

# Preparation of Eco-Friendly Fe<sub>2</sub>VAl-Based Thermoelectric Materials Using Cast Iron Scrap Chips as a Source Material

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This study investigates the upgraded recycling of cast-iron scrap chips in synthesising Heusler alloys Fe<sub>2</sub>VAl for thermoelectric materials. It mainly examines the microstructure and the thermoelectric performance of the products. The thermoelectric performance showed positive results, as the maximum power factor, the *PF* value, of p-type 2C.I.-V-Al prepared using cast-iron scrap chips (the prefix 'C.I.' presumably stands for 'cast-iron scrap chips') was 1604 μWm<sup>-1</sup>K<sup>-2</sup> at 200°C, the highest *PF* value. Meanwhile, the undoped 2C.I.-V-Al prepared using cast-iron scrap chips showed an approximate two-fold improvement in the power factor value, with 967 μWm<sup>-1</sup>K<sup>-2</sup> at 200°C, a higher *PF* value than those previously reported. Unfortunately, in this study, the n-type 2C.I.-V-Al specimen made from cast-iron scrap chips could not be fabricated due to the effect of unavoidable impurities in the cast-iron scrap chips. The use of cast-iron scrap chips to produce undoped and p-type 2C.I.-V-Al alloys can contribute towards eco-friendly and cost-effective production processes.

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## 1. Introduction

The effective use of waste heat, where more than 60% of the original energy is released into the environment, is being sought against the current social background—for example, in concerns over future energy supply and emerging environmental issues. Thermoelectric generation, which maintains the conversion of efficiency independent from the energy scale and can convert thermal energy to electricity, is the solution.<sup>1)</sup> Thermoelectric materials have recently attracted renewed interest for their potential application in a waste heat recovery system without any harmful emissions. Thermoelectric materials consisting of abundant and cheap elements are required for the widespread use of devices for ecological issues in production processes. Thermoelectric properties are typically evaluated by the dimensionless figure of merit, *ZT*, expressed as follows:

$$ZT = \alpha^2 \sigma / k(T) \quad (1)$$

where  $\alpha$  is the Seebeck coefficient,  $\sigma$  is the electrical conductivity and  $k$  is the thermal conductivity. Thermoelectric materials need to improve their *ZT*.

In Fe<sub>3</sub>Al, with the ordered D0<sub>3</sub> structure, V can be replaced with Fe in a wide compositional range. The addition of V in Fe<sub>3</sub>Al stabilises the D0<sub>3</sub> structure and the existence of the intermetallic compound, and Fe<sub>2</sub>VAl has been suggested as one of the Heusler alloys. The Fe-V-Si system is known as one of the Heusler alloys, and its crystal structure is described as L2<sub>1</sub>, a higher-ordered phase than the D0<sub>3</sub> type. A Heusler alloy, Fe<sub>2</sub>VAl, is one of the promising candidates for thermoelectric semiconductors due to its rich resources, robust mechanical strength and high electrical response.<sup>2)</sup>

Additionally, Fe<sub>2</sub>VAl has thermoelectric power generation close to room temperature because of its high-power factor compared to conventional thermoelectric materials, such as Bi-Te and Pb-Te.<sup>3)</sup> Fe<sub>2</sub>VAl is a semimetal group with a narrow pseudogap at the Fermi level.<sup>4,5)</sup> Thus, small stoichiometric deviations can drastically change transport properties.<sup>6)</sup> Methods for controlling the pseudogap in Fe<sub>2</sub>VAl, reported by several researchers, include changing its stoichiometry in chemical composition<sup>7,8)</sup> and substituting the fourth element.<sup>9-16)</sup> These proposed methods increase the absolute values of the Seebeck coefficient and reduce the electrical resistivity of Fe<sub>2</sub>VAl. However, the thermal conductivity is comparatively greater, resulting in a small *ZT*. Therefore, it is essential to decrease the thermal conductivity for obtaining a higher performance for Fe<sub>2</sub>VAl. The arc-melting method, based on ingot metallurgy,<sup>6-9)</sup> mechanical alloying<sup>14-16)</sup> and a combination of them<sup>10)</sup> are the conventional fabrication processes of Fe<sub>2</sub>VAl. Commercially, the cost of pure Fe grains (purity: 99.99%; 3–5 μm) from a local company is approximately 12,000 yen for 500 g and 20,000 yen for 1 kg.<sup>7)</sup> In these conventional methods, metal powders with high purity, such as pure Fe, are required as raw materials to synthesise Fe<sub>2</sub>VAl, resulting in escalated costs for the final products.

