Development of carbon porous electrode for Li-O₂/CO₂ battery using supercritical drying

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I. Introduction

Li-O₂/CO₂ battery is one of promising future batteries because it is superior to conventional Li-O₂ and Li-ion batteries with respect to CO₂ consumption [1]. In this battery, CO₂ which is a greenhouse gas is involved in discharge reaction and finally transformed into Li₂CO₃ discharge product thus environment and energy problem can be solved simultaneously. Most research on the cathode compartment of Li-O₂/CO₂ battery has focused on high surface area and light weight materials, such as carbon nanotubes, where a conventional evaporation method was applied to fabricate cathode composite. However, nanoparticles could aggregate during the drying process by evaporation, which reduces the pore size and decreases the gas dispersion capability of cathode.

Supercritical CO₂ drying which has nearly zero interfacial tension can be applied as drying method to fabricate free-standing cathode with porosity higher than 90% can be fabricated [2]. Moreover, supercritical CO₂ can be applied to impregnate ionic liquid (IL) into polymer binder by rediction of ionic liquid viscosity and expanding polymer matrix.

In this work, morphology and properties of electrodes fabricated from evaporation and supercritical drying along with effect of ionic liquid were investigated. Moreover, those fabricated electrodes were practically tested with O₂ and CO₂ flowing gas to investigate performance of battery.

II. Experimental

The cathode was prepared by dispersion of carbon black (CB), PVDF and MnO₂ in 1-methyl-2-pyrrolidone (NMP). This black viscous slurry was dried by 2 methods; which were evaporation at 80° C, 1 atm in ambient air and supercritical carbon dioxide (scCO₂) fluids at 40° C, 20 MPa. To fabricate porous electrode with polymer and ionic liquid (IL) binder, [bmim][Tf₂N] was added in black slurry and dried with scCO₂ at the same condition. The characteristics of cathodes were analyzed by SEM, DTG, porosity and conductivity measurement.

The as-prepared cathodes were assembled inside an argon atmosphere glovebox with humidity lower than 5% to create meshed coin cell batteries. Electrolyte used in this work was 1 M of LiTFSI in TEGDME. The initial discharge capacity and cyclability was tested with O2/CO2 mixing gases. Current density was set at 0.1 A.

III. Results and discussion

Morphology of cathodes was investigated by SEM which is showed in Fig. 1. The carbon particles in cathode dried by evaporation are clearly aggregated than cathode dried by supercritical drying. The porosity of cathode dried

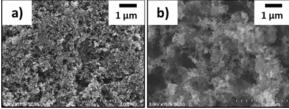


Fig. 1. SEM images of cathode dried by a) Evaporation and b) supercritical drying

by supercritical drying is 0.92 which is 1.77 times higher than cathode dried by evaporation. After applying ionic liquid in cathode, the porosity decreases to 0.83, however, sheet conductivity was 2.5 times higher. DTG analysis of cathodes shows that peak of cathode with ionic liquid binder shifts to lower temperature. This is the evidence that ionic liquid is impregnated inside polymer [3].

The initial discharge capacity of Li-O₂/CO₂ battery is shown in Fig. 2. It can be seen that battery using the cathode fabricated from supercritical drying gives higher discharge capacity than the cathode fabricated from evaporation. This can be considered that porosity of cathode affects discharge capacity due to increment of reaction gas diffusibility in cathode. Moreover, The cathode with polymer/IL binder gives highest discharge capacity at 15 mAh cm⁻², which is 3 times higher than cathode with normal polymer binder. This can be considered that cathode with polymer/IL binder gives higher ionic conductivity and reaction gas solubility than normal binder. Hence, surface area for discharge reaction increases after applying IL in cathode.

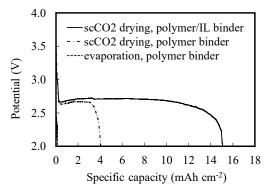


Fig. 2 initial discharge capacity of Li-O₂/CO₂ battery

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