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Enhanced Thermoelectric Properties of the $Zn_{1-x}Cd_xSb$ Prepared by Solid State Microwave Synthesis

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Abstract. This paper displays the fabrication of a thermoelectric (TE) generation module using n-ZnSb and p- $Zn_{0.25}Cd_{0.75}Sb$ bulk TE materials. TE properties of the $Zn_{1-x}Cd_xSb$ bulks with x= 0, 0.5 and 0.75, in terms of the electrical conductivity (σ) and Seebeck coefficient (S) were measured in the range of 300-500K. The higher power factor ($S^2\sigma$) values for n-ZnSb and p- $Zn_{0.25}Cd_{0.75}Sb$ bulks were obtained about 2.4×10^{-4}mW/mK^2 at 303K and 1.18×10^{-5} mW/mK^2 at 468K, respectively. By variation of the thermal conditions, the maximum output power (P_{max}) with two p-n couples generator module was 1.38×10^{-5} mW at hot side temperature of 355K and temperature difference (ΔT) of 40K. The internal (R_{in} = 0.17 mΩ) and contact resistances (R_c = 0.67 mΩ) between legs and electrodes were discussed below.

Introduction

Recently, ZnSb and CdSb have been famed as new TE materials with high performance at intermediate temperatures [1, 2]. The physical properties of the $Zn_{1-x}Cd_xSb$ compounds have been first investigated by Ugai et al. [3]. Most literatures refer the polycrystalline ZnSb, doped has been investigated as a p-type semiconductor [4, 5–8]. Therefore, to obtain a TE generation module, we need to pair two semiconductors of p and n-type. The Zn–Cd–Sb compounds have appearance more complex structure which resulted in enhanced TE properties. Furthermore, ZnSb and CdSb are grown as rhombohedral structure with R-3c space group at 300 K [9]. For the optimal results, ZnSb and CdSb can be mixed as $Zn_{1-x}Cd_xSb$ ternary alloys which is particularly interesting because of having narrow energy band gap is eminently suitable for various TE devices [1, 4]. Different methods have been utilized to prepare Zn–Cd–Sb bulk materials, such as: encapsulating sintering [10], quenching method [1], direct melting [4], vacuum casting [3], and solid state reaction [11].

Enhanced TE properties of semiconductors such as the σ and S properties are needful for development and fabricate of TE generation modules. Furthermore, the power factor is necessary obtained to provide information about the TE properties for the available materials. The aim of this research is to study the TE properties of new $Zn_{1-x}Cd_xSb$ (x=0, 0.5 and 0.75) bulk materials were prepared via a solid-state microwave method and fabrication of TE generation module using the optimal TE data

Experimental

High purity Zn, Cd and Sb as raw materials (99.999%) were used in this paper to perform two grams for each samples of $Zn_{1-x}Cd_xSb$ (x=0, 0.5 and 0.75) bulks using the solid-state microwave synthesis [12]. The TE bulk materials were ground onto fine powder and then pressed into disks by cold pressing method at 10 tons and the thickness of the disks were 0.2 mm. The TE characteristics were measured using a home-built equipment [13]. The variation of thermal conditions was desired to generate the open circuit voltage (V_{oc}) and P_{max}.

Results and Discussion

The TE properties of n-ZnSb and p- $Zn_{1-x}Cd_xSb$ (x=0.5 and 0.75) bulk materials such as σ, S and $S^2σ$ at the range 300-500K are shown in Figs. 1 to 3.The σ for all the TE bulk samples had the same (semiconductor) behavior, where increased with the temperature (Fig. 1).The σ value for the n-ZnSb changed from 817.6 S/m at 300 K to 1371 S/m at 475K, whereas σ for the p- $Zn_{1-x}Cd_xSb$ samples was from 271 and 138 S/m at 300 K for x=0.5 and 0.75 respectively, to 401 and 206 S/m at 475 K for x=0.5 and 0.75, respectively. These results were due to the existence of trapping states, which were capturing charge carriers.