Cast iron is essentially an iron-carbon alloy containing more than 2 mass% carbon, 1–3 mass% silicon, and minor elements such as manganese, sulphur and phosphorus, which markedly modify its structure and properties.<sup>17)</sup> It has been used in the oil and gas industry for making water conduit pipes, packer parts, boilers, valve bodies and valve parts. Cast-iron scrap chips are generated during mechanical processing. The recycling of cast-iron scrap chips is an interesting process because they can be used as base materials for preparing iron-based intermetallic compounds such as

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$\beta$ -FeSi<sub>2</sub> and Fe<sub>2</sub>VAl. We have already proposed the upgraded recycling of cast-iron scrap chips towards use in  $\beta$ -FeSi<sub>2</sub> thermoelectric materials.<sup>18–21</sup> Thus, this study promotes the upgraded recycling of cast-iron scrap chips towards preparing eco-friendly Fe<sub>2</sub>VAl thermoelectric materials. It evaluates the thermoelectric performance and the physical characteristics of Fe<sub>2</sub>VAl synthesised from cast-iron scrap chips. The purpose of the study is to reveal that the cast-iron scrap chips can be optimally used as a starting material for fabricating other iron-based materials, such as Fe<sub>2</sub>VAl, and promote thermoelectric performance comparable to that previously reported.

## 2. Experimental Procedure

The cast-iron scrap chips were prepared by drilling a cast-iron block in our laboratory. After collecting the cast-iron scrap chips in a beaker, they were cleaned using ethanol in an ultrasonic bath over four cycles of 20 min and then dried in a fume chamber (Yamato Scientific Co. Ltd., Japan). The specimens were then characterised using X-ray fluorescence (XRF) (Rigaku, Japan) and energy dispersion X-ray spectroscopy (EDXS) (EDAX Inc., USA) for elemental and chemical analysis, as mentioned in the previous paper. The numerical chemical compositions were cast iron (C.I.) or Fe:V:Al = 2:1:1 for undoped, cast iron (C.I.) or Fe:V:Ti:Al = 2:0.9:0.1:1 for p-type and cast iron (C.I.):V:Al:Si = 2:1:0.9:0.1 for n-type. The starting materials for undoped, p-type and n-type 2C.I.–V–Al were prepared by the solid-state reaction technique of cast-iron scrap chips (the prefix ‘C.I.’ presumably stands for ‘cast-iron scrap chips’; <100  $\mu$ m but easy to break during the ball-milling process), Fe grains (purity: 99.99%; 3–5  $\mu$ m), vanadium grains (purity: 99.99%; 75  $\mu$ m), powders of dopant element Ti (purity: 99%; 3  $\mu$ m) for p-type and powders of dopant element Si (purity: 99.99%; 3  $\mu$ m) for n-type. The powder mixture was prepared using a rotary dry ball milling for 4 h and then underwent a solid-state reaction at 1200°C for 12 h in a vacuum.

The solid-state reacted powder was prepared using a rotary dry ball milling for 1 d. Then, the reacted powder was directly put in a die and consolidated using a pulsed electric-current sintering technique at 950°C for 10 min in a vacuum under a uni-axial pressure of 80 MPa. Sintered specimens were annealed at 900°C for 2 d and 450°C for 6 h in a vacuum to homogenise the Heusler phase. The phase composition and morphology of specimens were investigated by a scanning electron microscope (SEM) and an electron probe micro-analyser (EPMA). The X-ray diffraction (XRD) analysis of specimens was then conducted for phase identification.

The thermoelectric properties of sintered Fe<sub>2</sub>VAl were evaluated by measuring the Seebeck coefficient,  $\alpha$ , electrical conductivity,  $\sigma$ , and thermal conductivity,  $k$ . The Seebeck coefficient and electrical conductivity were measured by a DC standard four-probe method. The thermal conductivity was calculated from the measured heat capacity and thermal diffusivity obtained by the laser flash method using the thermal constant analyser (LFA 457 Micro Flash) in a temperature range from room temperature to 500°C under the vacuum.

Table 1 Porosity data for the sintered Fe<sub>2</sub>VAl samples.

