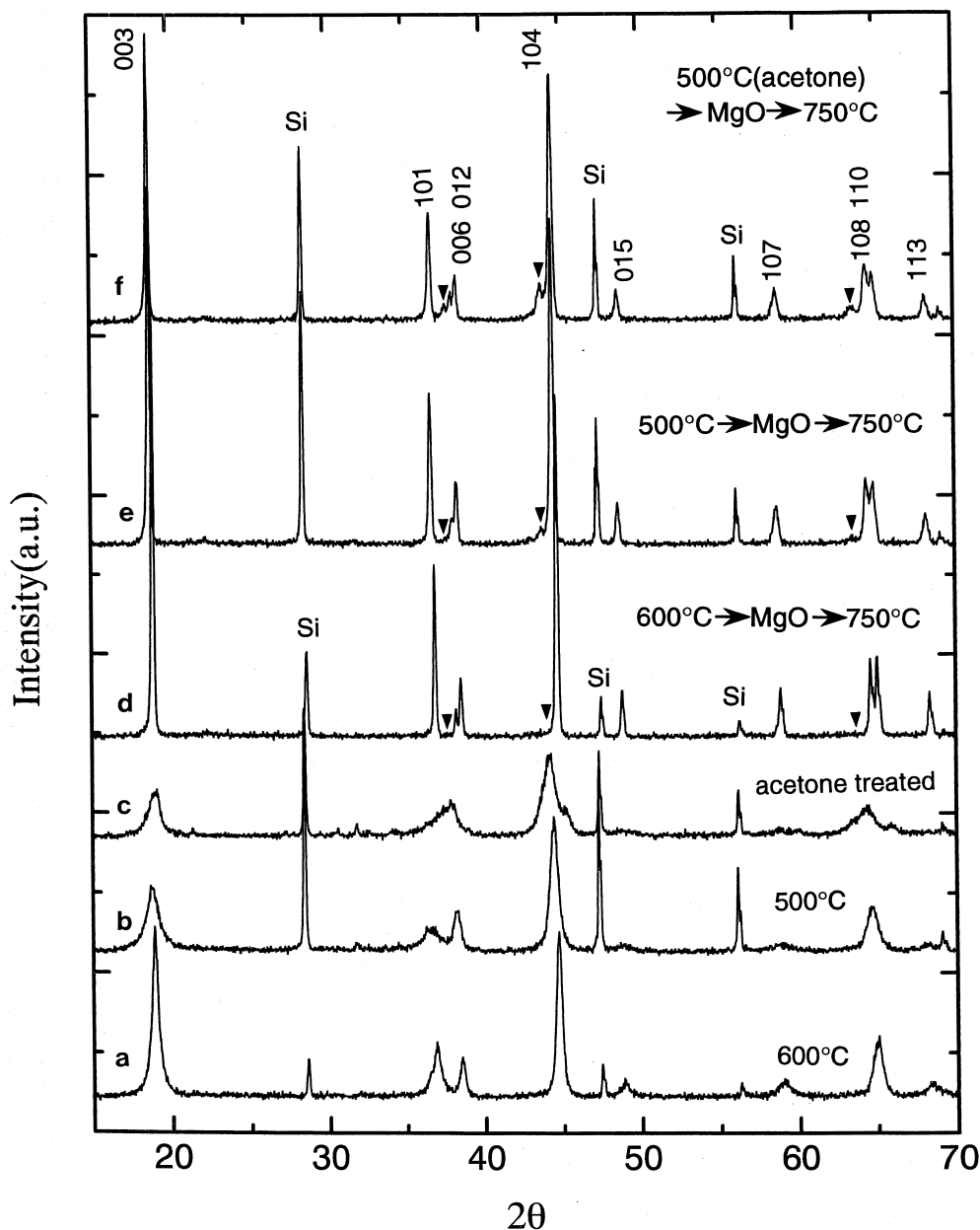


Fig. 3. XRD patterns for semi-crystalline core of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  which is prepared by heating hydroxide precursor at (a) 600°C, (b) 500°C, (c) at 500°C after addition of acetone. After coating  $\text{Mg}(\text{OH})_2$  on the samples, the surface is modified by further heating at 750°C. XRD patterns of fully crystalline powders of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , which are obtained by applying a coating on (d) 600°C-core, (e) 500°C-core, (f) acetone-treated 500°C-core peaks marked by closed triangles, show development of  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ .

poorer crystallinity. This observation suggests that those diffraction peaks relate to a mixed metal oxide of  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ , and not to  $\text{MgO}$ . If they were from  $\text{MgO}$ , the opposite trend would have been observed. Similarly, no diffraction peak was observed for  $\text{MgO}$  or  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$  when the 600°C-core was used (Fig. 3d). Apparently  $\text{MgO}$  does not diffusively react with underlying  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  and stays mostly at surface, presumably as nano grains.