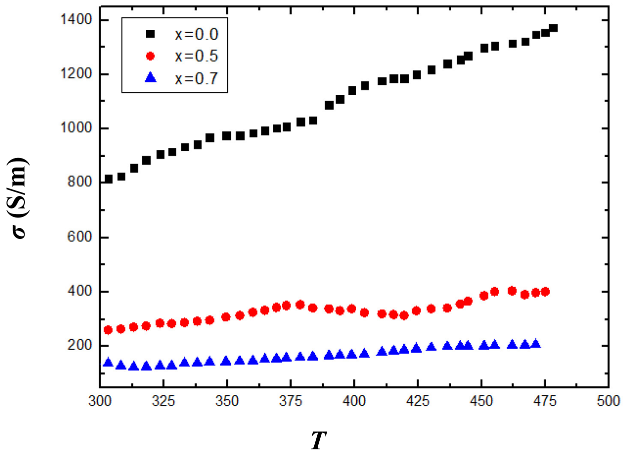


Fig. 1. σ as a function of T for $Zn_{1-x}Cd_xSb$ (x=0, 0.5 and 0.75) bulk TE materials.

From Fig. 2, S for the n-ZnSb was -542 μV/K at 300 K and for p- $Zn_{1-x}Cd_xSb$ samples was 190 μV/K (x=0.5) and 261 μV/K (x=0.75) at 500 K. At higher temperature, the value of S in the n-type sample decreased because of higher thermal excitation in the charge carriers than it is in the lower temperature region. The increasing in the value of S in the p-type sample is consistent with the Mott formula [13, 14].

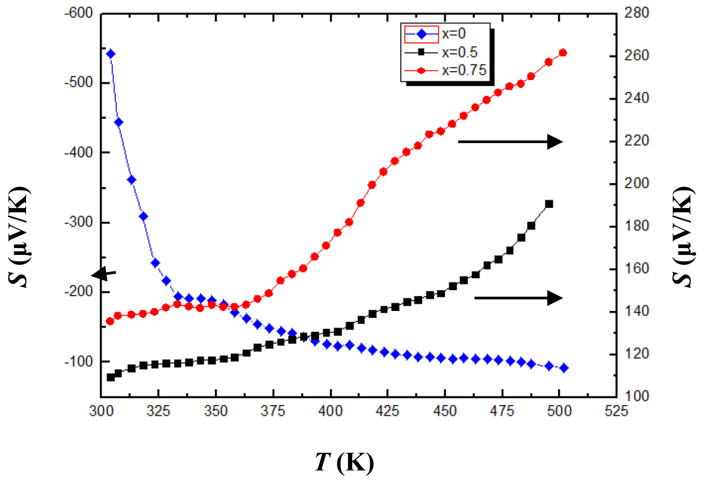


Fig. 2. S as a function of T for $Zn_{1-x}Cd_xSb$ (x=0, 0.5 and 0.75) bulk TE materials.

The $S^2σ$ for the $Zn_{1-x}Cd_xSb$ bulk samples as a function of the temperature range from 300 to 500 K was shown in Fig. 3. The $S^2σ$ can be calculated from σ and S properties, which posses the same behavior of the Seebeck coefficient, because the later property had the dominant value. The maximum calculated $S^2σ$ for the ZnSb (x=0) at 303K was 2.4×10^{-4} mW/mK^2, while, 1.1×10^{-5} mW/mK^2at 473K for the $Zn_{0.5}Cd_{0.5}Sb$ (x=0.5) sample and 1.18×10^{-5} mw/mK^2 at 468K for the $Zn_{0.25}Cd_{0.75}Sb$ (x=0.75) sample. At 300-400K, The $S^2σ$ the $Zn_{0.5}Cd_{0.5}Sb$ (x=0.5) sample higher than that for the $Zn_{0.25}Cd_{0.75}Sb$ (x=0.75) sample, which can be ascribed to the increased σ and moderate S for the sample with x=0.5. The optimal $S^2σ$ values for n-ZnSb and p- $Zn_{0.25}Cd_{0.75}Sb$ contribute to a P_{max} for our TE generator module.

