

# Grafted Polyrotaxane-based Solid Polymer Electrolytes for All-Solid-State Lithium Metal Batteries with Enhanced Properties

Laura Imholt<sup>a</sup>, Gunther Brunklaus<sup>a,b</sup>, Isidora Cekic-Laskovic<sup>a,b</sup>, Martin Winter<sup>a,b</sup>

<sup>a</sup> Helmholtz-Institute Münster, IEK-12, Forschungszentrum Jülich GmbH,  
Corrensstraße 46, 48149 Münster, Germany

<sup>b</sup> MEET Battery Research Center / Institute of Physical Chemistry, University of  
Münster, Corrensstraße 46, 48149 Münster, Germany

E-mail: l.imholt@fz-juelich.de

Solid polymer electrolytes (SPEs) and their potential use in high specific energy solid state batteries remain in the focus of exhaustive scientific research. Solvent-free polymer electrolytes exhibit advantages in terms of mechanical stability, operational safety and simplicity of cell design.[1] However, application of polymer electrolytes to all-solid-state lithium ion batteries (ASS-LIBs) and all-solid-state lithium ion batteries (ASS-LMBs), requires improvements in respect to lithium ion conductivity, especially at low temperature.

In this work, a new generation of Li<sup>+</sup>-conducting SPEs obtained from supramolecular self-assembly of polyethylene oxide (PEO), cyclodextrin (CD) and lithium salt has been designed and investigated for application in lithium metal batteries (LMBs) and lithium ion batteries (LIBs). The considered electrolyte comprises nano-channels for fast lithium-ion transport.[2] By means of synergistic and complementary experimental and simulation results, we could show that methylation of CD shifts the steady-state between lithium ions inside and outside the channels, thereby significantly improving the ionic conductivity. Therefore, within this work, we replaced the methyl groups by short flexible polymer chains, based on polycarbonates. Simultaneous ring-opening polymerization of  $\epsilon$ -caprolactone from both ends of the polymer backbone and functionalization of CD rings were applied to yield grafted polyrotaxanes with enhanced mechanical and electrochemical properties. This modification resulted in an increase in ionic conductivity by three orders of magnitude (up to 1 mS/cm at 60 °C) and the designed complexes yielded electrolytes suitable for galvanostatic cycling in LMBs with fast charging and discharging rates for more than 200 charge/discharge cycles.

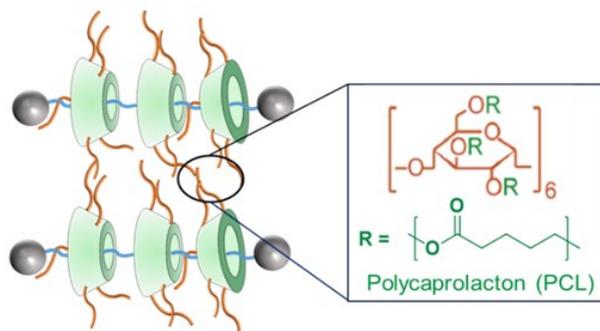


Figure 1. Structure of the investigated grafted polyrotaxane with CD (green) as host molecule, PEO (blue) as guest molecule and polycaprolactone-based polymer side chains (orange).

## References:

- [1] L. Long, S. Wang, M. Xiao, Y. Meng, J. Mater. Chem. A 4 (2016) 26 10038-10069.  
[2] L.Y. Yang, D.X. Wei, M. Xu, Y.F. Yao, Q. Chen, Angew. Chem. Int. Ed. 53 (2014) 14 3631-3635.