

TAILOR-MADE HIGH VOLTAGE STABLE HYBRID FLUOROPOLYMER ELECTROLYTE MATERIALS The widespread use of Ni-rich active cathode materials for next-generation lithium-ion batteries is strongly connected to the development of high-voltage stable electrolyte materials, as a way to avoid fast aging of the battery. Fluoropolymers are well known for their high voltage stability but need to be engineered smartly to meet the requirements of battery applications.

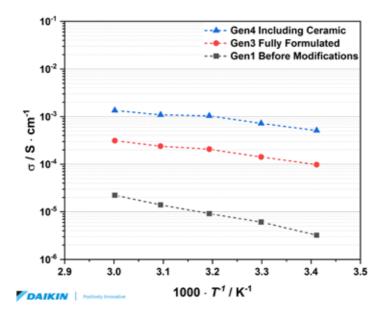
Polymer electrolytes are a key component of modern all-solidstate batteries. They not only make batteries safer, contributing to their inherent non-flammability, but also enable the use of key materials to increase the battery energy density. One class of these key materials is Ni-rich NCM cathode active material. By reducing the cobalt content of the NCM materials, it is possible to simultaneously improve the energy density and improve battery costs and sustainability. However, the consequence of using this material is a higher cell voltage. Current polymer electrolyte systems lack the necessary high-voltage stability and their degradation leads to very fast aging and poor cyclability of the battery

Fluoropolymer-based materials, which are well known as high voltage stable materials, do often lack other crucial performance values if they are to be efficiently used as electrolyte systems. Moreover, the complexity and variety of active materials, investigated and used for the mentioned high-voltage cathodes, pose an additional aspect to be considered. The electrolyte systems need to be flexible in their parameters and their implementation into various concepts. Our solution presents a hybrid electrolyte approach, which uses a high voltage stable, plasticized fluoropolymer electrolyte material as a matrix and host for a lithium-ion containing salt and various ceramic additives as performance enhancers.



As observable in the previous figure, high ionic conductivity is detrimental to the polymer electrolyte materials. In our development work, we modified the polymeric matrix and subsequently added various components to this modified polymer electrolyte matrix. The result is an easy-to-process, easyto-adapt, and fully tailorable set of materials, which can be used in many different concepts and in combination with many materials.

Of course, one of the main aspects of the electrolyte matrix is the ionic conductivity of the material. This property is very important to ensure a smooth operation of the battery system and to enable fast charging



and discharging processes. Going through several generations, we were able to obtain materials with best-inclass values for their ionic conductivity. Unusual values for fluoropolymers were obtained by tailoring the saltto-plasticizer-to-matrix ratio and in particular through ceramic additives, which enable a substantial increase in conductivity even when used in small fractions.

Of course, all developments were focused on true solid electrolyte concepts. And throughout the complete modification and formulation, the work performed on the highvoltage polymer electrolyte materials mechanical and thermal stability of our platform was monitored very closely. In this way, there is no need to trade off higher battery performance in terms of ionic conductivity with higher battery safety. Our developed electrolytes can be used as free-standing membranes or can also easily be cast from various non-toxic solvents. This allows a direct drop in-solution into existing processes for the manufacturers of solid-state batteries.

## Conclusion

The aim of for Daikin was to develop a modular catholyte with a fluoropolymer base, which is adaptable to various solid-state battery strategies and can be tailored according to the needs of the materials developed by all other partners inside the consortium. The NEOFLON family of polymers provides a unique platform fulfilling core necessities for a successful development implementation of the base material in all other parts of the project. Through extensive physicochemical and electrochemical characterization and benchmarking, we were able to determine one polymer as an ideal candidate for the catholyte matrix material. In short, it offers three key advantages that provide a lot of opportunities inside the ASTRABAT consortium.

Adaptability: The fluoropolymer is compatible with a variety of materials, including all lithium salts, plasticizers inorganic filler materials, active materials, and anolyte polymers which are developed by the consortium partners.

**Scalability:** The fluoropolymer provides a versatile platform, which is compatible with lab-scale and industry-scale processes due to its easy processability and favourable mechanical, thermal, and chemical stability. It is equally compatible with conventional LIB production processes and alternative routes which will be explored in the framework of ASTRABAT (e. g. 3D-printing).

**Customisability:** Due to the modular development approach for the catholyte polymer matrix, future development issues can be addressed by modifications in the formulation and by smart choices of additives, enabling further optimization of the SPE in the coming parts of the project and tailored to the role of the polymeric material inside the cell. In the case of unexpected performance shortcomings at different stages of the project, the base material can be adapted without changing processes and/or significant delays in the work.





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