

# PVdF/PVdF-HFP Based Gels and Polymer Blends for Li-ion Battery Electrolyte Applications

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Lithium ion batteries are important as power sources for many portable electrical devices (e.g. laptops and mobile phones) and the use of Li-ion batteries is predicted to grow substantially in the near future, whilst branching out into new application areas. Despite this rising demand, we presently lack of a safe, easy to process and thus low-cost, ideally mechanically flexible batteries. A key challenge is to resolve how to decouple the ionic transport from the segmental relaxation and thus to achieve good transport properties combined with mechanical rigidity. A decoupling between ionic transport and segmental polymer dynamics can be achieved by addition of solvent either as a plasticizer or to form a polymer gel [1].

Another route is the use of polymer blends [2] where an ion-conducting oligomer or polymer is combined with a polymer that contributes mechanical rigidity. For both polymer gel and polymer blend based electrolytes, efficient processing and manufacturing routes can be developed. However, a good understanding of the ion transport mechanisms, as well as the polymer structure, dynamics and rheology is needed to optimise these materials.

We will here present results on polymer gel and polymer blend electrolytes based on either polyvinylidene difluoride (PVDF) or polyvinylidene difluoride-hexafluoropropylene (PVDF-HFP). We have performed systematic studies of the ion transport, structure, dynamics, and rheology over a wide range of material compositions for blends based on PVdF-HFP ( $M_w=400\text{kg/mol}$ ) and di/poly(propylene glycol) (DiPG/PPG) of varying molecular weights (134~2000g/mol) based on two different salts, LiTFSI and LiBF<sub>4</sub>. We have also characterised the effects of addition of a plastic crystalline material as a plasticizer. On the other hand, we have performed a systematic study of PVDF-based gels based on the organic solvent propylene carbonate (PC) and the salt LiBF<sub>4</sub>, with the aim to determine the gel-formation mechanism and the associated structure-formation, rheology and ion transport over a wide range of polymer and salt compositions, to facilitate the design of efficient processing routes for PVDF-based polymer gel electrolytes.

We present results from a wide range of experimental techniques including broadband dielectric spectroscopy (BDS), calorimetry, dynamic mechanical thermal analysis (DMTA), shear rheology, (cryo-) scanning electron microscopy (SEM/Cryo-SEM), small angle neutron scattering (SANS), small/wide angle X-ray scattering (SAXS/WAX), nuclear magnetic resonance (NMR), and dynamic/static light scattering (DLS/SLS).

## References:

- [1] A. M. Stephan, Eur. Polym. J, 42 (2006) 31.
- [2] D. Cangialosi et al., Macromolecules, 41 (2008) 1565.