Tailored Organic Electrode Material Compatible with Sulfide Electrolyte for Stable All-Solid-State Sodium Batteries

Xiaowei Chi\textsuperscript{a}, Yanliang Liang\textsuperscript{a}, Fang Hao\textsuperscript{a}, Ye Zhang\textsuperscript{a}, Justin Whiteley\textsuperscript{b}, Hui Dong\textsuperscript{a}, Pu Hu\textsuperscript{a}, Sehee Lee\textsuperscript{b}, Yan Yao\textsuperscript{a}

\textsuperscript{a}Department of Electrical and Computer Engineering, University of Houston, Houston, Texas 77204, USA

\textsuperscript{b}Department of Mechanical Engineering, University of Colorado at Boulder, Boulder, Colorado, 80309, USA

E-mail: yyao4@uh.edu

All-solid-state sodium batteries (ASSSBs) have been recognized as a promising battery technology to address the safety and cost concerns of lithium-ion batteries with nonflammable solid-state electrolyte and ubiquitous sodium resources. However, the intrinsic mismatch between low anodic decomposition potential of superionic sulfide electrolytes and high operating potentials of conventional sodium-ion cathode materials leads to chemical reactions at the cathode-electrolyte interface and thus unstable cycling performance. Here we report for the first time an organic carbonyl compound, Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6}, as a high-capacity cathode material in all-solid-state batteries that is chemically and electrochemically compatible with sulfide electrolyte. A bulk-type ASSSB based on Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6} cathode, Na\textsubscript{3}PS\textsubscript{4} electrolyte, and Na\textsubscript{12}Sn\textsubscript{4} anode shows high specific capacity (184 mAh g\textsuperscript{-1}) and the highest specific energy (395 Wh kg\textsuperscript{-1}) among sodium intercalation compound-based ASSSBs. The cell shows capacity retention of 76\% after 100 cycles at 0.1C and 70\% after 400 cycles at 0.2C, representing the record cycling stability in ASSSBs reported to date. Electrochemical analyses confirm that the moderate redox potential of Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6} is crucial for the stability of cathode-electrolyte interface thus long cycle life. Additionally, Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6} could also function as an anode material with specific capacity of 187 mAh g\textsuperscript{-1}, thereby enabling a symmetric all-organic ASSSB with Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6} as both cathode and anode materials.

Figure 1. (Left) Potential–capacity plot for previously reported intercalation cathode materials and Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6} for ASSSBs. Shadow and blank bars represent the observed and theoretical specific capacities, respectively. Specific energy is calculated considering a sodium anode. (Right) Capacity and coulombic efficiency vs. cycle number at 0.1C for an ASSSB made of Na\textsubscript{4}C\textsubscript{6}O\textsubscript{6}[Na\textsubscript{3}PS\textsubscript{4}]Na\textsubscript{12}Sn\textsubscript{4}.

References: