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Decoupling of Ionic Transport from Segmental Relaxation in Polymer Electrolytes YANGYANG WANG, Oak Ridge National Lab, ALEXANDER AGAPOV, University of Akron, FEI FAN, University of Tennessee-Knoville, KUNLUN HONG, XIANG YU, Oak Ridge National Lab, JIMMY MAYS, University of Tennessee-Knoville, ALEXEI SOKOLOV, Oak Ridge National Lab — Polymer electrolytes provide elegant solutions to many difficulties in battery technology. However, their relatively low ionic conductivity has become the bottleneck for developing batteries with higher power density, shorter charging time, and better operations at low temperatures. In this work, we present detailed studies of the relationship between ionic conductivity and segmental relaxation in a set of specially-designed polymer electrolytes with systematic variation in chain rigidity. Our analysis shows that the ionic conductivity indeed can be decoupled from segmental dynamics in rigid polymers and the strength of the decoupling correlates with the fragility, but not with the glass transition temperature. These results call for a revision of the current picture of ionic transport in polymer electrolytes. We relate the observed decoupling phenomenon to frustration in packing of rigid polymers, which also affects their fragility. The principles demonstrated in this study may provide an alternative approach to design of highly conductive materials: by incorporating relatively rigid chain structures, it is possible to develop a new class of solid polymer electrolytes with strongly decoupled ionic conductivity.

Yangyang Wang
Oak Ridge National Lab

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