Numerical Chemical Composition	Archimedes Measurement	
	Density	Open porosity
2Fe-V-Al (Undoped)	6.34 g/cm <sup>3</sup>	0.1%
2C.I.-V-Al (Undoped)	6.14 g/cm <sup>3</sup>	0.1%
2Fe-0.9V-0.1Ti-Al (p-type)	6.23 g/cm <sup>3</sup>	0.4%
2C.I.-0.9V-0.1Ti-Al (p-type)	6.10 g/cm <sup>3</sup>	0.3%
2Fe-V-0.9Al-0.1Si (n-type)	6.20 g/cm <sup>3</sup>	0.4%
2C.I.-V-0.9Al-0.1Si (n-type)	6.09 g/cm <sup>3</sup>	0.2%

## 3. Experimental Results and Discussion

### 3.1 Characteristics of Fe<sub>2</sub>VAl prepared using cast-iron scrap chips

Table 1 shows the density and porosity data of sintered specimens for Fe<sub>2</sub>VAl prepared using pure Fe and cast-iron scrap chips. The values of open porosity for all samples were measured using Archimedes measurement. The open porosity observed for all samples was below 1% after sintering; thus, they were considered dense samples.

Figure 1 shows the XRD pattern of solid-state reacted powder and annealed samples of Fe<sub>2</sub>VAl prepared using pure Fe and cast-iron scrap chips. The prefix ‘C.I.’ presumably stands for ‘cast-iron scrap chips’, denoting the alloys formed from cast-iron scrap chips. The XRD pattern of solid-state reacted powder formed the D0<sub>3</sub>-type Fe<sub>3</sub>Al crystal structure. After the annealing treatment, the diffraction spectra in these alloys were identified in the expected L2<sub>1</sub> structure, while the Heusler-type appeared at a lower  $2\theta$  angle, such as (111) and (200). These results indicate that the Heusler structural Fe<sub>2</sub>VAl alloy was obtained. However, there were traces of an impurity phase or secondary phase, such as the oxides of Al<sub>2</sub>O<sub>3</sub>, V<sub>2</sub>O<sub>3</sub> and V<sub>2</sub>O<sub>5</sub>. The peaks of Fe<sub>3</sub>C or VC were obtained from the specimens made from cast-iron scrap chips, possibly due to the effect of impurity C, such as graphite, in the cast-iron scrap chips.

Figure 2 shows the SEM microstructures for annealed Fe<sub>2</sub>VAl alloys prepared using pure Fe and cast-iron scrap chips, respectively. The SEM images of annealed Fe<sub>2</sub>VAl prepared using cast-iron scrap chips revealed large intergranular segregation, which might be the V<sub>2</sub>O<sub>5</sub> and V<sub>2</sub>O<sub>3</sub> phases. On the other hand, several black particles of Al<sub>2</sub>O<sub>3</sub> were detectable in trace form in the Fe<sub>2</sub>VAl samples prepared using pure Fe and cast-iron scrap chips. The pore size observed from the SEM images was estimated to be less than 10  $\mu$ m. The open porosity as shown in Table 1 and observed in all samples was below 1% after sintering; thus, they were considered dense samples. To obtain perfectly homogenized Fe<sub>2</sub>VAl, a smaller sized V and a longer heat treatment are needed.<sup>22)</sup>

Figure 3 shows back-scattered electron (BSE) images and elemental mapping images of the undoped and p-type 2C.I.–V–Al prepared using cast-iron scrap chips. The presence of

