Impact of Cu doping and Mg-deficiency on Mg_2Sn thin films thermoelectric properties

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Thermoelectric materials have attracted global interest according to their ability to convert directly thermal to electrical energy, thus providing a clean and renewable supply of energy. The efficiency of thermoelectric materials refers to their dimensionless figure of merit (ZT), defined by equation: $ZT = \frac{\sigma\alpha^2}{K}T$, where α is the Seebeck coefficient (μV K^{-1}), σ the electrical conductivity (Ω^{-1} m^{-1}), K the thermal conductivity (W m^{-1} K^{-1}) and T the absolute temperature (K). Nowadays, Mg_2X (X = Si, Sn, Ge) semiconductor compounds and their solid solutions have drawn more attention due to their thermal stability, low cost, none toxicity, constituent abundance in the earth's crust, environmentally friendly, low density and providing both n and p-type conductions. Most of the studies about Mg_2X compounds are limited to n-type materials and the development of p-type compounds is still a challenging task. Many works were focused on bulk materials, while development of thin film materials with optimized ZT is required to reduce the size of current thermoelectric devices in order to address them in miniaturized applications like Micro Electronic and Mechanical Systems (MEMS) and Internet of Things (IoT). Mg_2Sn is a narrow band gap semiconductor and Mg_2Sn thin films have been not extensively investigated in the literature. Some strategies are developed to improve ZT of Mg-based thermoelectric materials such as alloying, solid solutions, nano-structuring, band engineering, and magnesium deficiency.

In the present work, the influence of Cu-doping and Mg-deficiency on ZT of Mg_2Sn was studied. In the first part, Mg-Sn thin films ($21 \leq$ at. % Sn ≤ 42.5) were synthesized by co-sputtering of Mg and Sn targets in an argon atmosphere. The structure and morphology of the films were characterized as a function of the film composition. Mg_2Sn structure was changed from stable face-centered cubic to metastable orthorhombic structure while the content of Sn in the films increased. The film carrier concentration and mobility were measured to explain the electronic transport behavior as a function of the film structural modifications. The influence of this structural modification on thermoelectric properties was discussed in a wide range of temperatures (30-200 °C). The highest ZT was obtained about 0.26 at 200 °C for the film with 36 at. % Sn.

In the second part, two groups of Mg_2Sn thin films were deposited by co-sputtering of Mg, Sn and Cu targets in an argon atmosphere. Group I with Mg: Sn (67:33) and group II with Mg: Sn (64:36) were doped by different at. % Cu (0, 0.5, 1.5). The structure and morphology of thin films were characterized as a function of the Cu atomic concentration. All films were crystalized in face-centered cubic structure. The film carrier concentration and mobility were measured to explain the electronic transport behavior as a function of the film composition. The influence of Cu-doping on the thermoelectric properties of thin films was discussed as a function of the temperatures. The highest ZT was reached about 0.08 at 180 °C, 0.27 at 200 °C for the group I and group II respectively.

The results show that the effect of Mg-deficiency on ZT improvement is stronger than doping.