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Article / Book Information

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著者(和文)	BELGIBAYEVA A
Author(English)	Ayaulym Belgibayeva
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（博士課程）
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論 文 要 旨

THESIS SUMMARY

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学生氏名： Student's Name	Belgibayeva Ayaulym	指導教員（主）： Academic Supervisor(main)	Prof. Taniguchi Izumi
		指導教員（副）： Academic Supervisor(sub)	

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Energy storage devices, particularly lithium batteries, play an essential role in developing renewable energy sources and electric vehicles as solutions for fossil fuel combustion-caused environmental issues. On the other hand, the current capacities of commercially available lithium batteries are limited by their electrode materials. Therefore, the development of new generation electrode materials with high power and safety is urgently needed. There are two ways to achieve high power: by increasing voltage and by increasing capacity. Among different candidate materials, $LiCoPO_4$ is attractive as high-voltage cathode material, which operates within the electrolyte stability window at 4.8 V (vs. 3.7 V for commercial $LiCoO_2$) and possesses higher thermal stability. Another attractive cathode material is sulfur, owing to its high theoretical capacity of 1675 mAh g^{-1} (vs. 148 mAh g^{-1} for commercial $LiCoO_2$).

Along with the improvement of cathode materials, consecutive development of anode materials is needed. SiO_2 has gained particular attention among different anode materials due to its high theoretical capacity (1965 mAh g^{-1}), natural abundance, low cost, and environmental friendliness. Common issues for $LiCoPO_4$, sulfur, and SiO_2 are low electronic and ionic conductivities. Furthermore, dissolution and shuttle of intermediate products of electrochemical reactions, polysulfides, remain another problem for lithium-sulfur batteries. The development of free-standing functional carbon composite nanofiber mats could solve all of the limitations mentioned above. Electrospinning is a straightforward and cost-effective method to synthesize such nanofibers. Since commonly used electrospinning template and carbon source PAN have restrictions on compositions (due to the low solubility of inorganic components in DMF solvent), more soluble PVP should be extensively studied as an alternative carbon source.

The present study systematically investigated the synthesis of different free-standing functional carbon composite nanofiber mats for different applications using PVP as a carbon source. The main conclusions are summarized as follows:

Chapter 2 reported the synthesis of free-standing SiO_2/C composite nanofiber mats (FS-SiO_2/C-CNFM) as anode materials for lithium-ion batteries. SiO_2/C composite nanofibers were successfully synthesized by electrospinning with two-step heat treatment, comprising the pre-oxidation at 280° C in the air followed by annealing at 700° C for 1 h in a reduced atmosphere, using PVP as a carbon source. The effect of pre-oxidation on the structure and electrochemical performance of SiO_2/C composites was studied. Mechanism of the PVP decomposition and carbonization under different heat treatment conditions was proposed. Furthermore, free-standing SiO_2/C composite nanofibers with different physical properties were synthesized. The effect of the fiber diameter and pore size on the electrochemical performance were evaluated.

Chapter 3 reported the synthesis of the free-standing $LiCoPO_4$/C composite nanofiber mats (FS-LCP/C) as high-performance cathode materials for lithium-ion batteries. Novel free-standing $LiCoPO_4$/C composite nanofiber mats were successfully synthesized by carbon coating $LiCoPO_4$ nanoparticles by electrospinning, followed by heat treatments using PVP as a carbon source. Synthesis conditions were optimized to get pure-phase fibrous, electrochemically active $LiCoPO_4$. The effect of pre-oxidation temperature on carbon content and physical properties of $LiCoPO_4$/C composites and their correlation to electrochemical properties were studied. Furthermore, the effect of SiO2 coating on the performance of $LiCoPO_4$/C composite nanofiber mats was evaluated.

Chapter 4 reported the effect of the FS-SiO_2/C-CNFM on the electrochemical performance of sulfur-based lithium batteries. To overcome the polysulfide dissolution and shuttle effect problems on lithium batteries with sulfur-based cathodes, applying the FS-SiO_2/C-CNFM interlayer, prepared by electrospinning with heat treatments in Chapter 2, was proposed. The application of the free-standing SiO_2/C composite interlayer, employed to lithium-sulfur battery without any additional coating procedure, was studied for the first time. The polysulfides adsorption and catalytic effect on polysulfides conversion for SiO_2/C composite

were studied compared to commercial carbon fiber paper. Polysulfide adsorption/conversion mechanisms were proposed based on the ex-situ physical characterizations.
Chapter 5 reported the synthesis of the free-standing $Co_xP/Co_3(PO_4)_2/C$ composite nanofiber mat as a multi-functional interlayer for lithium-sulfur battery. Novel free-standing $Co_xP/Co_3(PO_4)_2/C$ composite nanofiber mat, that was not reported so far, was successfully synthesized by electrospinning with heat treatments. Effect of combination of different cobalt phosphides and cobalt phosphate on electrochemical properties of lithium-sulfur batteries, when employed as free-standing interlayer, was studied for the first time. Effect of process parameters on the composition and physical properties of the composites was studied. Furthermore, effect of the interlayer composition on polysulfide chemisorption, conversion, and electrochemical properties of lithium-sulfur batteries were evaluated. The polysulfide trapping and catalytic conversion mechanisms were studied for the first time by ex-situ characterizations. According to the obtained results, each interlayer component played a vital role in trapping and converting polysulfides and comprehensively improved the electrochemical performance of lithium-sulfur batteries.
The obtained results confirmed that functional free-standing composite nanofiber mats could be successfully synthesized and applied to different energy storage devices, including lithium-ion batteries and lithium-sulfur batteries. The design of free-standing electrodes allowed increasing the energy density by omitting the coating procedure to electrochemically inactive heavy metal current collectors using inactive binders.