The stoichiometry of the metal constituents, which was analyzed by ICP/MS, is given in Table 1. Because a relatively large quantity of the product is required to fabricate several test cells of the battery, different batches

(A and B) of the reactant had to be used. This caused small differences in the Ni to Co ratio between the batches. The

Table 1  
Stoichiometry of constituent metals in samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$

Sample	Processing			Stoichiometry			
	1st	MgO	2nd	Li	Ni	Co	Mg
Batch A	500°C (5 h)	no	750°C (12 h)	0.86	0.85	0.15	–
	500°C (5 h)	yes	750°C (12 h)	0.81	0.85	0.15	0.075
Batch B	600°C (5 h)	no	–	1.05	0.83	0.17	–
	600°C (5 h)	no	750°C (12 h)	1.01	0.83	0.17	–
	600°C (5 h)	yes	750°C (12 h)	1.04	0.83	0.17	0.048

Table 2

Cell parameters calculated from XRD patterns for surface-modified  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ .  $\text{Mg}(\text{OH})_2$  is coated on semi-crystalline core of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , which is prepared at designated temperature

Temperature of preparation (°C)	$\bar{a}$ (Å)	$\bar{c}$ (Å)
600	2.866	14.145
500	2.887	14.197

same batch was maintained in a given heating condition, however, in order to obtain consistent results on the effect of surface modification on Li stoichiometry. A previous study indicated that heating reactants in a stream of dry air was accompanied by lithium loss through evaporation [18]. For a given batch of reactant, the loss was observed to occur to the same extent for a given condition. By applying a coating of MgO, however, an additional change in lithium stoichiometry occurred. The additional change apparently occurred in the opposite direction, depending on the crystallinity of the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  core. When the first heat treatment was carried out at 500°C (core with lower crystallinity), a coating of MgO apparently caused more lithium loss to occur and thereby decreased the lithium stoichiometry down to 0.81. By contrast, when the first heat treatment was carried out at 600°C (core with higher crystallinity), lithium loss by evaporation was blocked by applying a MgO coating, and the lithium stoichiometry remained nearly unchanged at 1.04.

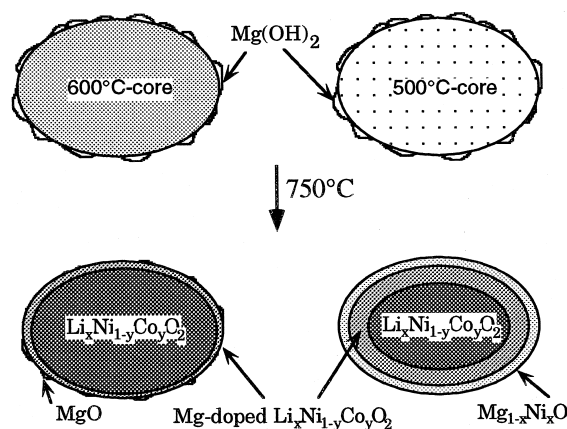
The above observations indicate that the coating of MgO influences significantly the crystallization process of the semi-crystalline core of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . When the crystallinity of the core is poor, it appears that Mg intercalates into the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  and replaces lithium near the surface, which is then lost by evaporation. During this process, the minor phase of de-lithiated  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$  is generated. By contrast, when the core is fairly crystalline, there is little diffusive intercalation of Mg. Instead, MgO on the surface actually blocks the escape of lithium by evaporation, and causes the lithium stoichiometry to stay unchanged during heating. Given these findings, it is worthwhile to revisit the XRD patterns in Fig. 3. It can be seen that the (108) and (110) diffraction peaks appear to merge as a minor phase of  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$  developed (compare Fig. 3d, e and f). This observation of broad diffraction suggests that the particles contain heterogeneous domains that exhibit a spectrum of interlayer distance. Given that Mg was provided externally, it was highly probable that the broadness of the diffraction is caused by a Mg gradient with depth.

Using only the peaks for  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , cell parameters were calculated and are provided in Table 2. The value of parameter  $c$  increases markedly when the sample is prepared by using a core of low crystallinity. Other workers have reported [19] that the value of parameter  $c$  increases significantly when  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is doped with

Mg [19]. Therefore, the increased value of the parameter  $c$  suggests that  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is doped with Mg which has originated from the MgO coating.

On the basis of the above experimental observations, a schematic of the cross-section of a particle of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is shown in Scheme 1. When MgO is applied to the crystalline core, the diffusion reaction between the core and MgO is minimal. On the contrary, when the crystallinity of the core is poor, MgO diffusively reacts with the underlying  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . The reaction generates  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ , which is devoid of lithium. During the process, some Mg intercalates into the lithium layer. This presumed scheme suggests that subtle differences in the crystallinity of the core can alter significantly the electrochemical properties of the cathode material when it is coated with MgO.

Crystalline powder of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  with a surface modified by MgO was used to fabricate test lithium-ion coin cells. All the manufacturing conditions were kept identical except for the cathode material. The electrochemical performance of the cells was investigated by measuring specific discharge capacity of the cells with cycling, Fig. 4. A continuous 100 cycles of charge–discharge was carried out mostly at a constant rate of 1 C. The initial discharge capacity at 0.1 C provides information on how much lithium is available, which is related to the amount of intercalated lithium in the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  (see Table 1). The slope of the curve at 1 C provides information on the stability of the two-dimensional layer structure of the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . If the structure is not stable, and collapses after lithium is de-intercalated, the amount of available lithium continues to decrease as the cycle continues. As can be seen in Fig. 4 and Table 3, when the surface is modified by a coating of MgO, the initial discharge capacity is decreased. At the same time, the slope of the curve is flattened, which indicates that the cycle-reversibility of the cell is improved. The decrease in capacity is exceptionally large when the crystallinity of the core is poor. Given that



Scheme 1.

