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Synthesis and characterization of $LiCo_{0.99}M_{0.01}O_2$ (M=Co, Mg, Zn, Mn, Al) by acid dissolution process

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During the last decade, various advanced chemical processes, such as sol-gel process, co-precipitation method, hydrothermal synthesis, ion exchange reaction, mechanical alloying, ultrasonic spray pyrolysis, reflux reaction, etc. have been tried to synthesize the high-performance cathode materials for the rechargeable lithium batteries. These solution-based processes are mostly able to produce materials with better homogeneity and higher electrochemical performance than the conventional solid-state reaction method, but there is still a need for better understanding of the effects of the different factors on the surface morphology, the chemical composition, and the crystallinity of the synthesized materials to commercialize these processes.

We have developed that new synthesis route called acid dissolution method is a promising process for preparing high-performance cathode materials for the rechargeable lithium batteries. And spray drying was introduced as a continuous mass production method of the precursor materials from the solution containing starting materials. In this method, insoluble starting materials such as metal carbonates or metal hydroxides are dissolved by strong organic acid, which also acts as a chelating agent. And then, the solvent of the solution containing starting materials is eliminated by spray drying or other drying method to obtain the xerogel of the initial solution, whose chemical form is expressed as $Li[MA_3]$, where M is transition metal atom, and A is the anion of the organic acid. The xerogel is then, calcined at the high temperature to obtain polycrystalline cathode materials.

In this work, the applicability of this method was demonstrated by synthesizing polycrystalline single phase $LiCoO_2$ and doped $LiCo_{0.99}M_{0.01}O_2$ (M=Co, Mg, Zn, Mn, Al) by the acid dissolution method using lithium carbonate, cobalt hydroxide, and dopant metal hydroxides as the insoluble starting materials and the acrylic acid as a chelating agent. The synthesized powders calcined at 800 ^oC showed high initial capacity and good cyclic performance in the half-cell test. We expect that similar procedure can be applied to synthesizing other types of cathode materials such as layered $LiNiO_2$, $LiNi_{0.8}Co_{0.2}O_2$, $LiCo_{1/3}Ni_{1/3}Mn_{1/3}O_2$, $LiNi_{0.5}Mn_{0.5}O_2$, spinel $LiMn_2O_4$, and so on. Since this method uses cost-effective starting materials and at the same time it has many advantages of wet process as well, we believe that powders synthesized by this method are to be economical as well as will show better electrochemical performance.

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Fig. 1. SEM image of powders synthesized at 800 ^oC

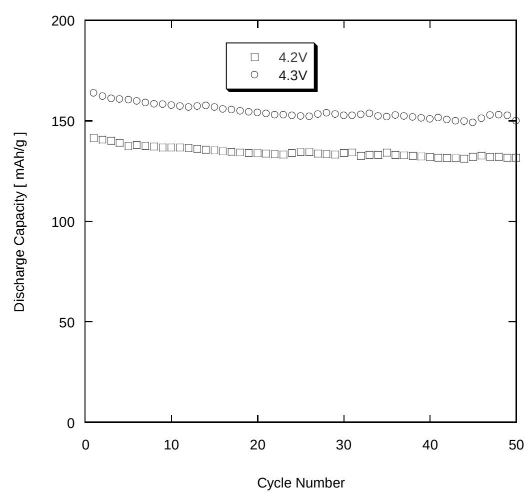

Fig. 2. Cyclic discharge capacities of the half cell, $LiCoO_2$/1M $LiPF_6$ in 1:1:1(v/v) EC/EMC/DMC/Li with voltage range of 3.0 ~4.2 or 4.3V, at 0.5 C rate, for the powders synthesized at 800 ^oC