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### **Simultaneously Ion- and Electron-Conducting Block Copolymer Binders for Battery Electrodes<sup>1</sup>**

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Lithium-ion batteries provide a portable, on-demand source of electrical energy and are comprised of multiple components for storing and releasing ions, transporting charges, and maintaining mechanical integrity. Polymeric binders, although representing only a fraction of the battery, are an important component for maintaining adhesion between different parts. Polymers that are simultaneously ion- and electron-conducting and redox-active are potentially ideal materials for use in electrodes, and here we show that such polymers can improve both mechanical and electrochemical properties of electrodes. First, flexible, carbon-free hybrid battery cathodes are prepared using poly(3-hexylthiophene)-*block*-poly(ethyleneoxide) (P3HT-*b*-PEO) as a binder. Only 5\_wt % polymer was required to triple the flexibility of V<sub>2</sub>O<sub>5</sub>, and electrodes comprised of 10\_wt % polymer had unusually high toughness (293\_kJ/m<sup>3</sup>) and specific energy (530\_Wh/kg), both higher than reduced graphene oxide paper electrodes. Next, we present work on self-doped conjugated polymeric binders, which provide stable conductivities and are fully water-processable. These materials are incorporated into V<sub>2</sub>O<sub>5</sub> cathodes and suppress the crystallization of V<sub>2</sub>O<sub>5</sub>, even at thermal annealing temperatures above 400 C, maintaining the more favorable aerogel structure. Finally, we discuss the design and development of conjugated and redox-active polymers in Silicon anodes. These results highlight the importance of tradeoffs between mechanical and electrochemical performance in the design of conjugated polymeric binders.

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