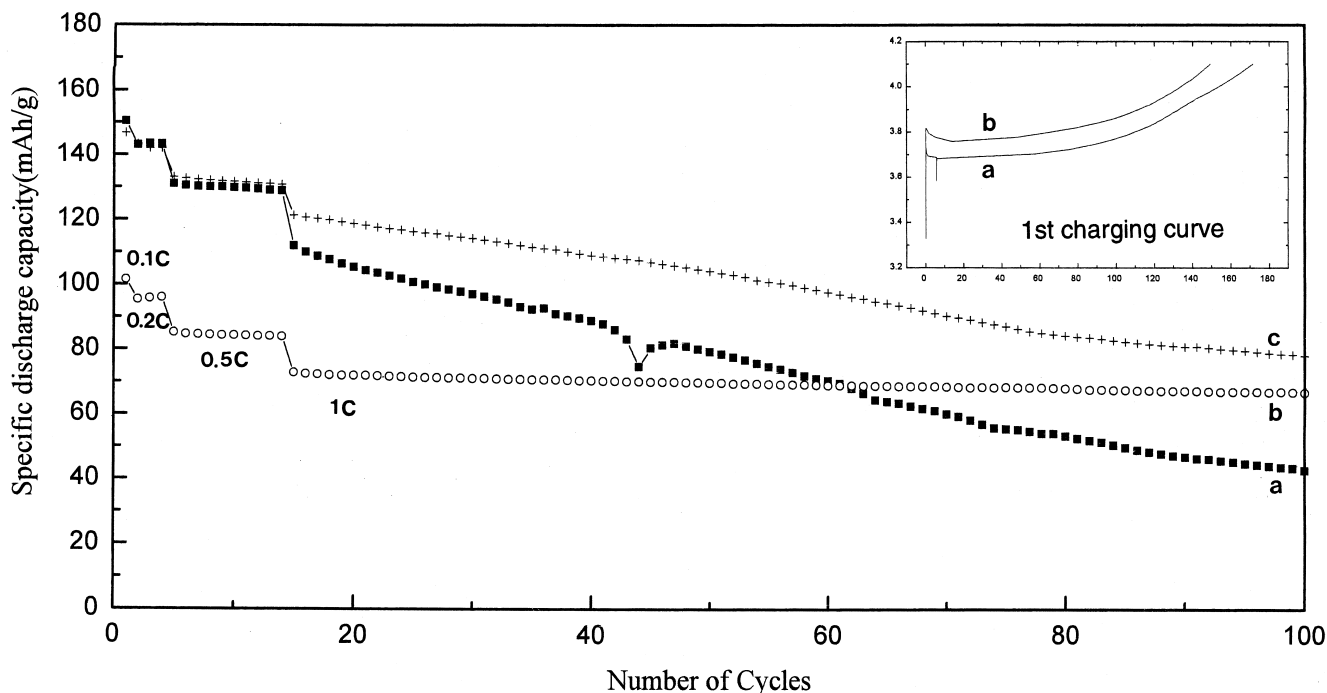


Fig. 4. Variation of specific discharge capacity of lithium-ion cell which uses a cathode made by using  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  with a modified surface. The battery is pre-conditioned by step-wise increase in current rate, from 0.1 to 0.5C. (a) Semi-crystalline  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , prepared by heating precursor at 500°C, further heated at 750°C without coating (pristine sample). (b) Semi-crystalline 500°C-core coated with  $\text{Mg}(\text{OH})_2$  and further heated at 750°C. (c) Same as (b) with 600°C-core. First charging curves are for (a) and (b) are compared in the insert. It shows that charging potential increases as the surface is modified.

the difference in crystallinity (which was qualitatively assessed by XRD patterns) is not that significant, the drop in capacity appears to be larger than anticipated. The initial charging curves for pristine and coated  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  are compared in the insert of Fig. 4. The increase in potential indicates that the surface resistivity is raised by applying MgO, probably because of the surface layer of  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ .

In order to determine whether the Mg dopant would bring about such a large drop in capacity, several samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  were doped homogeneously with Mg by adding the reactant from the beginning of the synthesis. The initial discharge capacities of the test cells, which were fabricated by using the Mg-doped  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  as cathode materials, are listed in Table 4. Apparently, the capacity drop is not linear with respect to the dopant level

and decreases to a large extent when even small amount of Mg is doped. The XRD patterns taken from doped samples do not contain any minor peaks that could be related to  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ . Therefore, the observed variation in the initial discharge capacity by applying a coating of MgO suggests that not only  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$  is generated, but also some Mg is doped into the lithium layer, i.e., Scheme 1. When the core of the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is obtained at 600°C, the drop in initial discharge capacity is very small. Even though the initial discharge capacity at 0.1 C is decreased (from 150 to 146 mAh/g on the first cycle) by surface modification, the capacity at 1 C is actually increased (from 112 to 121 mAh/g on the 15th cycle) as the result of the improved cycle-reversibility. These observations suggest that the performance of the cell can be tailored by adjusting the properties of the cathode materi-

Table 3

Electrochemical properties of test lithium-ion cell which is equipped with a cathode made by using surface-modified  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$

Sample	Initial capacity at 0.1 C (mAh/g)	Initial capacity at 1.0 C (mAh/g)	Capacity drop at 1.0 C (mAh/g)
(Not modified) 500°C → 750°C	150	112	−70
(Modified) 600°C → MgO → 750°C	146	121	−40
(Modified) 500°C → MgO → 750°C	103	73	negligible

Table 4

Initial discharge capacity of cells at 0.1°C for samples of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , which are homogeneously doped with Mg

$\text{LiNi}_{0.74}\text{Co}_{0.26}\text{Mg}_x\text{O}_2$	$x = 0$	$x = 0.01$	$x = 0.03$
Initial discharge capacity	173 mAh/g	156 mAh/g	149 mAh/g

als through control of the history of the heat treatment during surface modification by MgO.

#### 4. Conclusions

Applying a coating of MgO on the surface of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  alters the characteristics of the material, and thereby affects the electrochemical performance of a battery which uses the material as a cathode. The variation in the properties can be tailored to a limited extent by controlling the crystallinity of the  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  before it is coated with  $\text{Mg}(\text{OH})_2$ . When  $\text{Mg}(\text{OH})_2$ -coated  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  is further heated at 750°C, the reaction between MgO and  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  proceeds to a different extent as determined by the crystallinity of the core. As underlying  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$  becomes less crystalline, the reaction generates more of the minor phase, which appears to be  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ . With the generation of the minor phase, it is suggested that some Mg is doped during the reaction. Such modification of cathode material by a MgO coating has a marked influence on the electrochemical properties of  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ . Subtle changes in the crystallinity of the semi-crystalline core during the processing cause a relatively large variation in the performance of the battery. The generation of the minor phase of  $\text{Mg}_{1-x}\text{Ni}_x\text{O}$ , presumably near the surface, lowers the initial discharge capacity of the battery. At the same time, however, the cycle-reversibility of the battery is greatly improved. In view of the application, such surface modification could provide a means to tailor the properties of the battery.<sup>3</sup>

<sup>3</sup> Our prime goal is to improve the safety of the battery by modifying the surface of the cathode material.

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