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| Abstract |  |

Aqueous rechargeable zinc-ion batteries (ARZIBs) have drawn enormous attention because of their low-cost and eco-friendly cell components. However, designing high-performance cathode materials towards practical application of ARZIBs remains a major challenge. Therefore, in this contribution, a comprehensive study on K+ intercalated V2O5 (KVO) nanorods with exposed facets as a high-performance cathode for ARZIBs is presented. The KVO cathode exhibits remarkable discharge capacities of 439 and 286 mAh g−1 at current densities of 50 and 3000 mA g−1, respectively. Furthermore, it recovers 96% of the capacity after 1500 cycles at 8000 mA g−1. Impressively, the Zn/KVO battery offers a specific energy of 121 W h kg−1 at high specific power of 6480 W kg−1. The storage mechanism of the KVO cathode in an ARZIB is systematically elucidated using in operando synchrotron X-ray diffraction, ex situ synchrotron X-ray absorption spectroscopy, ex situ TEM analyses and first-principles calculations. The superior performance of the cathode is attributed to its unique exposed layer structure with high surface energy, high conductivity and low migration barrier for Zn2+ migration. This study provides insight into designing high-performance cathode materials for ARZIBs and other electrochemical systems.