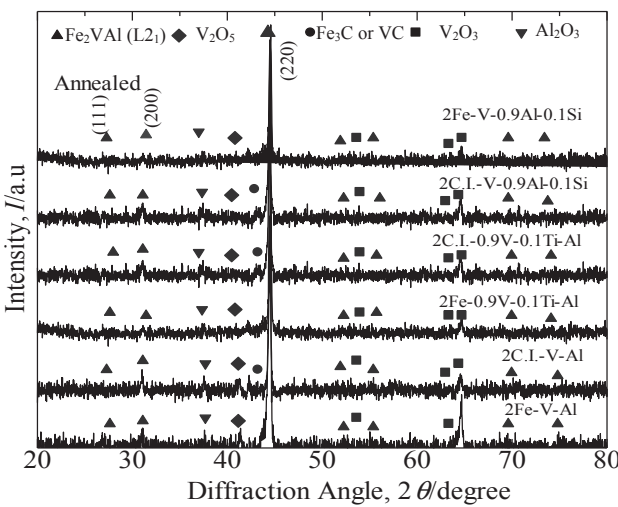
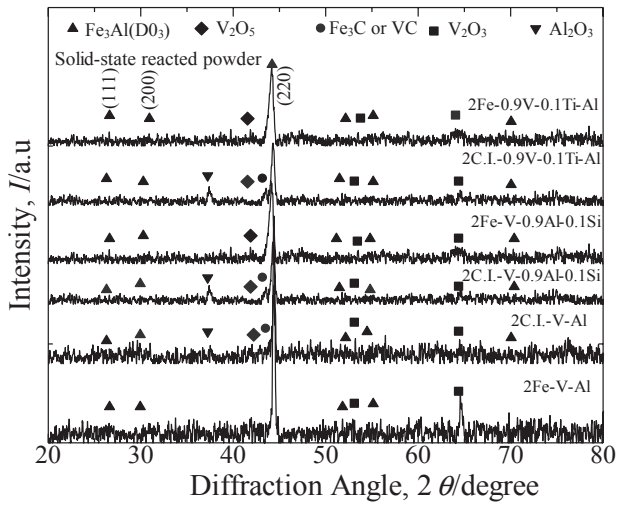


Fig. 1 XRD patterns of the solid-state reacted powder and annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

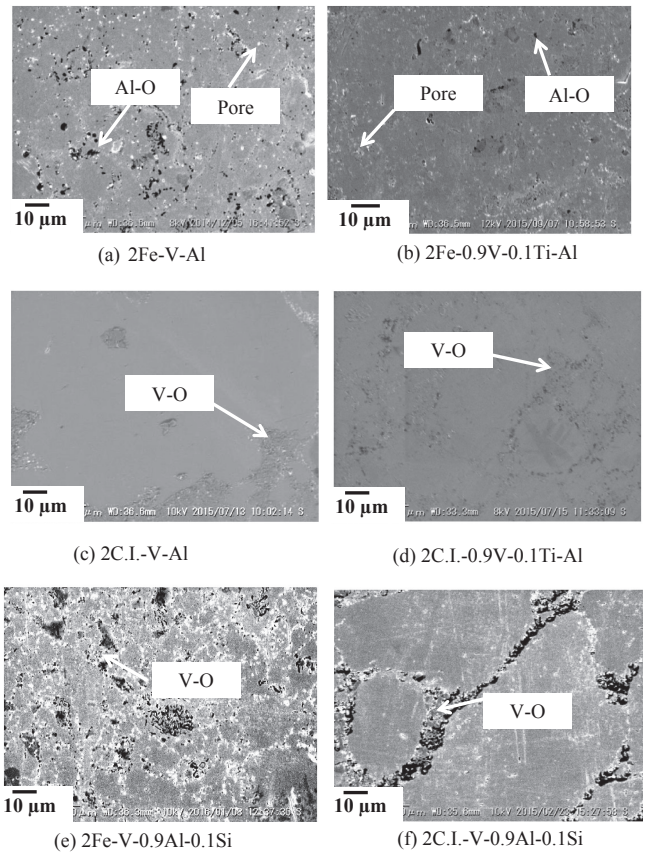


Fig. 2 SEM microstructures of the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

oxide in the matrix was considered to be caused by the ease with which the Al or V element is oxidised during sintering, as the cast-iron scrap chips are strongly oxidised by machining. This was most likely the oxidation of the

