Gas Atomization and Consolidation of Thermoelectric Materials

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Abstract

The n-type (95%Bi_2Te_3-5%Bi_2Se_3) compound was newly fabricated by gas atomization and hot extrusion, which is considered to be a mass production technique of this alloy. The effect of powder size on thermoelectric properties of 0.04% SbI_3 doped 95%Bi_2Te_3-5%Bi_2Se_3 alloy were investigated. Seebeck coefficient (α) and Electrical resistivity (ρ) increased with increasing powder size due to the decrease in carrier concentration by oxygen content. With increasing powder size, the compressive strength of 95%Bi_2Te_3-5%Bi_2Se_3 alloy was increased due to the relative high density. The compound with ~300 ㎛ size shows the highest power factor among the four different powder sizes. The rapidly solidified and hot extruded compound using 200~300 ㎛ powder size shows the highest compressive strength.

Keywords : Thermoelectric Materials, Gas Atomization, Powder Metallurgy, BiTe-BiSe

1. Introduction

Thermoelectric materials have been widely used in both cooling devices of electronic parts such as computer IC chips, laser diodes, infrared detective devices etc and electronic cooling devices of car refrigerator, cool and hot clean water machine. Semiconducting bismuth telluride (Bi_2Te_3) and antimony telluride (Sb-Te) alloys regards as one of the attractive materials at medium temperature thermoelectric conversion [1-2]. The commercial thermo elements used in the thermo modules are mainly prepared by the unidirectional solidification such as single crystal. It was already reported that new process such as gas atomization and extrusion process can improve the strength and thermoelectric properties [3]. However, the wide size distribution in gas atomized powder is commonly observed and cooling rate varies depending on powder size. It is expected that microstructure and oxygen content of consolidated alloy vary with initial alloy powder size and this effects will be able to affect the thermoelectric properties.

In this work, the effect of powder size on the thermoelectric properties and compressive strength of n-type 95%Bi_2Te_3- 5%Bi_2Se_3 compound was studied.

2. Experimental and Results

Prepared high purity (99.99%) Bi, Te and Se element were melted in the high frequency induction in graphite crucible under argon atmosphere, and atomized by N_2 gas. Fabricated 95%Bi_2Te_3-5%Bi_2Se_3 alloy powders with 0.04 wt% SbI_3 were sieved the size of ~45 ㎛, 75~125 ㎛, 200~300 ㎛ and ~300 ㎛, respectively. Sieved powders were reduced in the H_2 reduction furnace at 360 °C for 4 h. Reduced powders were respectively consolidated in Al alloy can by cold compaction at room temperature and degassed at 450℃ for 1h by vacuum pump. Extrusion was performed at 450℃ with a extrusion ratio of 16:1. For the microstructure analysis of powder and extruded bar, Scanning Electron Microscope and Optical Microscope were used. Phase analysis of powders and extruded bars were characterized by conventional X-ray diffraction using monochromatic Cu-K_α radiation. Thermoelectric properties were measured at parallel section to extrusion direction. Seebeck coefficient(α) of the extruded materials were measured to form 5×5×10mm at room temperature. Electrical resistivity (ρ) was measured to form 2×2×10mm by four point probe at room temperature. Compressive strength of extruded bars were measured by Material Test System (MTS).

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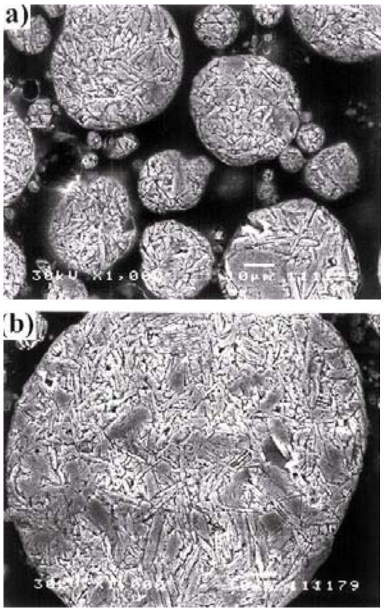

Fig. 1. SEM micrographs of 95%Bi_2Te_3-5%Bi_2Se_3 alloy powders: (a) ~45, (b) 75~125 μm.

Fig. 1 shows the typical morphology of the gas atomized 95% Bi_2Te_3-5%Bi_2Se_3 alloy powders with powder size as observed by SEM. The microstructures of the gas atomized powders show a homogeneously distributed needle shape compounds. With increasing the powder size, the width and length of needle shape compounds are increased. The microstructure of extruded bar shows a homogeneous and fine distribution of grains in all extruded bars. The grain size tends to be increased with increasing powder size. Fig. 2 shows the relative density of hot extruded bar with powder size. The relative density of extruded bar is increased with increasing the powder size. The different hardness of the initial powder is likely to affect the consolidation density. The oxygen content of the gas-atomized powder decreases with increasing powder size because of low specific area at the coarse powder. The Seebeck coefficient(~45 μm:195, 75~125 μm:243, 200~300 μm:252, ~300 μm:225 |α| [μV/K]) of extruded bar increases with increasing the power size. The tendency can be explained by decrease of carrier concentration that is proportional to the oxygen content. The Seebeck coefficient was increased with increasing powder size indicating decrease in the oxygen content. The electrical resistivity of the extruded bar as a function of powder size is increased with increasing powder size. The increase of electrical registivity with increasing powder size is because of decreasing of carrier concentration by oxygen content increase. The power factor(~45 μm:3.4, 75~125 μm:3.5, 200~300 μm:2.1, ~300 μm:3.6 [$\times 10^{-3}$ w/mK]) of 95%Bi_2Te_3-5%Bi_2Se_3 alloy is increased with increasing powder size and the highest powder factor was obtained at ~300 μm powder size range.

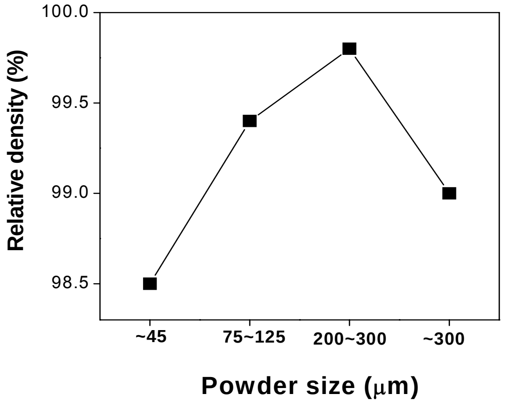

Fig. 2. Variation of density of extruded bars with powder size.

3. Summary

The effect of powder size on thermoelectric properties of extruded 95%Bi_2Te_3-5%Bi_2Se_3 alloy was investigated. The oxygen contents of the gas atomized powder decrease with increasing powder size due to low specific area at the coarse powder. The oxygen content of the reduced powder at 360 °C for 4h shows the same trends as with increasing powder size, the oxygen content increase. The Seebeck coefficient of extruded bar increases with increasing the power size. The tendency can be explained by decrease of carrier concentration that is proportional to the oxygen content. The highest value of powder factor is 3.6 $\times 10^{-3}$ w/mK at the extruded bar from ~300 μm powder. With variation of powder size to ~45 μm, 75~125 μm, 200~300 μm and ~300 μm compressive strength shows the value of 207.6, 265.7, 338.2 and 231.3 MPa, respectively.

4. References

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