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

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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, $Na_4C_6O_6$, 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 $Na_4C_6O_6$ cathode, Na_3PS_4 electrolyte, and $Na_{15}Sn_4$ anode shows high specific capacity (184 mAh g^{-1}) and the highest specific energy (395 Wh kg^{-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 $Na_4C_6O_6$ is crucial for the stability of cathode-electrolyte interface thus long cycle life. Additionally, $Na_4C_6O_6$ could also function as an anode material with specific capacity of 187 mAh g^{-1}, thereby enabling a symmetric all-organic ASSSB with $Na_4C_6O_6$ as both cathode and anode materials.

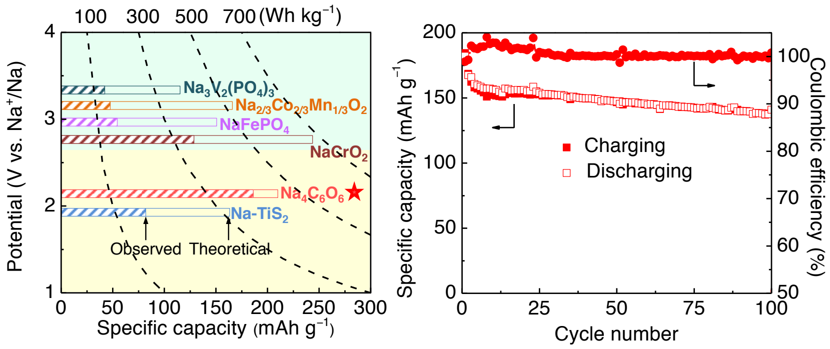

Figure 1. (Left) Potential–capacity plot for previously reported intercalation cathode materials and $Na_4C_6O_6$ for ASSSBs. Shadow and blank bars represent the observed and theoretical specific capacities, respectively. Specific energy is calculated considering a sodium anode. (Right) Capacity and coulombic efficiency vs. cycle number at 0.1C for an ASSSB made of $Na_4C_6O_6|Na_3PS_4|Na_{15}Sn_4$.

References:
[1] X. Chi, Y. Liang, F. Hao, Y. Zhang, J. Whiteley, H. Dong, P. Hu, S. Lee, Y. Yao, *Angewandte Chemie*, (2018) in press.