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The Influence of Spark Plasma Sintering Temperature on the Microstructure and Thermoelectric Properties of Al,Ga Dual-Doped ZnO

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ZnO dual-doped with Al and Ga was prepared by spark plasma sintering using different sintering temperatures. The microstructural evolution and thermoelectric properties of the samples were investigated in detail. The samples obtained with sintering temperature above 1223 K had higher relative densities and higher electronic conductivity than the sample sintered at 1073 K. These results were supported by the solid-state reaction completion rate, which suggested that sintering temperature above 1223 K would be preferable for complete solid-state reaction of the samples. The sintering mechanism of ZnO particles and microstructure evolution at different sintering temperatures were investigated by simulation of the self-Joule-heating effect of the individual particles.

Key words: Thermoelectric oxide, spark plasma sintering, Al,Ga-codoped ZnO, sintering temperature

INTRODUCTION

Thermoelectric materials have received increasing attention for their wide applications such as in radioisotope thermoelectric generators used for space probes or systems for energy recovery from waste heat.[1] The conversion efficiency of thermoelectric modules depends on the operating temperature, with higher temperature yielding higher efficiency, as well as the material figure of merit (ZT). Thus, development of high-temperature thermoelectric materials may be of considerable value in practical applications. High-temperature-stable, nontoxic, low-cost zinc oxide (ZnO) is a promising candidate n-type thermoelectric material for such applications.[2–5] It has been reported by Ohtaki et al.[6] that dual doping of ZnO with Al and Ga resulted in a large enhancement of ZT to 0.47 at 1000 K and 0.65 at 1247 K for the composition $Zn_{0.96}Al_{0.02}Ga_{0.02}O$ made using conventional pressing techniques. However, conventional sintering processes such as hot pressing of doped ZnO require long processing time at high temperature, which limits grain size control. Spark plasma sintering (SPS) is an alternative, advanced sintering method. It simultaneously applies a high-intensity pulsed direct current and uniaxial pressure to the sample during the sintering process, which offers the possibility to densify the samples within a short time at a relatively lower temperature compared with the conventional sintering method.[7]

The present study aims to investigate the influence of the SPS sintering temperature on the thermoelectric properties and microstructure evolution of $Zn_{0.96}Al_{0.02}Ga_{0.02}O$, and to understand the sintering mechanism. This is done by relating the experimental observations to theoretical analysis of solid-state reaction kinetics and the temperature distribution in the particles.

EXPERIMENTAL PROCEDURES

The starting materials used in these investigations were ZnO (99.9%, 200 mesh powder; Alfa-Aesar), γ-Al_2O_3 (99.5%, 40 nm to 80 nm APS

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