

# 学位論文の要約

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学位論文題目				
Solid polymer electrolytes for a water stable lithium electrode in aqueous lithium air batteries (和訳 水系リチウム空気電池に用いる水に安定なリチウム電極用固体ポリマー電解質)				
主論文の要約 (図表・写真は除く)				
<p><b>導入 (Introduction)</b></p> <p>In recent years, lithium air and lithium sulfur batteries have attracted increasing attention in the worldwide academic and industrial fields, due to their far higher theoretical energy density. The use of lithium metals in both promising systems should address inevitable issues: a lithium dendrite formation and propagation, which could lead to the short-circuit and even the explosion of batteries. The underlying reasons for lithium dendrite formation are mainly related to the generation of spatially inhomogeneous and heterogeneous surface film on the lithium metal in contact with aprotic organic electrolytes with high reactivity and the inhomogeneous current distribution across the interface inducing the preferred lithium deposition on the special spots. Solid polymer electrolytes possessing a lower reactivity with lithium metal and a moderate mechanical strength have been considered to reduce the lithium dendrite formation.</p> <p><b>背景 (Background)</b></p> <p>It has been suggested that interfacial resistance between polymer electrolytes and lithium metal is a key factor in initiating lithium dendrite formation. Generally, conventional polymer electrolytes show the lithium dendrite formation for a short period at a higher current density of such as 1 mA cm<sup>-2</sup>. To suppress the lithium dendrite formation at a high current density, polymer electrolytes with additives, such as ionic liquids, nanofillers and oligomeric ethers, have been proposed and investigated in terms of electrical conductivity and ionic transport number of the polymer electrolyte, and interface resistance between lithium and the polymer electrolyte. However, the polymer electrolytes reported previously are not satisfactory.</p> <p><b>目的 (Objectives)</b></p> <p>This study is intended to correlate the cell resistance and the limiting diffusion current density with polarization performance. The ultimate purpose of this research is to provide guidance and suggestions for design and synthesis of polymer electrolytes that can suppress lithium dendrite formation and are suitable for protective layers in water-stable lithium electrodes (WSLEs).</p> <p><b>方法 (Methods)</b></p> <p>Effects of additive type and additive content on the electrical conductivity were examined by alternating current (a.c.) impedance method in a pouch cell of Au/CPEs/Au. Interfacial stability between polymer electrolytes and lithium metal was evaluated in a Li/CPEs/Li pouch cell in conjunction with electrochemical impedance spectroscopy (EIS). Effects of</p>				

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current density, additive types, thicknesses, and the combination of LTAP and polymer electrolytes on the lithium dendrite formation were studied by optical microscope, chronopotentiometry and EIS in a variety of cells, such as visualization cells, pouch cells and beaker cells. Electrode resistance and over potentials for lithium deposition and dissolution of WSLE were evaluated by different additives.

#### 結果 (Results)

Addition of an ionic liquid of PP13LiTFSI and low molecular weight oligomer ethers of PEGDME and TEGDME into the polymer electrolyte were found to be effective in reducing the resistance of the surface layer between lithium and the polymer electrolyte and suppressing the lithium dendrite formation. Addition of PEGDME and TEGDME into the polymer electrolyte was also revealed to reduce the resistance of the charge transfer reaction at the lithium electrode surface, and enhance lithium ion transport property significantly. The composite polymer electrolyte with low molecular weight of oligomer ether of TEGDME showed the best polarization performance. Limiting current density of the polymer electrolyte interlayer is demonstrated to being a dominant factor in influencing the lithium deposition and dissolution overpotential of WSLE at high current densities.

#### 考察 (Consideration)

We have attempted to investigate three impact factors on the lithium dendrite growth at the lithium metal and polyethylene oxide (PEO)-Li(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>N (LiTFSI) electrolyte interface: (1) interfacial resistance at the lithium metal interface, (2) chemical diffusion coefficient of lithium ion in the polymer electrolyte, and (3) thickness-dependence of the polymer electrolyte. The role of composite polymer electrolytes as the interlayer of (WSLE) in the Li/polymer electrolyte/LTAP/LiCl aqueous solution/Pt. air cell has been examined.

#### 結論 (Conclusion)

Addition of ionic liquid into PEO<sub>18</sub>LiTFSI reduced the resistance of the passivating layer formed at the interface. Addition of low molecular weight of oligomer ethers was found to not only reduce the resistance of passivating layer but also of charge transfer resistance at the interface between lithium metal electrode and the polymer electrolyte. Both additives can effectively reduce the passivating film resistance and lead to the homogenous distribution of current density across the interface and thus the impression of lithium dendrites.

Lithium electrodes protected by water-stable fast lithium ion conductor of LTAP and the polymer electrolytes have been investigated in terms of interfacial resistance, polarisation performance and cycling performance. The composite polymer electrolytes with the ionic liquids and oligomer ethers could reduce the dominant part of electrode resistance, interfacial resistance between lithium metal and polymer electrolytes. The most low and stable electrode resistance of 85 Ω·cm<sup>2</sup> was achieved by addition of TEGDME into PEO<sub>18</sub>LiTFSI.