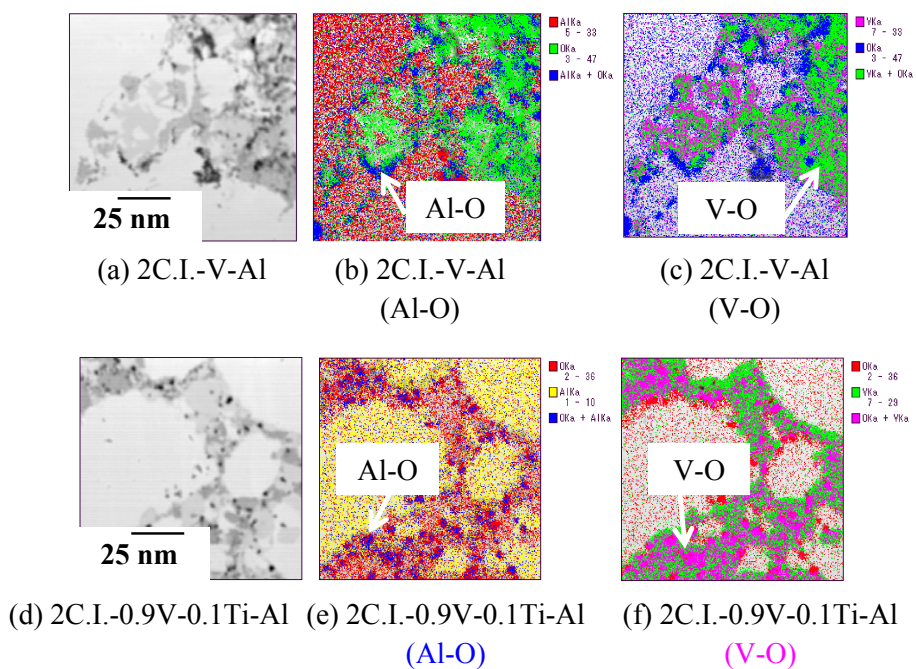


Fig. 3 EPMA analysed, elemental mapping images for O, V and Al of the annealed Fe<sub>2</sub>VAl samples prepared by utilizing cast iron scrap chips.



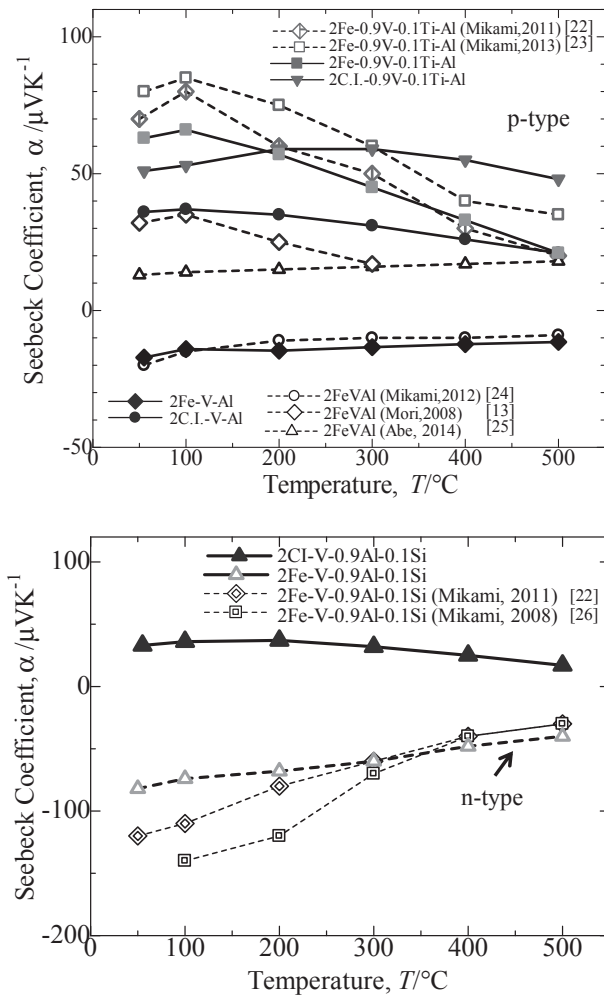


Fig. 4 Temperature dependence of Seebeck coefficient,  $\alpha$ , for the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

undoped and p-type 2C.I.-V-Al samples made from cast-iron scrap chips that were caused by impure oxygen in the cast-iron scraps.

### 3.2 Thermoelectric performances of Fe<sub>2</sub>VAl prepared using cast-iron scrap chips

Figure 4 shows the temperature dependence of the Seebeck coefficient,  $\alpha$ , of the annealed Fe<sub>2</sub>VAl specimens between room temperature and 500°C. The values of  $\alpha$  of the undoped and p-type 2C.I.-V-Al prepared using cast-iron scrap chips were positive in the entire temperature range in this study. These findings suggest that the dominant carriers were the hole-type in Fe<sub>2</sub>VAl, which is consistent with previous results.<sup>6,8)</sup> This is also in agreement with the band-structure calculations, which revealed the existence of large hole pockets near the Fermi level density of states (DOS) in Fe<sub>2</sub>VAl.<sup>4,5)</sup> On heating, intrinsic electrons and holes are excited. If the holes have slightly higher mobility than the electrons in these materials, the p-type carriers will eventually govern the thermal transport, leading the positive  $\alpha$  values to high temperatures. Also, the upward shift of  $E_F$  will cause higher activated energy for the hole carriers thermally excited across the pseudogap. The Seebeck coefficient values of undoped 2C.I.-V-Al prepared using cast-iron scrap chips are positive and centred approximately 20  $\mu\text{V/K}$ , as most carriers

