Fabrication and Characterization of Thermoelectric Thick Film by Using Bi-Te-Sb Powders

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Abstract

Thermoelectric thick film was fabricated by screen printing process with using p-type Bi-Te-Sb powders. The powder was synthesized by melting, milling and sintering process and hydrogen reduced to enhance the thermoelectric property. The thick film of Bi-Te-Sb powder was fabricated by screen printing method and baked at the optimized conditions. The thermal conductivity, the electrical resistivity and Seeback coefficient of thick film were measured and the thermoelectric performance was analyzed in terms of film characteristics and its microstructure. Finally, the feasibility of thermoelectric thick film into micro cooling device on CPU chip was discussed in this study.

Keywords : Thermoelectric, Thick film, Screen printing, Micro cooling, Bi-Te-Sb thermoelectric powders

1. Introduction

Thermoelectric cooling is a suitable technique for the local cooling of micro-sensor and devices as it does not require any moving parts and can be microelectronically integrated. While the search for thermoelectric materials compatible with solid-state electronics material continues, tellurium compounds currently have the highest cooling performance aroud room temperature.

Thick films (10-50 μm) of tellurium alloys have been deposited using electroplating for fabrication of thermoelectric coolers where the current and heat flow perpendicular to the film plane (column-type design) [1,2]. Yao et al. [3] have increased the performance of this design by optimizing the geometry and using suitable materials for both the thermoelectric legs and the substrate.

Due to the parasitic conduction heat transfer between hot and cold juctions, and the thermal and electrical contact resistances, thin film (less than 1 mm) have not been used in conventional column-type thermoelectric coolers. These problems can be minimized, if thicker films (2-10 mm) are used.

In this study, we tried to thermoelectric thick film of P-type Bi-Te-Sb powders by screen printing process which is feasible to pattern to the thermoelectric module. The structure and thermoelectric properties of Bi-Te-Sb thick film have been investigated with the change of processing parameters.

2. Experimental and Results

The raw materials of 20%Bi_2Te_3 + 80%Sb_2Te_3 for p-type thermoelectric powder was used in this study. The constituent elements with purity of 99.999% in the proper proportions were melted at 700°C in a sealed and evacuated quartz tube. A rocking furnace at a certain frequency was used to ensure the composition homogeneity without any segregation. The melted ingots were crushed into powder using a ball mill for 24 hours and sieved with 100 mesh to classify the powder size of less than 320 μm. The material media of ball (7.76 mm diameter) and jar was stainless steel, and the mixing ratio of powder to ball was 1:7 in mass (refer to Fig 1). The ball milled powders were mixed with binders and printed on alumina subtrate with 8×8 mm square type screen. Drying of the as-printed samples was carried out in air for 5 hours and fired in programmed muffle furnace in air at 400°C for 2 hours with the heating rate of 2°C/min. This sample was sintered by hot press and spark plasma sintering (SPS) processes. SPS was carried out at 350-400°C under a pressure of 10-70 MPa for 5 min. Also hot press was carried out up to 400°C under a pressure of 60 MPa in hydrogen atmosphere. Before hot press, the samples were pre-sinted at 250°C for 30 min. The electrical conductivity was measured using Hall measurement system and the concentration of carrier and its mobility was analysed with sintering conditions.

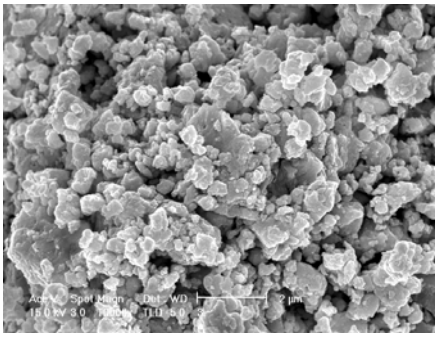

Fig. 1. SEM micrograph of ball milled Bi-Te-Sb p-type thermoelectric powder.

3. Results and Discussions

Fig. 2 shows the micrographs of screein printed thermoelectric module (a) and the cross sectional area of the screen printed and sintered sample by SPS process (b). In this figure, Ag electrode layer of around 10 μm in thickness was coated on alumina subtrate and Bi-Te-Sb p-type thermoelectric powder of around 200 m in thickness was deposited on Ag electrode layer.

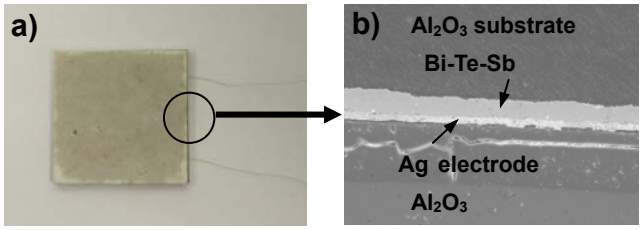

Fig. 2. The micrograph of screen printed thermoelectric module (a) and its cross sectional area sintered at 350°C for 5 min. by SPS process.

The density of SPSed samples increased with the increasing of pressure and the change of electrical resistivity was depicted in Fig. 3 with the applied pressure. The electrical resistivity of SPSed thick film drastically decreased with the increasing of applied pressure. The lowest electrical resistivity of SPSed sample was 15.11×10^{-5} Ωm at 70 MPa pressure condition that is 10 times higher than bulk materials (aroud 1.2×10^{-5} Ωm). This high electrical resistivity thought to be resulted from the relatively lower sintered density than that of bulk and the residual organic materials remaining after debinding process.

However, more lowder electrical resistivities could be obtained by hot pressed samples. The sample hot pressed at 400°C has lower electrical resistivity of 7.08×10^{-5} Ωm which is quite close to that of bulk materials. But the residual organics should be controlled during sintering process as well as sintered density in further study.

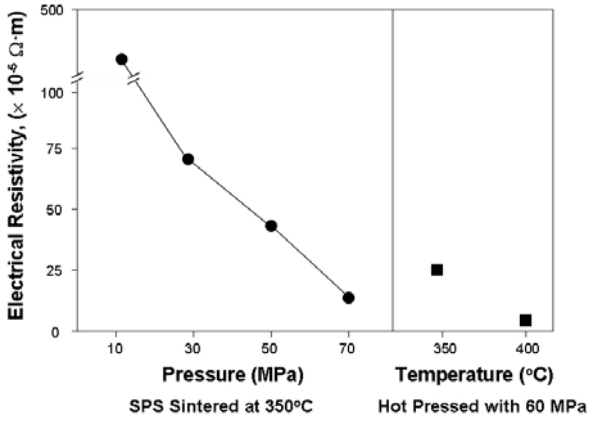

Fig. 3. The variation of electrical resistivities of Bi-Te-Sb screen printed materials sintered by SPS and hot press process.

4. Summary

The thick film of p-type Bi-Te-Sb thermoelectric materials was successfully fabricated by screen printing process and sintered by SPS and hot press. The thickness of film was around 250 mm and showed sound micrograph of layers. The electrical resistivity decreased with increasing of applied pressure during SPS sintering and with increasing of sintering temperature during hot press process. But the values of electrical resistivities have much higher than that of bulk materials due to the relatively low sintered density and the residual organics after debinding process. The designing for improving the electrical properties should be done in further works.

5. References

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