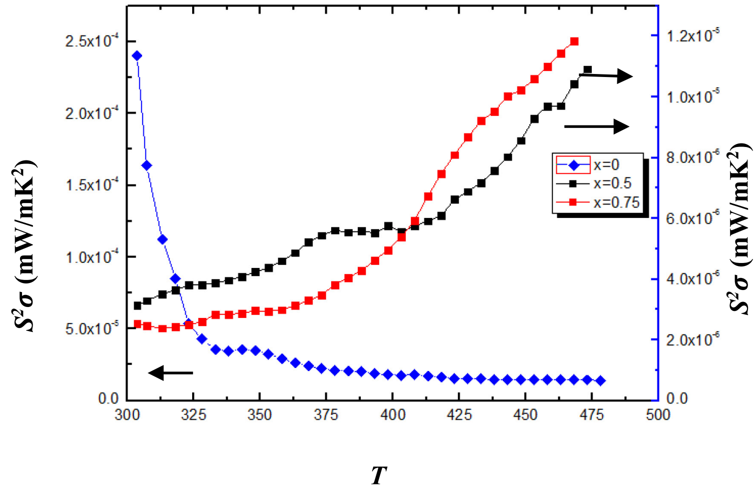
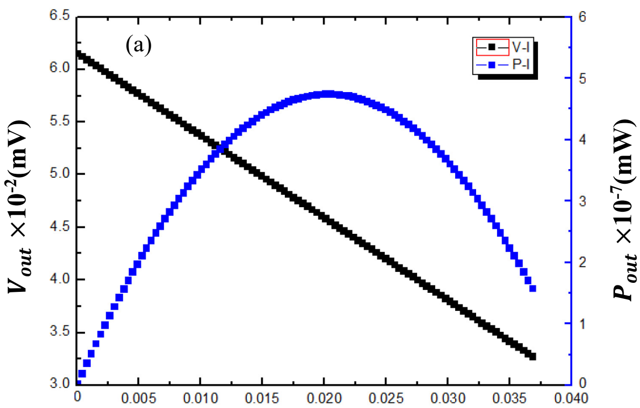


Fig. 3. $S^2\sigma$ as a function of T for $Zn_{1-x}Cd_xSb$ (x=0, 0.5 and 0.75) bulk TE materials.

From Fig. 4 and Table 1, the output voltage (in mV) and the output power (in mW) of the two (n- and p- types) couples as a function of the current (in mA) were measured at several temperature conditions (ΔT=10, 20, 30 and 40K). From I-V lines the V_{oc} can be calculated at I=0 and were reached 6.2×10^{-2}, 8.5×10^{-2}, 17.4×10^{-2}, and 20.4×10^{-2} mV (Table 1) at ΔT=10, 20, 30, and 40 K, respectively, which were lower than that calculated curves in Fig. 2. This is because the alumina substrate had low thermal conductivity or the junctions between the two legs and the electrodes were unfavorable [13].

Table 1: V_{oc} and P_{max} at various ΔT for the TE generator module.

ΔT (K)	V_{oc} (mV)	P_{max} (mW)
10	6.2×10^{-2}	4.75×10^{-7}
20	8.5×10^{-2}	2.08×10^{-6}
30	17.4×10^{-2}	7.28×10^{-6}
40	20.4×10^{-2}	1.38×10^{-5}



