Li/LiPON/Li-conducting glass ceramic composite for Li-air batteries with non-aqueous electrolyte

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The highest specific energy density is the main attraction of rechargeable Li-air batteries. The best results for the Li-air battery were obtained when pure oxygen is used. However, in practical applications for electric vehicles, air containing moisture should be used. The exclusion of water from air is difficult using a conventional membrane filter, which makes the Li-air battery unsafe for use due to vigorous reaction between lithium metal and water. Thus, the protection of the Li-metal anode from water is the most critical issue for long term stability. The use of non-aqueous electrolyte can solve the problem of water from electrolyte. However, issue related to the water entering into the cell by means of air remains unsolved. Furthermore, the prominent problem for rechargeable lithium-air batteries is the lithium dendrite formation during repeated charge and discharge cycles; the dendritic Li-metal causes an internal short circuit. Li-metal can be protected by use of water stable NaSICON type lithium-ion conducting solid electrolyte. However, this solid electrolyte is unstable in contact with lithium. The interlayer needed between the lithium ion conductor and Li-metal, which should be stable with Li-metal and should have high lithium ion conductivity.

In this study, we have proposed the LiPON as an interlayer between the Li-metal anode and water stable solid electrolyte.

 B_2O_3 added $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$ (LAGP) glass ceramic was prepared by melt-quenching method. The LAGP glass ceramic crystallized for 5 h at 825 °C showed the highest conductivity of 6.9 x 10⁻⁴ S·cm⁻¹. The LiPON as an interlayer deposited on the prepared B_2O_3 added LAGP solid electrolyte by RF sputtering technique. The ionic conductivity of LiPON/solid electrolyte porecting layer was of the order 10⁻⁶ S·cm⁻¹ at room temperature. The AC impedance of Ag/B₂O₃ added LAGP/Ag, Li/ B₂O₃ added LAGP /Li, Ag/LiPON/ B₂O₃ added LAGP /LiPON/Ag, and Li/ LiPON/ B2O3 added LAGP /LiPON/Li were measured in frequency range of 1 Hz ~ 1 MHz at voltage amplitude of 100 mV at room temperature. The LiPON/Solid electrolyte showed good stability in contact with lithium metal. The AC impedance, cyclic voltammograms and charge-discharge performance measurements for the Li-O2 cell (Li/LiPON/B2O3 added LAGP/LiTFSI-TEGDME/Cathode) were also carried out. The reversible lithium oxidation and reduction was observed in Li-O2 cell. This cell exhibited a favorable discharge and charge performance, indicating good reversibility of the electrode reaction on the Li-metal electrode with a solid electrolyte, thus showing a good exchange reaction of the ions between the solid electrolyte and liquid electrolyte. Li-O2 cell exhibited the capacity of 1100 mAh·g⁻¹ at 0.2 mA·cm⁻² current density.



Fig.1. First charge-discharge performance of Li/LiPON/B₂O₃ added LAGP/LiTFSI-TEGDME/Cathode at 0.2 mA·cm⁻² and at 30 °C.