Structural and Electrochemical Characterization of $Li[Co_{1-2x}(Li_{1/3}Mn_{2/3})_x(Ni_{1/2}Mn_{1/2})_x]O_2$ Solid Solutions

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$Li[Co_{1-2x}(Li_{1/3}Mn_{2/3})_x(Ni_{1/2}Mn_{1/2})_x]O_2$ (x=1,2,3, 4, and 5 for samples (a), (b), (c), (d), and (e), respectively) were synthesized using a sol-gel method. Shown in Fig. 1 are X-ray diffraction (XRD) patterns for the prepared $Li[Co_{1-2x}(Li_{1/3}Mn_{2/3})_x(Ni_{1/2}Mn_{1/2})_x]O_2$ samples with Miller indices. All peaks were indexed based on a hexagonal α-NaFeO2 structure with space group $R\bar{3}m$ (No. 166) except the supper lattice ordering peaks between 20° and 25°. The impurity peaks resulted from the short range ordering of Li, Co, Ni and Mn atoms on the transition metal layers, indicating a formation of Li_2MnO_3 structure. All samples showed the values of (003)/(104) intensity ratio higher than 1.2, indicating no cation mixing.

Figure 2 shows the capacity *vs.* voltage profiles of the fabricated $Li[Co_{1-2x}(Li_{1/3}Mn_{2/3})_x(Ni_{1/2}Mn_{1/2})_x]O_2$/Li cells, measured at a current density of 0.4 mA/cm^2 in a cut-off voltage range of 2.5 - 4.5V. All samples exhibit smooth and monotonous charge/discharge curves, although the discharge curve shape was a little bit different. In addition, the structure of the samples were not transformed from layered to spinel during the cycling. All samples except sample E delivered high discharge capacity without capacity fading. In case of sample E, discharge capacity gradually increased with cycle number. This unique phenomenon was intensively discussed on the basis of various experimental data. The samples (a), (b), (c), (d), and (e) delivered initial discharge capacities of 172, 178, 181, 178 and 126 mAh/g, respectively. The subsequent capacities of samples were 143, 165, 179, 190 and 172 mAh/g after 50 cycles, respectively.

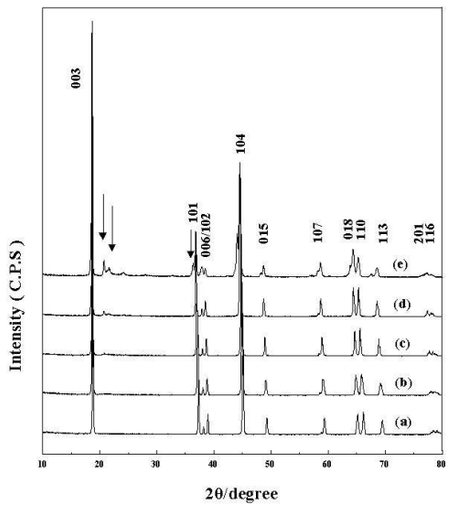

Fig. 1. XRD patterns of $Li[Co_{12x}(Li_{1/3}Mn_{2/3})_x\ Ni_{1/2}\ Mn_{1/2})_x]O_2$ powders.

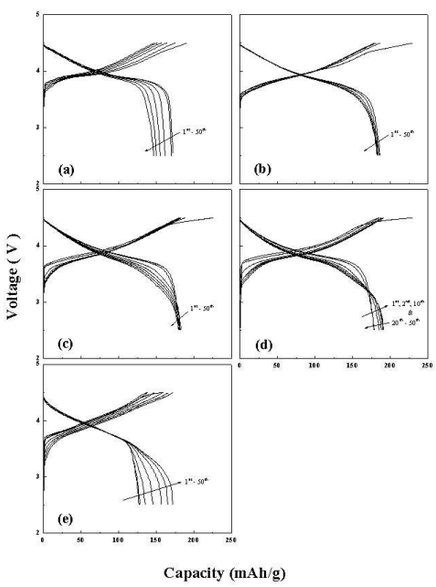

Fig. 2. The voltage as a function of capacity for Li/$Li[Co_{1-2x}(Li_{1/3}Mn_{2/3})_x(Ni_{1/2}Mn_{1/2})_x]O_2$ cells.