SUMMARY OF Ph.D. DISSERTATION

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Title

Preparation of functional metal oxides by an aqueous solution process and their application to electronic devices

Abstract

Nanoscale metal oxides are interesting materials due to their unique properties and great potential for application to electronic and optical devices. A smart technique for the fabrication of nanoscale metal oxides is required for a further development of materials chemistry. Aqueous solution processes using crystal growth have attracted attention as a novel strategy for nanoscale processing. In an aqueous solution, metal oxide crystals are spontaneously constructed. The nanostructural control of metal oxides is achieved through self-organized crystal growth by adjusting the degree of supersaturation and addition of organic molecules in aqueous solutions. However, it is generally difficult to prepare multicomponent oxides and to precisely control nanostructures of products by aqueous solution processes. The strict control of crystal growth in aqueous solutions is desired for the industrial application. This study addresses the control of the components and oxidation state of metal oxides prepared in aqueous solutions, and the preparation of nanoarchitectures by a multistep process through an intermediate phase. The reaction mechanisms and device performances of the products were discussed toward the practical use of an aqueous solution process.

Chapter 1 summarizes the backgrounds and previous works of aqueous solution processes, and describes the objective of this study.

Chapter 2 describes the compositional control of rutile-type $Ti_xSn_{1-x}O_2$ solid solutions in aqueous solutions of Ti^{4+} and Sn^{4+}. We successfully prepared rutile-type $Ti_xSn_{1-x}O_2$ solid solutions, and achieved the control of the composition of solid solutions in the whole range (x = 0.0–1.0). The solid solutions had a higher photocatalytic activity than that of the commercial anatase-type TiO_2.

Chapter 3 mentions the control of the oxidation state of tin oxides crystals in solutions of Sn^{2+}. The selective preparation of SnO_2 and SnO crystals was achieved by adjusting the nucleation rate in response to the oxidation of Sn^{2+} by dissolving oxygen molecules. The morphologies of the products was controlled by varying the pH and [SnF_2] in the precursor solutions.

Chapter 4 shows the preparation of nanostructured SnO crystals by a multistep aqueous solution process through $Sn_6O_4(OH)_4$. The hierarchical architectures of SnO consisting of oriented units were obtained by adjusting the dissolution rate of $Sn_6O_4(OH)_4$. The novel aqueous solution process using the intermediate phase has a great advantage as a technique for the fabrication of nanoscale functional materials.

Chapter 5 describes the preparation of a nanoscale meshed electrode consisting of single-crystalline SnO and its application to an anode for lithium-ion batteries. The nanoscale meshes prepared by the multistep aqueous solution process improved the cycle performance of SnO electrodes.

Chapter 6 mentions the preparation of porous spinel-type $LiMn_2O_4$ using an aqueous solution process, and the cathode performance for lithium ion batteries. Porous $LiMn_2O_4$ was obtained from hierarchical architectures of $MnCO_3$ prepared in agar gel matrix containing Mn^{2+} ions. The porous framework of the products improved the electrochemical properties as a cathode.

Chapter 7 described the summary of this study and the future outlook of aqueous solution processes.