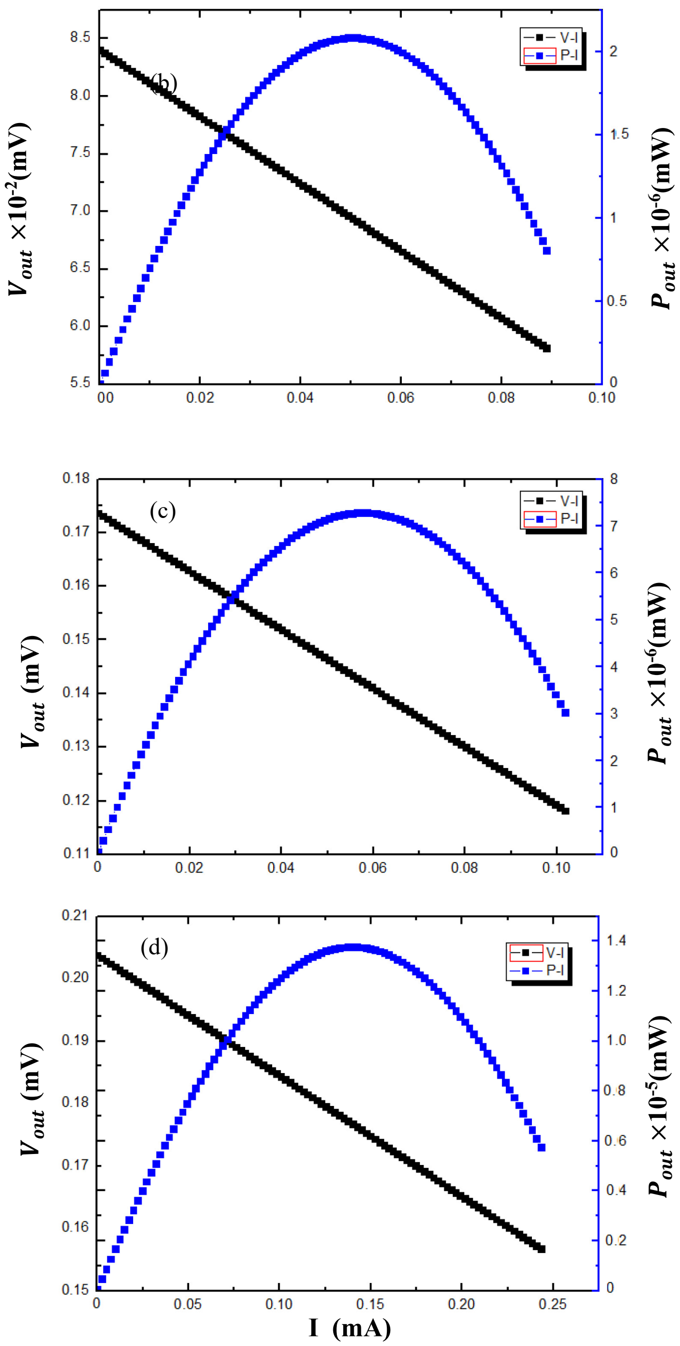


Fig. 4. V_{oc} and P_{max} as a function of I of the TE generator module, where (a) $\Delta T = 10$ K, (b) $\Delta T = 20$ K, (c) $\Delta T = 30$ K, and (d) $\Delta T = 40$ K.

The parabolic P-I curves are illustrated in Fig. 4. The P_{max} values were 4.75×10^{-7}, 2.08×10^{-6}, 7.28×10^{-6}, and 1.38×10^{-5} mW (Table 1) at ΔT=10, 20, 30, and 40K, respectively, which means these results improved by increasing the temperature. From the I-V lines slope, the R_{in} of TE generator module was calculated by the measured system. The R_c can be achieved by: $R_c = R_{in} - R_{id}$, where R_{id} is the ideal internal resistance was obtained by sum of the p- and n-type samples resistance values. The resistance values of the TE generator module are 0.17 and 0.67 mΩ for R_{in} and R_c , respectively. The minimized value of the R_c plays a key role in the performance of TE generator module.

Conclusion

TE bulk materials n-ZnSb and p- $Zn_{0.25}Cd_{0.75}Sb$ were prepared via solid-state synthesis method. The good properties of TE generator module originally came from high TE properties (σ, S and $S^2\sigma$) of n- and p-type samples. At ΔT=40K, the V_{oc} and P_{max} values were reached to 20.4×10^{-2} mV and 1.38×10^{-5} mW, respectively, which means these results improved with increasing the temperature. The enhanced TE performances of the TE generator module proposes the considerable potential of low temperature $Zn_{1-x}Cd_xSb$ TE materials towards future applications.

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