LITHIUM-ION BATTERIES

Entropy stabilization

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For Li-ion batteries (LIBs), due to the positive electrodes generally having smaller capacities than the negative electrodes, the development of high-performance positive electrode materials is the bottleneck for further improvement of the amount of energy that can be stored. Positive electrodes in LIBs are typically made of transition metal oxides. They have long been thought to function solely based on cationic redox, and it was only recently revealed that the redox of O^{2-} anions also participates in battery electrochemistry. As both anionic and cationic redox contribute to the overall capacity, exploring materials in which both can occur is a promising strategy for designing high-energy electrodes. Now, Qingsong Wang, Horst Hahn, Ben Breitung and colleagues in Germany and India develop an oxyfluoride compound, $Li_x(Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})OF_x$, as a potential positive electrode material featuring multi-anionic and multi-cationic redox behaviour.

The oxyfluoride is fabricated by ball-milling a mixture of $(Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})O$ and LiF , and possesses the same rock-salt crystalline structure as its oxide precursor. Through X-ray diffraction and nuclear magnetic resonance measurements, the researchers suggest that Li and F species are randomly distributed on their respective cation and anion lattice positions, thus giving rise to a number of possible microstates and, consequently, high configurational entropy. This stabilizes the material and is further suggested to play a key role in maintaining structure and capacity stability during cycling of the oxyfluoride electrode. Furthermore, the F species help to increase the electrode working potential to 3.4 V versus Li^+/Li , higher than that of most rock-salt oxyfluorides, as well as to suppress oxygen release, a phenomenon often seen during battery cycling. The theoretical specific capacity of the oxyfluoride is reported to be 222 mAh g^{-1} , somewhat lower than many oxide-based positive electrode materials.

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