Nanomaterials Design for Lithium-Sulfur Batteries

A thesis presented for the award of the degree of

Doctor of Philosophy

From

University of Technology Sydney

By

Yi Chen, B.Eng., M.Eng.

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CERTIFICATE OF ORIGINAL AUTHORSHIP

I, Yi Chen declare that this thesis, is submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the School of Mathematical and Physical Sciences, Faculty of Science at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise reference or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

This document has not been submitted for qualifications at any other academic institution.

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DEDICATION

This thesis is dedicated to my family. Appreciate their love and support.

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ABSTRACT

Lithium-sulfur (Li-S) batteries, which rely on the redox reactions, show great promise for next-generation energy storage owing to their high theoretical energy density, environmental benignity and low cost of sulfur. However, the practical application of Li-S batteries has been largely impeded by the low conductivity of sulfur and the shuttle effect of polysulfides. One of the most effective strategies to overcome these problems is to disperse insulating sulfur active material within other conductive matrixes that are capable of physically adsorbing and/or chemically binding sulfur and its intermediate polysulfides. In this thesis, we designed two types of host materials that can be used to improve the electrochemical performance of Li-S batteries.

A new self-standing host enabled by a 3D hierarchically-porous titanium monoxide-graphene composite film was designed to overcome the main challenges of Li-S batteries. The hierarchically porous graphene scaffold can not only facilitate rapid lithium ion and electron transport, but also provide sufficient spaces to accommodate sulfur species. In addition, the ultrafine and polar titanium monoxide nanoparticles embedded in the three-dimensional graphene networks show strong chemical anchoring for polysulfides, and their inherent metallic conductivity accelerates the redox reaction kinetics. Benefiting from this attractive architecture, the freestanding titanium monoxide-graphene/sulfur cathode demonstrated superior electrochemical performance for Li-S batteries.

Uniform Co-Fe mixed metal phosphide (Co-Fe-P) nanocubes with highly interconnected-pore architecture were synthesized as sulfur host for Li-S batteries. With the

highly interconnected-pore architecture, inherently metallic conductivity and polar characteristic, the Co-Fe-P nanocubes not only offer sufficient electrical contact to the insulating sulfur for high sulfur utilization and fast redox reaction kinetics, but also provide abundant adsorption sites for trapping and catalyzing the conversion of lithium polysulfides to suppress the shuttle effect. As a result, the sulfur-loaded Co-Fe-P (S@Co-Fe-P) nanocubes exhibited superior electrochemical performances both in coin cells and pouch cells.