

# Dual lithium-salts mediated lithium transfer in viscous cellulose acetate gel-polymer electrolyte

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Since gel polymer electrolytes (GPEs) have been introduced by Feuillade and Perche in 1975<sup>[1]</sup>, the studies on electrolytes based on biopolymers have been actively progressing due to the desirable properties of biopolymers and the concern on environmental issues. Furthermore, GPEs enable improved safety and mechanical stability as well as design flexibility, which meet the requirements for the automotive sector, medical health sector or portable electronics. The state-of-the-art GPE-based lithium-ion batteries (LIBs) are usually realized by mixing organic electrolytes in minimal amounts of polymer hosts, such as poly-(ethylene oxide) (PEO)<sup>[2-3]</sup> or poly-(vinylidene fluoride) (PVDF)<sup>[4]</sup>. This approach is pursued in order to have Li-ion transport properties as similar as possible to those of the liquid electrolyte fraction, while preventing leakage and improving the adhesion, *i.e.*, the separator and electrolyte assembly<sup>[5]</sup>. The utilization of current GPEs has proven to be competitive with liquid electrolyte-based LIB cells. For instance, Sony already introduced Li-ion batteries including GPEs on the market in 1999 under name ‘LiPo’ batteries. Yet there are concerns due to the fluorinated compounds and the biodegradability. Decomposition of the fluorinated compounds or contact with moisture can lead to the formation of toxic HF or organo-(fluoro)-phosphates<sup>[6]</sup>, which has an impact on the safety and cell performance, but also on the recycling cost. Additionally, the polymers are fossil fuel-derived and not biodegradable, therefore, the use of sustainable GPE alternatives is highly desired.

In this work, biopolymers become accessible by using solely organic solvents without the usage of water or acid in order to prevent impurities in the product and avoiding side reactions. Further, this approach enables an improved control on the reaction path. The designed gel-biopolymer electrolytes are investigated within laboratory-scale cells in terms of their reductive and oxidative stability, as well as under practicable commercial conditions in LIB full-cells. In particular, this work includes the investigation of a natural bio-derived GPE (cellulose-derivative), which is designed to combine different functional properties to enhance the battery cell performance, and to increase the safety of the current LIB technology.

## References:

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