Regular Paper

Effects of Two-step Spark Plasma Sintering on the Microstructures and Thermoelectric Properties of Pure/In-doped ZnO

Ahrong JEONG* and Byung-Koog JANG*

Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, 6-1 Kasuga-koen, Kasuga-shi, Fukuoka 816-8580, Japan

Received Oct. 11, 2022; accepted Nov. 9, 2022

Abstract

Pure and In-doped zinc oxide thermoelectric ceramics $[Zn_{(1-x)}In_x]O$ (x = 0, 0.005) were prepared by spark plasma sintering (SPS) with one-step/two-step (OS/TS) temperature profiles. The strategy of the TS temperature profile was sintering at a higher temperature (T_1) followed by prolonged sintering at a lower temperature (T_2) for densification while suppressing grain growth. In the first step, the temperature was 1150 °C for 15 min, and the second step was kept at 900, 950, 1000, 1050, and 1100 °C for 30 min. Through a combination of TS and SPS processes (TS-SPS), high relative densities of 94.6–98.3% were achieved without significant grain growth. Electron backscatter diffraction (EBSD) indicated noticeable grain growth suppression of 14.0 and 21.5% for the pure/In-doped TS-SPS samples as compared with OS-SPS samples, respectively. The highest power factor of 1.0 mW K^{-2} m^{-1} was achieved for the In-doped ZnO, attaining a dimensionless figure of merit (ZT) of 0.194 at 773 °C.

Keywords: Thermoelectric, ZnO, Doping, Two-step sintering, Spark plasma sintering

1. Introduction

With increasing global concern for energy and environmental problems, thermoelectric (TE) technology has attracted attention because it can convert waste heat into electrical power [1]. Intermetallic compounds such as Bi_2Te_3 and PbTe can give the best performance as TE materials [2–4]. However, their operating temperatures (27–227 °C and below 627 °C, respectively) are inherently limited by their melting points, which are 580 °C and 924 °C, respectively. Even below the melting points, these materials are not attractive because surface oxidation and vaporization can easily occur at such high temperatures. Furthermore, their constituting heavy metal elements are mostly toxic, scarce, and expensive. The efficiency [5] of a TE material is defined by a dimensionless figure of merit, $ZT = S^2\sigma T / \kappa$, where S, σ, T, and κ are the Seebeck coefficient, electrical conductivity, absolute temperature, and thermal conductivity, respectively. The ZT values can be enhanced by maximizing the power factor ($S^2\sigma$, PF) and reducing the thermal conductivity.

The zinc oxide (ZnO) ceramic [6–10], which is low-cost, abundant, and environmental friendly, has attracted considerable attention as a new TE material for high-temperature applications due to its excellent thermal and chemical stability in air. Nowadays, most researchers are focused on decreasing the thermal conductivity of ZnO-based TE materials [11–13]. Nevertheless, the enhancement of the PF is also important for optimizing the ZT values. The undoped ZnO ceramics exhibit a high S (~340 V K^{-1}); however, the σ (~10 S m^{-1}) is still too low to be used [14]. One obstacle to increasing σ is the limited solubility of donor dopants such as aluminum (Al), indium (In), and gallium (Ga), which is too low to achieve a sufficient carrier concentration for maximizing the PF of ZnO-based TE materials [14–17]. Therefore, increasing the solubility of donor dopants is needed to optimize the PF. However, the enhancement of the carrier concentration should be carefully designed because there is a trade-off between S and σ. A higher carrier concentration

*Corresponding author: jang.byungkoog@kyudai.jp