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Negative Transference Numbers in Polymer Electrolytes
DANIELLE PESKO, KSENIA TIMACHOVA, NITASH BALSARA, University of California, Berkeley — Energy density and safety of conventional lithium-ion batteries is limited by the use of flammable organic liquids as a solvent for lithium salts. Polymer electrolytes have the potential to address both limitations. The poor performance of batteries with polymer electrolytes is generally attributed to low ionic conductivity. The purpose of our work is to show that another transport property, the cation transference number, t+, of polymer electrolytes is fundamentally different from that of conventional electrolytes. Our experimental approach, based on concentrated solution theory, indicates that t+ of mixtures of poly(ethylene oxide) and LiTFSI salt are negative over most of the accessible concentration window. In contrast, approaches based on dilute solution theory suggest that t+ in the same system is positive. In addition to presenting a new approach for determining t+, we also present data obtained from the steady-state current method, pulsed-field-gradient NMR, and the current-interrupt method. Discrepancies between different approaches are resolved. Our work implies that in the absence of concentration gradients, the net fluxes of both cations and anions are directed toward the positive electrode. Conventional liquid electrolytes do not suffer from this constraint.

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