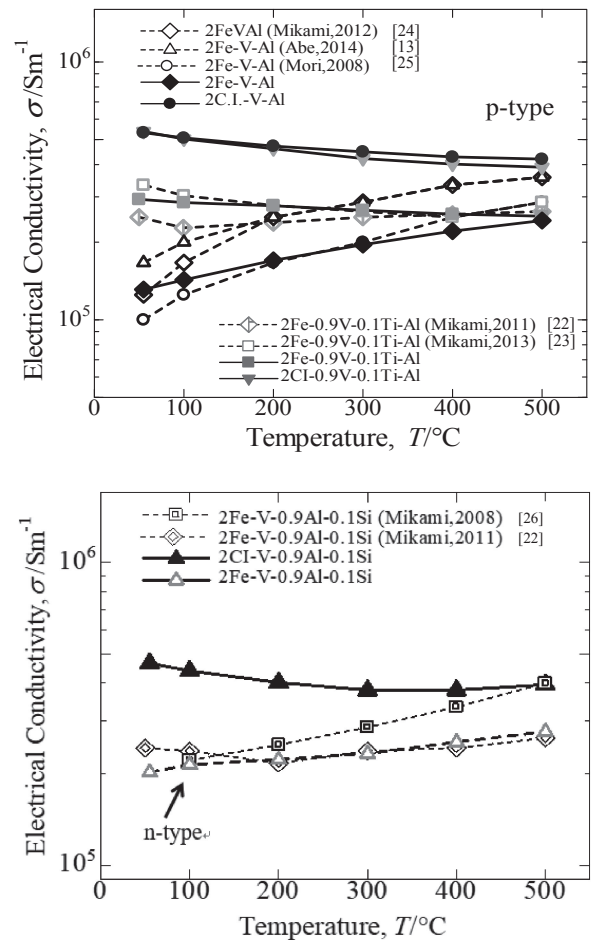


Fig. 5 Temperature dependence of electrical conductivity,  $\sigma$ , for the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

are holes; however, the number of carriers and hole pockets are nearly comparable.<sup>26)</sup> Furthermore, the Seebeck coefficient values of the p-type 2C.I.-V-Al prepared using cast-iron scrap chips were approximately 10% less than those previously reported.<sup>22,23)</sup> This reduction was attributed to the small difference in the off-stoichiometry of the specimen since the composition of cast-iron scrap chips contains impurities.

However, in this study, the n-type 2C.I.-V-Al specimen made from cast-iron scrap chips could not be fabricated due to the effect of impurities in the cast-iron scrap chips, such as O, C and Si. The effect of these impurities could change the conduction type of the 2C.I.-V-0.9Al-0.1Si specimen prepared using cast-iron scrap chips, from n-type to p-type. For example, Mn or C might be considered acceptor dopants by substituting the V site; however, Si is considered a donor dopant by substituting the Al site. Thus, further investigation is needed to study the effect of these impurities. For example, the n-type Fe<sub>2</sub>VAl sample made from pure Fe could be experimentally prepared and doped it with these impurities (C, Mn or Si). Thus, we could understand the effect of each impurity in the cast-iron scrap chips on the conduction type.

Figure 5 shows the temperature dependence of the electrical conductivity of the annealed Fe<sub>2</sub>VAl specimens between room temperature and 500°C. The electrical conductivity,  $\sigma$ , of the p-type 2C.I.-V-Al specimens prepared

using cast-iron scrap chips exhibits a rather semiconductor-like dependence on temperature. Furthermore, the electrical conductivity values of p-type 2C.I.-V-Al prepared using cast-iron scrap chips were approximately 30%<sup>13,24,25)</sup> higher than those previously reported.<sup>22,23)</sup> The temperature dependence of the electrical conductivity could be attributed to carrier mobility, which decreased with the temperature increase, since the acceptors were exhausted due to the movement of the carrier by lattice vibration. Thus, it is confirmed that the 2C.I.-V-Al specimens made from cast-iron scrap chips have positive effects on conductivity compared with those reported in other studies,<sup>13,22-25)</sup> where cast-iron scrap chips were also used as a starting material.

