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Ion-transport in battery relevant polymers - challenges in analysis

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Li metal-based batteries are foreseen as future energy storage devices. However, a major hindrance to their industrial application is the uneven Li deposition on the anode that results in dendritic growth and poor columbic efficiency (CE).

Our previous work had revealed that batteries with a mechanically rigid M1I0 polymer coating on anode show a comparatively lower CE than those with a a viscolelastic M1I3 polymer coating [1]. Moreover, while anodes with M1I0 polymer coatings still showed Li dendrite growth, those with M1I3 polymer coating had smooth and homogeneous Li deposition.

In this work, we try to further study these polymers to understand the interplay between polymer dynamics and CE in batteries and the influence of Li salt. The polymer network in these polymers consists of a PFPE backbone interconnected by urethane units containing H-bonds, with H atoms involved in N-H bond being responsible for polymer rigidity.

When Li salt is added, Li cations bind to the oxygen atoms in the urethane units. In fact, FTIR revealed a lengthening of the C=O bond on addition of Li salt, which was attributed to weakening of H-bond, as Li cation formed Lewis acid complexation with the basic carbonyl oxygen site. This interaction is known also to increase barrier heights to rotation.

At the highest energy resolution QENS revealed localized dynamics in all polymers. Amongst other fast motions, these could be a due to constrained motions of the hydrogen-bonded units. While the extracted correlation times for M1I0, M1I3 and Li_M1I0 are similar, the correlation time for Li_M1I3 was twice as long. A similar trend is observed at the intermediate resolution.

Interestingly, at the lower energy resolution which probes slightly faster dynamics, the pristine polymers still show localized motions, whereas polymers with Li salt show diffusive motions. At these temporal scales, H-bond breaking is possible which may result in decoupling of Li-ions, facilitating thereby cation migration, also across the hydrogen-bond connected chains (intrachain migration).

The QENS signal contains a significant coherent contribution which limits the accessible Q-range for incoherent dynamics. At Q values below 1 Å-1, we account for the coherent contribution with a linear background. However, at higher Q values, we require characterizing the ratio of incoherent to coherent via polarisation methods.

[1] Z. Huang, S. Choudhury, N. Paul, et. al. Adv. Energy Mater. 12 (2022) 2103187

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