

Electrochemical Properties of Thixotropic Gel Polymer Electrolytes Based on Poly(vinylidene fluoride-co-hexafluoropropylene)/Hydroxypropyl Cellulose Blend for Screen-Printing Process of 4V-Class Flexible Lithium Rechargeable Batteries

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Flexible lithium rechargeable batteries have received a lot of attention together with the rapid development of bendable or even foldable electronic devices such as flexible display, e-paper, foldable/bendable cellular phone, wearable PC, and so on. Recent efforts on unique and flexible battery designs are working towards advanced batteries that can be applied to devices and applications without limiting their form factors [1]. Recently, several groups have demonstrated promising results in the area of flexible lithium rechargeable batteries using various types of materials and processes [2-7].

Importantly, liquid electrolytes limit choices in cell design of flexible lithium rechargeable batteries due to their fluidic characteristics and the need of separators in cell assembly even though their excellent electrochemical performance and good physical contact with electrode [8].

This situation motivates us to develop self-supporting electrolytes paste that can be prepared on the electrode surface directly via roll-to-roll screen-printing process.

In this work, we have newly designed novel polymer electrolytes with thixotropy based on Poly(vinylidene fluoride-co-hexafluoropropylene)(PVdF-HFP)/Hydroxypropyl cellulose(HC) blend. The thixotropic gel polymer electrolytes pastes were composed of PVdF-HFP/HC blend, Al_2O_3 nanoparticles, and liquid electrolytes which is 1M LiPF₆ in EC/PC/EMC(1.5/1/1.5, w/w). To control the solvent evaporation time(less than 30min.) and medium viscosity(up to 50,000cP), NMP/Acetone(5/5, w/w) co-solvent was used. Adhesive gel polymer electrolytes of less than 30 μm thickness were prepared on the electrode surface throughout direct screen-printing. The maximum ionic conductivity is $3.6 \times 10^{-3}\text{S}/\text{cm}$ at 25°C and no significant decomposition of any components in the thixotropic gel polymer electrolytes takes place below 4.5V vs. Li/Li⁺.

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Acknowledgements

This research was supported by the Converging Research Center Program through the National Research Foundation of Korea(NRF) funded by the Ministry of Science, ICT & Future Planning(2012K001259) .

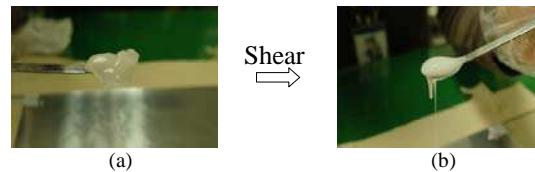


Figure 1. Thixotropic gel polymer electrolytes prepared:
(a) gel (before shear) (b) sol (after shear)



Figure 2. Thixotropic gel polymer electrolytes which is directly screen-printed on the electrode surface

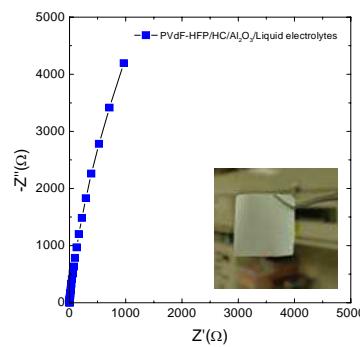


Figure 3. Casted film and impedance spectrum of thixotropic gel polymer electrolytes