Studies on performance improvement routes for wide band gap oxide thermoelectric materials

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論 文 内 容 の 要 旨

Thesis Summary

The purpose of this doctoral thesis (Studies on performance improvement routes for wide band gap oxide thermoelectric materials) is to develop and examine new oxide thermoelectric materials. The development includes the utilization of unusual (supercritical drying of oxide gels) as well as new synthesis methods (microwave-assisted emulsion synthesis). Examination of the materials is done by direct measurement of synthesized samples properties via scanning electron microscopy, powder x-ray diffraction (XRD), four-point probe measurements of electrical properties and laser-flash measurements of thermal properties. The graduation thesis is composed of six chapters, each of them dealing with a different stage of the research and conclusions that can be drawn from the obtained results.

Chapter 1 serves as an introduction and describes the history and current state of the knowledge on thermoelectric materials. The chapter is subdivided into four parts. Part 1 describes the history and explains what profits can be gained with utility thermoelectric materials. Part 2 deals with the explanation of the basics of the thermoelectric effect. Part 3 explains the thermoelectric effect in detail, taking into account the effects that exist in real materials. Part 4 presents classes of known high-temperature thermoelectric materials with their limitations and advantages.

Chapter 2 presents the results of research into introducing nano-porosity into ZnO samples with a bottom-up approach. It was discovered that unlike silica oxide, zinc oxide does not form hollow spheres when microwave-assisted emulsion synthesis is utilized. However asymmetrical ZnO micro-crystals with unreported before morphology, were obtained and studied in detail. Samples with range of zinc ion concentrations from 0.1 M to 1.0 M have been prepared and contrasted with samples prepared without surfactant. Difference in morphology was illustrated by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) imaging. Crystal structure of obtained crystals have been studied using powder XRD. Crystals were cut with Focused Ion Beam (FIB) and crystal cell studied with scanning precession electron diffraction (SPED). Location of atoms in the crystals was determined using annular dark and bright field scanning transmission electron microscopy (STEM).

Chapter 3 explores the synthesis of conductive ZnO aerogel. The chapter presents a methodology that is successfully utilized for silica aerogel fabrication and applies it to zinc oxide. As a result, it is discovered that zinc oxide gel, heated above 270°C crystallizes and disassembles into powder. This chapter provides

evidence strongly suggesting that creation of zinc oxide thermoelectric aerogel is likely impossible, due to innate properties of the material.

Chapter 4 explores the possibility of utilization of indium oxide as a thermoelectric material. By optimizing the carrier concentration, $ZT \approx 0.75$ can be expected. The ZT values comparable to that of silicon-germanium alloys used in radioisotope thermoelectric generators can be expected for this material at the amorphous limit of the thermal conductivity, which has already been reported in the literature

Chapter 5 explores indium oxides with intrinsically low thermal conductivity. Using custom python program using Python Material Genomics (Pymatgen) library, and standard Ionic Radii method, candidate oxides and their dopants are selected and empirically tested. Screening of solubility of dopants was performed by combination of XRD pattern analysis and energy-dispersive x-ray spectroscopy (EDS) maps. As a result, stable candidate oxide (InNbO4) is found and soluble dopants are found.

Chapter 6 gathers the conclusions drawn from the experimental results and discussions.

The main aim of the graduation thesis has been reached. The author suggests that additional research into unconventional synthesis methods and new oxide materials holds the promise of useful and interesting discoveries.