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Boosting lithium-selenium batteries

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Lithium-selenium (Li-Se) batteries have been recognized as one of the most promising candidates for nextgeneration energy storage devices due to their high energy density and high conductivity of Se^[1,2]. However, the shuttle effect has become a major shackle restricting their commercial development^[3,4]. Confining active material selenium inside the cathode is an effective method to suppress this phenomenon^[5]. Moreover, rapid transportation of electrons and ions is indispensable for achieving high electrochemical performance Li-Se batteries^[6].

Combining the microporous carbon particles that provide Se loading space and adsorption to polyselenides with a highly conductive meso/macropores network is a design direction to suppress the shuttle effect and achieve high reaction kinetics^[7]. Microporous-rich metal-organic framework (MOF) and high conductivity CNTs become the ideal compositions. The current work presents tunable size zeolitic imidazolate framework-8 (ZIF-8) derived microporous carbon particles strung by MWCNTs (ZIF-8-C@MWCNTs-X, X = 1-5) with a simple *in-situ* solvothermal reaction [Figure 1]^[8]. The active Se is dispersed in the designed ZIF-8-C@MWCNTs host materials by a simple melt-diffusion method.

Compared with Se@ZIF-8-C cathode battery, the electrochemical performance of Se@ZIF-8-C@MWCNTs achieves high improvement due to the accelerated electron/ions transfer brought by the interconnected MWCNTs. Moreover, the size of ZIF-8-C particles is a critical factor for electrochemical kinetics in the charge/discharge process. For too small particles of ZIF-8-C, the problem of shuttle effect is serious due to



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Figure 1. Schematic mechanism of the (A) Se@ZIF-8-C, (B) ZIF-8-C@MWCNTs-1 and (C) Se@ZIF-8-C@MWCNTs-5 electrodes for lithium ion and electron movement. (D) Cycling performance of Se@ZIF-8-C and Se@ZIF-8-C@MWCNTs-3 at high current density of 1 C, (E) summary of the specific capacity and impedance of the Se@ZIF-8-C@MWCNTs-X (X = 1-5) positive electrodes according to the particle size of the internal ZIF-8 derived porous carbon.

the easily diffused out polyselenide. Otherwise, too large particle size leaves the ions transfer difficult. At an optimized particle size of 300-500 nm, both the suppressed shuttle effect and the rapid ions transfer are achieved. Se@ZIF-8-C@MWCNTs-3 with the optimized ZIF-8-C size exhibited the best cycle stability with a high capacity of 468 mAh g⁻¹ at 0.2 C after 200 cycles. Even at the high rate of 1 C, the capacity of Se@ZIF-8-C@MWCNT-3 can remain at 206 mAh g⁻¹ after 500 cycles.

In the designed Se@ZIF-8-C@MWCNTs system, ZIF-8-C particle functions as a reaction block, where the Se confined inside of the micropores undergoes a rapid electrochemical reaction. While the interconnected MWCNTs works as transfer block to ensure the supply of required electrons and ions. This kind of block mode design realizes both the confinement of the reaction and the rapid mass transportation. Compared with the conflated design system, the modular system enables the electrochemical reaction inside the battery to proceed in an orderly manner as living organisms. The efficient synergy of different modules is the premise for the system to maximize its value, which is reflected in the particle size of ZIF-8-C nanocarbons

in the Se@ZIF-8-C@MWCNTs system. The Se@ZIF-8-MWCNTs system obtains the best match at the size of 300-500 nm.

The design concept of coordinated development of work blocks has a significant effect on the working mechanism of the system. It could give a guiding for other reaction environments. The strategy in this work realized the *in-situ* interconnection of the ZIF-8 derived reactor and MWCNTs highways. The effect of particle size on the reaction kinetics provides suggestions for further structural design and high performance Li-Se batteries.

DECLARATIONS

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