Improvement of Thermoelectric Properties of Transparent Conducting Oxide Ceramics by Nanostructuring.

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Highlights

- Nanoparticles have been synthesized by precipitation in aqueous medium.
- Dense ceramics with nano-sized grains have been sintered by SPS.
- Thermal conductivity is drastically reduced for ceramics with nanograins.

1. Introduction

The energy request is ever increasing due to technical evolution and increase of human population. Moreover, fossil fuel contributes to global warming. Many human activities are generating large amounts of waste heat, such as incinerators, power plants, steel mills, vehicles. Using this waste heat to produce electric energy would be an important contribution to sustainable development as it would allow reducing both greenhouse gas emissions and fossil fuel consumption. Thermoelectric materials are able to transform directly heat in electric energy or inversely electric energy in cold (Seebeck and Peltier effects) without any emissions (CO_2, other gases, radiations, …), vibrations or moving parts. The efficiency of a thermoelectric device η depends on the thermoelectric figure of merit $ZT = S^2\sigma T/\kappa$, where T is the absolute temperature, S is the Seebeck coefficient, σ the electrical conductivity and κ the thermal conductivity.

The ideal thermoelectric material should be stable in an oxidizing atmosphere in the targeted temperature range, easy to produce in large quantities, cheap and readily available, not toxic for environment and human health and of course presenting high efficiency. Nowadays, this material simply does not exist. Oxides, that meet these criteria, are considered as promising thermoelectric materials since Terasaki et al. have reported a large thermopower and power factor in the metallic oxide Na_xCoO_2 [1]. However, nowadays, the best oxide ceramic ZT value (0.5 at 1000 K [2]) at still remains too low for applications.

Hicks & Dresselhaus have proposed in 1993, theoretical pioneer works [3, 4] that showed that reducing the dimensionality from 3D bulk compounds to 2D or 1D nanostructures of Bi_2Te_3 should lead to improved ZT values (up to x 13). Since these pioneer works, many models proposed by various authors have predicted higher ZT values for nanostructured compounds [5, 6]. Clear experimental evidences of the ZT improvement due to nanostructuration of thermoelectric materials have been obtained by Poudel et al [7] who have reported a ZT improvement from 1 to 1.5 by using tellurides nanopowders.

Therefore, the aim of this work is to nanostructure oxide ceramics by SPS and study its influence on thermoelectric properties of transparent conducting oxides such as ZnO and SnO_2.

2. Methods

First, nanoparticles of undoped and Al-doped ZnO and undoped SnO_2 have been synthesized by aqueous precipitation [8, 9]. The influence of SPS parameters on the density, grain growth, transport properties and thermal conductivity has been studied.

3. Results and discussion

The Al-doped powders and the pure ZnO have been respectively sintered at 500°C and 600°C with a pressure of 100 MPa during 5 minutes. The pure ZnO ceramic exhibits a very low relative density of 65%, whereas the Al doped ceramics reached a relative density between 90-96%. Depending of the temperature, different grain sizes can be reached. Small grain size can drastically reduces thermal conductivity (figure. 1) and therefore increases ZT values. Similar results have been obtained for SnO_2 ceramics sintered at 950°C with a pressure of 100-150 MPa during 5-10 minutes. The median grain size is 60-70 nm and the thermal conductivity is divided by a factor of 5-7 (at high and low temperature, respectively) compared to micro-grained ceramics.

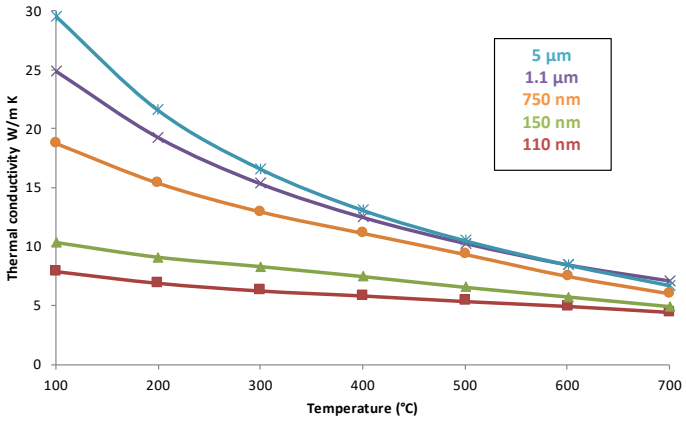

Figure 1. Thermal conductivity of Al-doped ZnO ceramics with different grain sizes.

4. Conclusions

Nanoparticles of ZnO, Al-doped ZnO and SnO_2 have been synthesized by precipitation at low temperature in aqueous medium. SPS has allowed sintering dense ceramics with nano-sized grains. This study clearly evidences the influence of nanostructure on the thermoelectric properties of transparent conducting oxide ceramics. Thermal conductivity is drastically reduced for ceramics with nanograins, leading to improved ZT values.

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Keywords
Thermoelectric ; Oxide ; Nanostructuring.