

## Improvement of battery cycle characteristics by introducing a protective layer to Li(FSA)(SN)<sub>2</sub> composite electrolyte - LiCoO<sub>2</sub> positive electrode interface

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Solid-state Li<sup>+</sup> batteries, which utilize solid electrolytes instead of liquid electrolytes, have garnered significant attention as next-generation energy-storage devices. Nevertheless, solid electrolytes generally exhibit inferior performance compared with liquid electrolytes in terms of Li<sup>+</sup> conductivity and interfacial adhesion. We have focused on molecular crystal electrolytes, primarily considering their soft interfacial adhesion. We have reported the battery operation using Li(FSA)(SN)<sub>2</sub> molecular crystal, composed of lithium bis(fluorosulfonyl)amide (LiFSA) and succinonitrile (SN) (Li<sup>+</sup> conductivity of  $1.1 \times 10^{-4}$  Scm<sup>-1</sup>, Figure a)<sup>[1]</sup>. Furthermore, we recently found a higher conductivity of  $1.1 \times 10^{-3}$  Scm<sup>-1</sup> with a composite electrolyte composed of Li(FSA)(SN)<sub>2</sub> and 20 wt % SiO<sub>2</sub> [2]. However, Li<sup>+</sup> battery operation using this composite electrolyte has not been reported.

In this study, we report the performance of a battery using a composite electrolyte. The battery consisted of a Li foil negative electrode (0.1 mm), LiCoO<sub>2</sub> thin-film positive electrode (40 nm), and composite electrolyte pellet (0.3 mm). Cyclic voltammetry (CV) measurements revealed a decrease in the reaction current (Figure b). To prevent such battery degradation, we inserted a non-composite Li(FSA)(SN)<sub>2</sub> protection layer between the composite electrolyte and LiCoO<sub>2</sub>. The CV curves of the battery with protective layer overlapped over 10 cycles (Figure c). These results show that battery degradation originates from the Li(FSA)(SN)<sub>2</sub>-SiO<sub>2</sub> composite electrolyte | LiCoO<sub>2</sub> positive electrode interface and that the non-composite Li(FSA)(SN)<sub>2</sub> layer can suppress degradation.

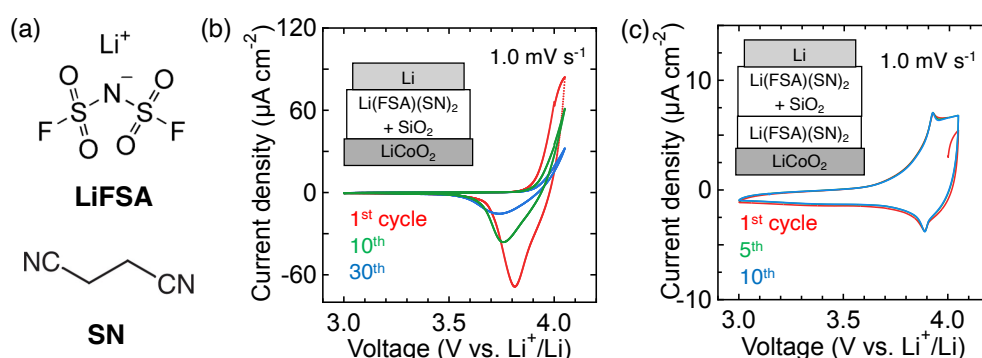


Figure. (a) Structural formula of lithium bis(fluorosulfonyl)amide (LiFSA) and succinonitrile (SN). (b) Cyclic voltammetry (CV) curves of the battery using Li(FSA)(SN)<sub>2</sub>-SiO<sub>2</sub> composite electrolyte. (c) CV curves of the battery with a Li(FSA)(SN)<sub>2</sub> protective layer.

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