第68回応用物理学会春季学術講演会 講演予稿集 (2021 オンライン開催)

Thermoelectric properties of p-type Si-Ge-(Fe,Ni,V,Ti)-B alloys

○(PC)Omprakash Muthusamy[1], Saurabh Singh[1,3], Masahiro Adachi[2], Yoshiyuki Yamamoto[2], Tsunehiro Takeuchi[1,3,4,5]

[1]Research Center for Smart Energy Technology, Toyota Technological Institute, Nagoya 468-8511, Japan
[2]Sumitomo Electric Industries, Ltd., Hyogo 664-0016, Japan.
[3] CREST, Japan Science and Technology Agency, Toyota Technological Institute, Tokyo 102-0076, Japan.
[4] MIRAI, Japan Science and Technology Agency, Toyota Technological Institute, Tokyo 102-0076, Japan.
[5] Institute of Innovation for Future Society, Nagoya University, Nagoya 464-8603, Japan.

E-mail : omprakashmuthusamy@gmail.com

Introduction

In order to improve the figure of merit *ZT*, many strategies have been reported. For example, nano-structuring and modulation doping approach has been utilized to improve the thermoelectric properties of Si-Ge alloy by decreasing thermal conductivity without degrading the power factor [1].

Our previous report showed that B-doped Si-Ge-Au thin film and bulk sample possessed *ZT* value of 1.38 and 1.63 at 1000K. We found that the electron transport properties were constructively improved using Au-doping to form an impurity states near the valence band top, and B-doping to control the Fermi level. Very small thermal conductivity ~ 1.5 $Wm^{-1}K^{-1}$ was obtained due to nanograins. However, since Au is an expensive element, that avoid us to move further device application. Therefore, we need to find other metals which cheap, non-toxic, and environmentally friendly.

Yamada *et al.*, Calculated the DOS of Si-Ge by various transition metal doping [3]. It revealed that large Seebeck coefficient could be obtained by making sharp peak near valence or conduction band edge with different transition metal doping such as Mn, Ni, Co, Cu, Fe, and Zn.

These metals are cheap, non-toxic, and environmentally friendly. In this study, therefore, we synthesized bulk noncrystalline p-type Si-Ge alloy with a small amout of one of the 3d-transition metal elements together with boron. Thermoelectric properties (TE) of bulk nanocrystalline samples were investigated as a function of temperature 300 – 1000 K, respectively.

Experimental Procedure

High purity metal, Ge (5N), Si (5N), and B powders were sealed in a Zirconia container with Zirconia balls in the glove box under pressurized Ar gas atmosphere. The mixed powders were milled by using high energy planetary milling system. Afterwards, the powders were subsequently sintered with a relatively high pressure of 400 MPa at 873 ~ 1003 K for 4 h, respectively.

Results and discussion

The powder XRD indicated that the single phase of diamond structure was obtained after the ball-milling. After sintering, The size of nano-crystals in the bulk sample was estimated to be 10 – 30 nm from the XRD peaks and the Scherrer equation. We found that grain size is controllable with sintering condition, but sample density is also strongly affected by it.

One of the transition metal substituted Si-Ge shows the large Seebeck coefficient of 321 $\mu V^{-1}K^{-1}$ at 1000 K presumably due to presence of impurity states near the valence band. The electrical resistivity was 4.4 mΩcm at 1000 K. As a consequence, a very large power factor reaching 2.3 $mWm^{-1}K^{-2}$ together with low thermal conductivity 1.47 $Wm^{-1}K^{-1}$ allow us to obtain high *ZT* = 1.59 at 1000 K, Which was closed to the previously reported Au and B co-doped (*ZT* = 1.63) [2].

The TE properties of various transition metal substituted Si-Ge will be discussed together with the best condition leading to the best balance of grain size and density.

References

[1] N. Mingo *et al.*, *Nano Letters* **9**, 71, (2009).
[2] M. Omprakash *et al.*, *JJAP.* **58**, 125501 (2019).
[3] R. Yamada *et al.*, *Solid State Comm.* **323** (2021) 114115.