Large evolution of *ZT* in p-type nanocrystalline bulk Si-Ge

Muthusamy Omprakash[1], Ghodke Swapnil[1], Kévin Delime-Codrin[1],

Masahiro Adachi[2], Yoshiyuki Yamamoto[2], and ◦ Tsunehiro Takeuchi[1],

[1]Toyota Technological Institute, Hisakata 2-12-1, Tempaku, Nagoya 468-8511, Japan

[2]Sumitomo Electric Industries, Ltd., Konyo Kita 1-1-1, Itami, Hyogo 664-0016, Japan.

E-mail : omprakashmuthusamy@gmail.com

Introduction

The Si-Ge alloys are a cheap and non-toxic thermoelectric materials usable for high temperature applications. In order to improve its figure of merit $ZT = S^2\sigma/\kappa$, we succeeded in decreasing thermal conductivity using nano-structring by means of high-energy ball milling and low temperature sintering. [1] However, the *ZT* value wasn't improved mainly because of its low power factor.

We tried to improve power factor of Si-Ge thermoelectric cmaterials by use of impurity elements that produce impurity states near the chemical potential with keeping the low thermal conductivity using the nano-structuring. As a result, we succeeded in obtaining ZT =1.88 for a n-type Si-Ge material. [2]

In this study, therefore, we tried to develop corresponding p-type Si-Ge bulk thermoelectric materials possessing a large *ZT*-value far exceeding unity by using the same techniques.

Experimental Procedure

Sampled consisting of nano-crystalline powder were prepared using ball milling at 600 rpm for 6 h at the composition of of $Si_{0.55}Ge_{0.35-x-y}Au_xB_y$. The prepared powders were sintered using moderately high pressure of 500 MPa at low temperature below 700°C with pulse current sintering technique in vacuum atomsphere. The density was measured by Archimedes' method with ethanol as the working liquid.

Thermal conductivity was measured by laser flash method from 300 K to 1000 K. Seebeck coefficient *S* and electrical resistivity was measured in a hand-made apparatusover the temperature range from 300 K to1100 K.

Results and discussion

From the first principle culations, we confirmed that Au 5d orbitals produce impurity states in tbe band gap. When the Au concentration was small enough to be a few at.%, the impurity states are rather isorated to make sharp peaks in the electronic density of states. As a typical example, the electronic density of states calculated for the solidi solution of Si containing 0.9at.%Au is plotted in Fig.1.

We confirmed that the averaged grain size in the sintered sample was less than 20 nm, and the lattice thermal conductivity was reduced less than 1.4 $Wm^{-1}K^{-1}$. We also confirmed that the holes were effectively introduced by B-doping, and that the electrical resisitivty possessed tyipical hebavior of disordered metal showing neglegiblly small temperature dependence with rather large magnitude of ~10mΩcm. Seebeck coefficient was increasing with increasing temperature up to 1000 K with showing no evidence of bi-polar effect. The magnitude was also kept larger than 400 μVK^{-1}. These thermoelectric properties led a significant increase in *ZT*-value, that will be shown in the presentation.

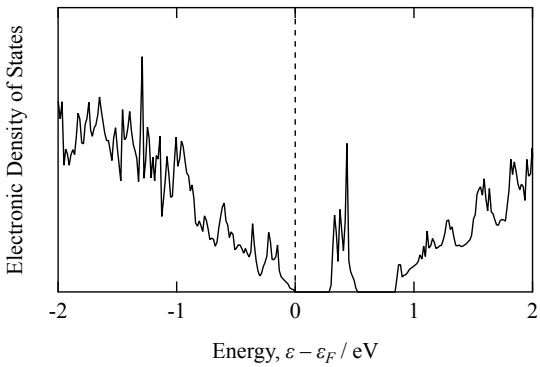

Figure 1 : Electronic density of states calculated for $Si_{143}Au$ of diamond struvture. An intense peak was produced by Au 5d orbitals.

References

[1] M. Omprakash et al., *J. Electron. Mater* **47**, (2018).

[2] K. Deline-Codrin, submitted to APEX.