



Design of new liquid electrolyte materials for next-generation rechargeable batteries

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Abstract

Lithium metal is an ideal negative electrode for rechargeable batteries. It has been studied for various next-generation battery concepts such as lithium metal batteries, lithium sulfur batteries, and lithium oxygen batteries. However, the high reactivity (strong reducing power) of lithium metal causes electrolyte decomposition, resulting in low charge-discharge efficiency. We demonstrated a new method to control the reactivity of lithium metal by electrolyte design. Based on this concept, we proposed several electrolyte materials that can weaken the reactivity of lithium metal and lead to high charge-discharge efficiency of over 99%.

Background & Results

Lithium-ion batteries are indispensable devices in our daily lives because they are widely used in smartphones, notebook PCs, household storage batteries, and electric vehicles (EVs). However, the energy density (stored energy per unit weight or volume) of lithium-ion batteries is reaching a theoretical limit, and various next-generation battery concepts are being studied to achieve much higher energy density. Lithium metal negative electrodes have been a central subject of research and development because the theoretical capacity of lithium metal (plating/stripping reaction: $\text{Li}^+ + \text{e}^- \rightleftharpoons \text{Li}$) is 3,860 mAh/g, which is about 10 times higher than that of graphite (372 mAh/g) used in commercial lithium-ion batteries. The major issue of lithium metal that hampers its practical application is low charge-discharge efficiency due to electrolyte decomposition caused by the high reactivity (strong reducing power) of lithium metal. It has been shown that charge-discharge efficiency strongly depends on the electrolyte formulations, but it is thought to be due to the difference in passivation films formed on the surface of lithium metal by electrolyte decomposition. There has been no quantitative discussion on how charge-discharge efficiency is dominated by electrolyte formulations.

In this study, we found that the reaction potential (reactivity or reducing power) of lithium metal differs significantly depending on the electrolyte formulations and that the charge-discharge efficiency is closely correlated with the reaction potentials. The charge-discharge efficiency of lithium metal tends to increase when its reaction potential is high (low reactivity or low reducing power) in a specific electrolyte formulation. In addition, we investigated the relationship between the reaction potentials of lithium metal and the liquid structures of electrolyte solutions. We found that the ion-pairing state of lithium ion and counter anion dominates the reaction potential. By designing the electrolyte with extensive ion pairing, the reaction potential of lithium metal can be increased, which in turn increases the charge-discharge efficiency. Based on the proposed mechanism, we proposed several electrolyte materials that achieved high charge-discharge efficiencies of over 99%.

Significance of the research and Future perspective

This finding provides a clear guideline for the electrolyte design of next-generation batteries that employ lithium metal negative electrodes and will greatly accelerate research and development toward their practical application. Replacing commercial graphite negative electrodes for lithium metal will increase the theoretical energy density of lithium-ion batteries by approximately 1.4 times. Furthermore, the energy density may be increased by several times when lithium metal negative electrodes are paired with next-generation high-capacity positive electrodes such as sulfur or oxygen. Such next-generation rechargeable batteries with high energy density are promising for use in EVs. They can achieve a cruising range comparable to those of gasoline cars.

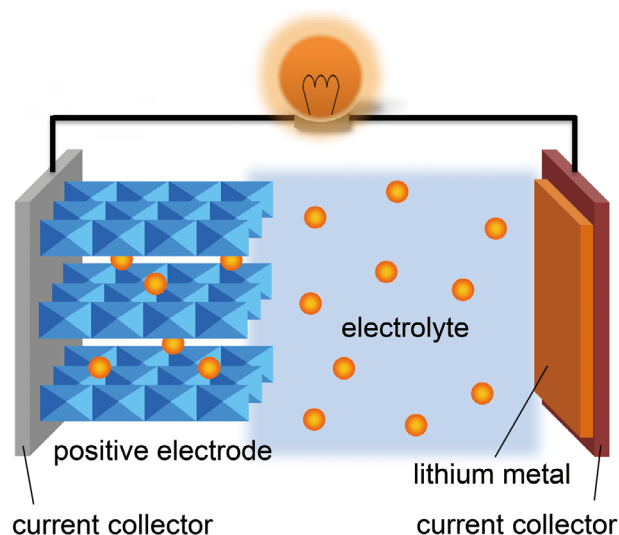


Fig.1

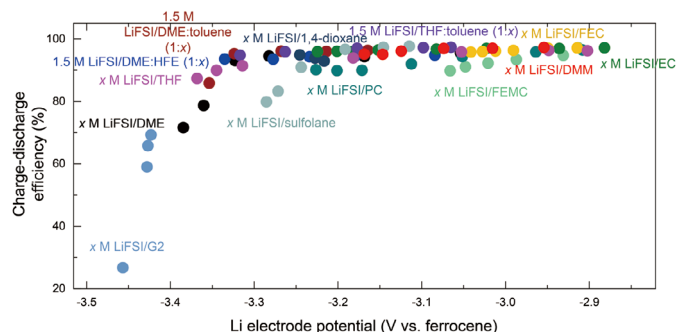


Fig.2

Patent Japanese Patent Application No. 2021-164244**Treatise**Ko, Seogjae; Yamada, Atsuo; Yamada, Yuki et al. Electrode potential influences the reversibility of lithium-metal anodes, *Nat. Energy*, 2022, 7, 1217,

doi: 10.1038/s41560-022-01144-0

Yamada, Yuki et al. Advances and issues in developing salt-concentrated battery electrolytes, *Nat. Energy*, 2019, 4, 269, doi: 10.1038/s41560-019-0336-z**U R L**<https://www.sanken.osaka-u.ac.jp/labs/eem/en/>**Keyword**

rechargeable battery, lithium metal, electrolyte, high energy density