Synthesis of Porous Si/Carbon Composites for High-Energy Lithium-ion Battery

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In order to improve the energy and power densities of lithium-ion batteries (LIBs), the low-capacity and low-rate graphitic carbon anodes have to be replaced with advanced anode materials. Silicon is one of the most attractive anode materials due to its high theoretical capacity (4200 mAh/g) and low working voltage (~0.4 V) [1]. However, Si anode undergoes large volume expansion during Li alloying/de-alloying processes causing electrode pulverization and thereby short cycle life. To solve these problems, we prepared mesoporous silicon (mpSi) by magnesiothermic reduction (MR) of commercial zeolites. MR is an effective process to prevent severe sintering of silicon due to its lower reduction temperature (~650℃) than that of conventional carbothermic reduction (>2000℃) [2]. However, it also generates huge exothermic heat during reaction causing the sintering of pre-reduced silicon particles. In order to obtain the mpSi in high yield, high purity and with small primary Si particles, various reduction conditions including reduction temperature, reactants ratio, reactor system (closed or open) and with or without heat scavenger of NaCl etc., were tested. The prepared mpSi was coated with carbon to form mpSi/C composites for use as anodes for lithium-ion batteries.

The SEM images of mpSi in Figure 1b reveal that each particle had similar size dimensions to the parent zeolite particles in Figure 1a yet were highly porous with a considerable roughened surface. The TEM images of the single mpSi particle in Figure 1c clearly indicate its porous structure with pore sizes in the range of 20–40 nm. The primary silicon particles are 20–30 nm in diameter and are highly interconnected. The selected area electron diffraction (SAED) pattern in the inset of Figure 1c further verifies that mpSi is comprised of crystalline silicon. As seen in Figures 1e-1g, the mpSi/C composite exhibited a high reversible capacity of over 1100mAh g^{-1} at a current density 100 mA g^{-1}, excellent rate capability and cycle stability with high coulombic efficiencies up to 200 cycles. The mpSi/C composites were also applied to fabricate a full LIB cell and details on its electrochemical properties are to be presented.

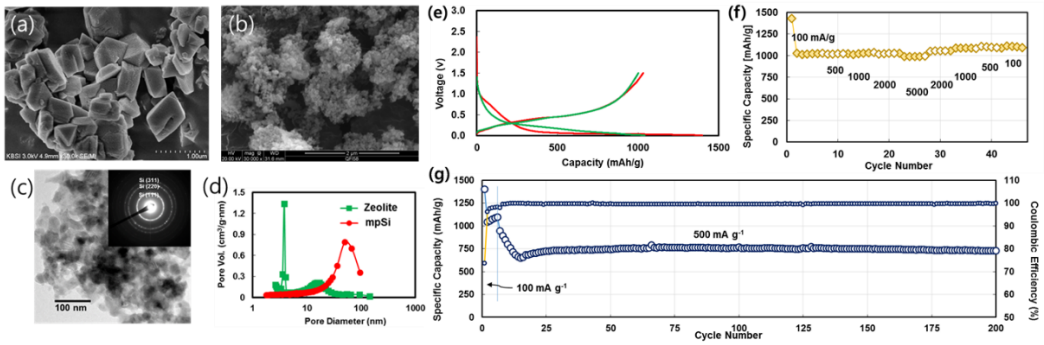

Figure 1. SEM images of (a) zeolite and (b) mpSi, (c) TEM image of mpSi with SAED pattern in the inset, (d) pore size distribution of zeolite and mpSi, and (e) charge/discharge voltage profiles, (f) rate responses and (g) cycling stability of mpSi/C.

References:

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[2] N. A. Liu, K. F. Huo, M. T. McDowell, J. Zhao, Y. Cui, Sci. Rep. 2013, 3, 10.1038/srep01919