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# Effect of Heavy Element Substitution and Off-Stoichiometric Composition on Thermoelectric Properties of $Fe_2$VAl-Based Heusler Phase

TSUNEHIRO TAKEUCHI,[1,2,3,4,6] YUICHI TERAZAWA,[3] YUKIHIRO FURUTA,[3] AKIO YAMAMOTO,[4] and MASASHI MIKAMI[5]

1.—EcoTopia Science Institute, Nagoya University, Nagoya 464-8603, Japan. 2.—Precursory Research for Embryonic Science and Technology, Japanese Science and Technology Agency, Tokyo 102-0075, Japan. 3.—Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan. 4.—Department of Crystalline Materials Science, Nagoya University, Nagoya 464-8603, Japan. 5.—National Institute of Advanced Industrial Science and Technology, Nagoya 463-8560, Japan. 6.—e-mail: takeuchi@nuap.nagoya-u.ac.jp

The thermoelectric performance of $Fe_2VAl$-based alloys was improved by using the effects of (a) heavy element substitution and (b) off-stoichiometric (Fe/V $\neq$ 2) composition. The former method led to a significant reduction of lattice thermal conductivity, whereas the latter to an evolution of the Seebeck coefficient. As a result of sample preparation, we confirmed that the dimensionless figure of merit with $n$-type behavior was increased up to 0.25 at 420 K for the sample obtained at the optimized composition of $Fe_{1.98}V_{0.97}Ta_{0.05}Al_{0.9}Si_{0.1}$. Electronic structure calculations revealed that the increase of the Seebeck coefficient observed for Fe-poor samples was caused by a reduction of the density of states near the chemical potential.

**Key words:** Seebeck coefficient, lattice thermal conductivity, dimensionless figure of merit, first-principles calculations

## INTRODUCTION

Recently, thermoelectric generators have attracted considerable interest because of their ability to recover energy from waste heat. Since a large part of waste heat is released to the environment at low temperatures below 500 K, it is of great importance to develop thermoelectric generators usable in this particular temperature range. Since a large number of generators must be used for effective recovery of energy from low-temperature waste heat that is lost diffusively, the materials used in such generators must also be harmless and of low cost.

We considered that the Heusler-type $Fe_2VAl$ alloy is one of the most plausible candidates for such thermoelectric materials, because it consists solely of ubiquitous, harmless, cheap elements and possesses a large power factor PF = $S^2\sigma$ exceeding $3 \times 10^{-3}$ W m$^{-1}$ K$^{-2}$ at around 300 K to 400 K.[1–3] Here, $S$ and $\sigma$ indicate the Seebeck coefficient and electrical conductivity, respectively. Unfortunately, the magnitude of $ZT$ observed for Heusler-type $Fe_2VAl$-based alloys[4] is limited to small values below 0.1 due to the large magnitude ($\sim$20 W m$^{-1}$ K$^{-1}$) of the lattice thermal conductivity $\kappa_{lat}$. If the magnitude of $\kappa_{lat}$ of Heusler-type $Fe_2VAl$-based alloys were to be effectively reduced without greatly affecting the power factor, such materials could be widely used as environmentally friendly, practical thermoelectric materials.

To effectively reduce the magnitude of the lattice thermal conductivity, we employed, in our previous study,[5,6] partial substitution of heavy elements for the constituent elements in $Fe_2VAl$-based alloys. This strategy was employed because the less obvious temperature dependence of electrical resistivity at high temperatures above 300 K[1] suggested that

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