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Improved Electrochemical Performances of NH₄AlF₄-coated LiCoO₂ Cathode Material for lithium ion battery

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Introduction

Layered LiCoO₂ electrode has higher reversible capacity, ease preparation, and excellent cycle stability within the voltage range of 3.0 - 4.2 V. Therefore, LiCoO₂ cathode material is typically used at the voltage up to 4.2 V (vs. Li); the specific discharge capacity is about 140 mAh g⁻¹, which is only a half of the theoretical capacity about 273 mAh g⁻¹. Because the amount of Li that can be extracted is limited to only 0.5 mol/mol of LiCoO₂, beyond which the layered rhombohedral structure collapses and gives way to an electrochemically inactive monoclinic phase. Also, higher potential than 4.2 V for LiCoO₂, a capacity fading occurred mainly due to the increase in the amount of cobalt dissolution. Improve the electrochemical properties is to coat on the cathode surface with metal oxide. A coating technology is very effective to enhance the electrochemical performances of cathode materials though the reason for the improvement effect is not fully understood yet. Extensive studies have been studied to search for new coating species and coating method. Recently, our group reported that AlF₃ coating on LiCoO₂, Li[Ni_{1/3}Co_{1/3}Mn_{1/3}]O₂, and Li[Ni_{0.8}Co_{0.15}Al_{0.05}]O₂ showed noticeably improved cycling performance and rate capability at a high cut-off voltage above 4.5 V vs. Li metal.[1] In this work, we study the effect of surface modification of LiCoO₂ by NH₄AlF₄ on electrochemical properties. Even the coating method was very simple, the performances of NH₄AlF₄-coated LiCoO₂ electrode were quite improved.

Experimental

For preparation of coating material, ammonium fluoride (NH₄F), and aluminum nitrate (Al(NO₃)₃) were separately dissolved in distilled water. The prepared ammonium fluoride solution was slowly fed into the aluminum nitrate solution [2]. The pH of the solution was controlled using NH₄OH for formation of (NH₄)₃AlF₆ phase. The mixed solution was constantly stirred for 30 min, and the prepared bulk coating powder was washed and filtered. Dried (NH₄)₃AlF₆ powder was heated at 300 °C for 5 h in air to form the NH₄AlF₄. A LiCoO₂ cathode material, NH₄AlF₄ bulk powder and zirconia ball were mixed and ball-mill for coating. The SEM images of the powders were observed using a scanning electron microscope (SEM, JSM 6400, JEOL, Japan). Charge-discharge cell test was done using a coin type half cell (CR2032) with Li-metal as the negative electrode. The electrolyte solution was 1 M LiPF₆ in ethylene carbonate (EC) - diethyl carbonate (DEC) in 3:7 volume ratio (Cheil Industries, Inc., Korea).

Results and discussion

Figure 1 shows the SEM images of pristine LiCoO₂ and NH₄AlF₄-coated LiCoO₂, respectively. From Fig 1a and 1b, it is clear that the surface of the particles with NH₄AlF₄. The NH₄AlF₄-coating layer of 30 - 50 nm was formed on the surface of LiCoO₂ that was observed. Discharge capacity of Li/pristine LiCoO₂ and NH₄AlF₄-coated LiCoO₂ cells with a current of 90 mA g⁻¹ (0.5 C-

rate). The pristine LiCoO₂ showed a gradual decrease in capacity, leading to capacity retention of 38% after 50 cycles in Fig 2a. Meanwhile, the NH₄AlF₄-coated LiCoO₂ had remarkably enhanced capacity retention of 75% after same cycling period in Fig 2b.

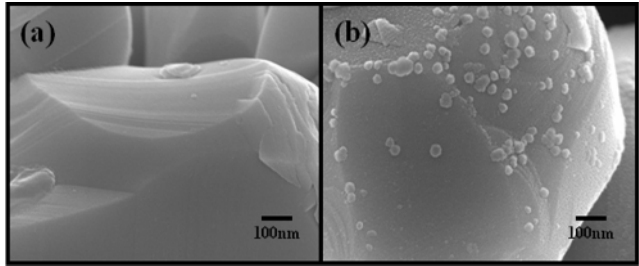


Figure 1. SEM image of (a) LiCoO₂ Pristine and (b) NH₄AlF₄ coated LiCoO₂.

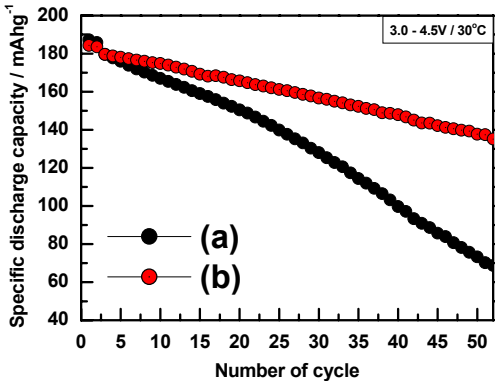


Figure 2. Cycling performances of (a) LiCoO₂ Pristine and (b) NH₄AlF₄ coated LiCoO₂.

Acknowledgements

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