Li で部分置換した Mg_2Sn 単結晶の結晶構造と熱電特性

Crystal Structures and Thermoelectric Properties of Li-doped Mg_2Sn Single Crystals

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Mg_2Sn is a promising mid-temperature thermoelectric (TE) material consisting of earth-abundant, low cost, and less-toxic elements. We successfully prepared Mg_2Sn single crystals (SCs) with Mg vacancy (V_{Mg}) by melting method under Ar pressure [1]. The V_{Mg} has an effect of lowing lattice thermal conductivity (κ_{lat}) of Mg_2Sn, which is lower than those of the Mg_2Sn polycrystals (PC) ever reported. Furthermore, we prepared n-type $Mg_2Sn_{1-x}Sb_x$ (x = 0.005, 0.01, and 0.02) SCs and investigated the effect of V_{Mg} and Sb atoms on their crystal structures and TE properties [2]. The higher carrier mobility of $Mg_2Sn_{1-x}Sb_x$ SCs led to a higher power factor (PF = 5.1 $mW \cdot m^{-1} \cdot K^{-2}$) than that of the PC, which resulted in the maximum dimensionless figure of merit (zT_{max}) for the $Mg_2Sn_{0.99}Sb_{0.01}$ SC (~ 0.72 at 650 K).

Compared with n-type Mg_2Sn, p-type Mg_2Sn shows a lower zT value. So far, Ag, Li, Na, and Ga doping has been used to obtain p-type Mg_2Sn by increasing the hole carrier concentration. The zT_{max} was obtained for an Ag-doped Mg_2Sn large-grain PC with finely dispersed Mg_2Sn + MgAg eutectic clusters (zT = 0.3 at ~500 K) [3] and for a Li- doped Mg_2Sn PC (zT = 0.3 at 700 K) [4]. Recently, we prepared $Mg_2Sn_{1-x}Ga_x$ SCs and investigated their TE properties. Although we obtained relatively low κ_{lat} for $Mg_2Sn_{0.98}Ga_{0.02}$ SC, the zT value (zT_{max} = 0.18@450 K) was still not high enough because of the low carrier concentration ($n = 5.92 \times 10^{19}$ cm^{-3}) and low PF (PF_{max} = 1.5 $mW \cdot m^{-1} \cdot K^{-2}$) [5].

In this work, a series of $Mg_{2-x}Li_xSn$ (x = 0, 0.005, 0.01, 0.02, 0.03) SCs with V_{Mg} were prepared by the melting method, whose morphology and TE properties were investigated with a particular emphasis on the effects of doped Li atoms and V_{Mg}. With Li content x increased, σ increased over the whole temperature range. The electrical conductivity of the x = 0.02 SC was higher than that of the Ga-doped SC [5] and was similar with that of the Li-doped Mg_2Sn SC [4]. Significantly, the Seebeck coefficient of the x = 0.02 SC was higher than that of the Li-doped Mg_2Sn PC [4]. We assumed that this behavior was affected by a larger carrier effective mass (m^*). As a result, the PF (~ 2 $mW \cdot m^{-1} \cdot K^{-2}$ for x = 0.02 SC) was higher than that of the Li-doped PC [4]. In addition, the κ was lower than that of the Li-doped PC [4] due to the enhanced phonon scattering by the introduced V_{Mg}. Finally, the zT_{max} value of 0.36 was achieved for $Mg_{1.98}Li_{0.02}Sn$ at 600-650 K, which was 20% higher than that of the reference[4].

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