Role of the solid electrolyte interphase on a Li metal anode in a dimethylsulfoxide-based electrolyte for a lithium-oxygen battery

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Lithium-oxygen (Li-O_2) batteries are anticipated to be useful in large-scale energy storage because of their theoretically high energy density. To improve the cycling performance, the use of dimethylsulfoxide (DMSO) as a solvent has been widely studied in recent years. However, the Li metal anode is reported to be unstable in DMSO solvent due to formation of an unstable solid electrolyte interphase (SEI) on the lithium surface caused by side reactions during cycling. Therefore, a stable SEI should be formed on the lithium surface before cycling. However, a chemical structure of the SEI that is suitable for DMSO solvents has not yet been explored. This study examined the effects of SEI compounds on a lithium surface on the coulombic efficiency of a lithium anode in 1 M LiTFSI/DMSO electrolyte by following the procedure proposed by Koch et al. To prepare the lithium metal anodes with different SEI structures, excess amount of lithium (5.1C cm^{-2}) was electrodeposited onto a nickel substrate at 2 mA cm^{-2} in the various electrolyte solutions 1 M $LiClO_4$/EC-DEC, 1 M $LiPF_6$/EC-DEC or 1 M $LiNO_3$/DMSO. A part of the electrodeposited lithium (1.0C cm^{-2}) was dissolved/deposited during cycling in 1 M LiTFSI/DMSO at 2 mA cm^{-2}.

Fig. 1 shows the effects of main compounds on the SEI surface and interior on the coulombic efficiency of a lithium metal anode. The SEI's compounds were analyzed by XPS with and without Ar^+ sputtering. This result suggests that SEI compounds and their location strongly affect the coulombic efficiency of a lithium anode. Formation of inorganic compounds (Li_2CO_3, Li_2O and LiF) to only dominant proportions on the surface and in the interior of the SEI layer leads to enhanced coulombic efficiency (> 85%), while a lower efficiency was observed when organic compounds ($ROCO_2Li$ and polycarbonate) exist either on the SEI surface or its interior as the main constituents. Similarly, this enhanced coulombic efficiency by inorganic compounds was observed even when an O_2-saturated 1 M LiTFSI/DMSO was used for the cycling test. Also, the protective function of the inorganic SEI layer against H_2O constituent in the electrolyte was confirmed by the H_2O content of 1,000 ppm (Fig. 2). From this study, the lithium surface should be protected by inorganic compounds prior to cycling to prevent it from undergoing side reactions with the electrolyte and H_2O during cycling in the DMSO-based electrolyte.

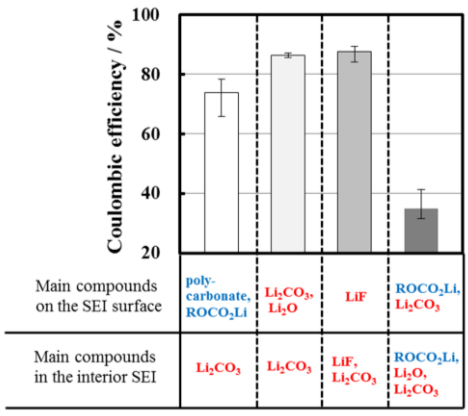

Fig. 1 Effects of main compounds on the SEI surface and interior on the coulombic efficiency of a lithium metal anode in 1 M LiTFSI/DMSO solution.

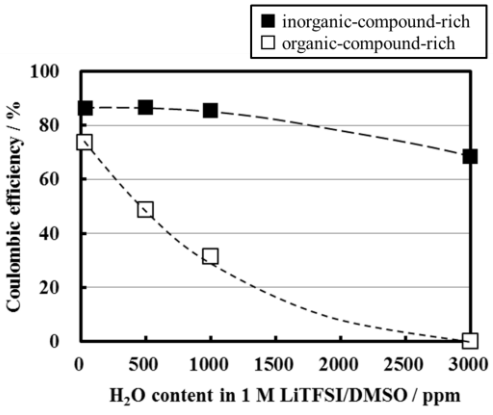

Fig. 2 Dependence of the coulombic efficiency of a lithium metal anode on the H_2O content of 1 M LiTFSI/DMSO electrolyte.

Reference:

[1] N. Togasaki, T. Momma, T. Osaka, *J. Power Sources*, **294**, 588-592 (2015).