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Induced electrochemical stability of quasi-solid-state electrolyte containing SiO₂ nanoparticles for Li-O₂ battery**Soonchul Kwon, Taeyoon Kim, Junebae Lee and Yongju Kwon**
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A stable electrolyte is required for use in the open-packing environment of a Li-O₂ battery system. Herein, a gelled quasi-solid-state electrolyte containing SiO₂ nanoparticles was designed, in order to obtain a solidified electrolyte with a high discharge capacity and long cyclability. We successfully fabricated an organic-inorganic hybrid matrix with a gelled structure, which exhibited high ionic conductivity, thereby enhancing the discharge capacity of the Li-O₂ battery. In particular, the improved electrochemical stability of the gelled cathode led to long-term cyclability. The organic-inorganic hybrid matrix with the gelled structure played a beneficial role in improving the ionic conductivity and long-term cyclability and diminished electrolyte evaporation. The experimental and theoretical findings both suggest that the preferential binding between amorphous SiO₂ and polyethylene glycol dimethyl ether (PEGDME) solvent led to the formation of the solidified gelled electrolyte and improved electrochemical stability during cycling, while enhancing the stability of the quasi-solid state Li-O₂ battery. We initially examined the morphology and appearance of the SiO₂ gellant PEGDME 500 electrolytes containing 5 wt.% SiO₂, as shown in Figures 1a and b. The transmission electron microscopy (TEM) image shows solid SiO₂ nanoparticles that are 10 nm in diameter. With an addition of 5 wt.% SiO₂ nanoparticles, the PEGDME 500 electrolyte becomes paste-like. We assembled the Li-O₂ battery cell by stacking the Li metal, electrolyte-soaked Celgard® film, Li-conducting LTAP solid electrolyte, cathode and gas diffusion layer, in order from the bottom to top, as shown in the schematic illustration in Figure 1d. All the components were pressed uniformly to form a Li-O₂ cell and the configuration. Using the assembled Li-O₂ cells, we compared the electrochemical performance of the gelled and conventional PEGDME 500 cathodes. The former yielded a significant specific capacity during the full discharge/charge process (Figure 2a). Note that the cells were maintained in a constant-current discharge with a current density of 500 mA·g⁻¹ and cutoff voltage of 1.8 V. A stable discharging plateau was observed at 2.67 V for both the cells. During charging, the cells were subjected to a constant current of 500 mA·g⁻¹ to 4.6 V, and maintained at 4.6 V until the current decreased to 10% of the initial current density. Interestingly, the first discharging capacity for the gelled cathode is higher by 13% (to 4600 mAh·g⁻¹) compared to the PEGDME 500 cathode (~4000 mAh·g⁻¹). From the cyclic performance results, it is worth noting that the gelled cathode delivers as much as 50% higher cyclability over its PEGDME 500 and 1000 counterparts (Figure 2d).

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