Nevertheless, in this study, the n-type 2C.I.-V-Al specimen prepared from cast-iron scrap chips would not have been possible to manufacture due to the effect of impurities in the cast-iron scrap chips, such as O, Si, Mn and C. These impurities might affect the conduction type of the 2C.I.-V-0.9Al-0.1Si specimen prepared using cast-iron scrap chips. Hence, further investigation regarding the chemical analysis of this sample is necessary to understand the effect of the impurities.

Figure 6 shows the thermal conductivity  $k$  of the annealed Fe<sub>2</sub>VAl specimens evaluated from room temperature to 500°C. The value of  $k$  for Fe<sub>2</sub>VAl is rather high, typically 28 W/mK,<sup>13)</sup> which is almost an n-type order of magnitude higher than that of conventional thermoelectric materials,<sup>22,23)</sup> such as Bi<sub>2</sub>Te<sub>3</sub>. However, the thermal conductivity for 2C.I.-V-Al specimens prepared using cast-iron scrap chips could be considered decreased, perhaps because of the off-stoichiometry effect. The reduction of the thermal conductivity of the 2C.I.-V-Al specimens prepared using cast-iron scrap chips is believed to be related to an increase in phonon scattering by the increase in grain boundary regions due to the dispersal of impurity phases and the small difference in stoichiometry in the composition.<sup>25)</sup> Thus, the thermal conductivity values of undoped and p-type 2C.I.-V-Al prepared using cast-iron scrap chips were about 10% smaller than those previously reported.<sup>22-25)</sup> Here, as well, cast-iron scrap chips were used as the base material.

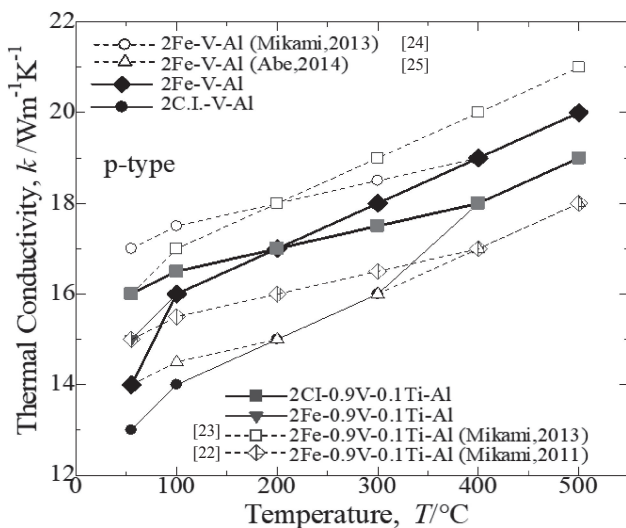


Fig. 6 Temperature dependence of thermal conductivity,  $k$ , for the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

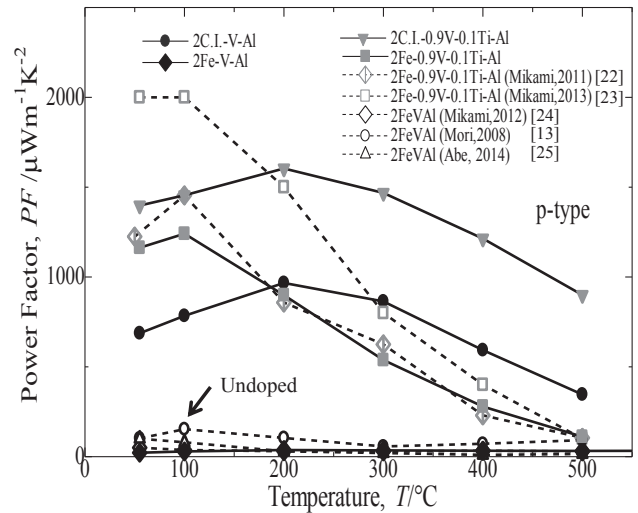


Fig. 7 Temperature dependence of power factor,  $PF$ , of the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

The power factor,  $PF$ , represents the electronic part of the thermoelectric performance, which is proportional to  $\alpha^2\sigma$ . Figure 7 shows a comparison of  $PF$  across Fe<sub>2</sub>VAl specimens, prepared using pure Fe and cast-iron scrap chips, as a function of temperature. The  $PF$  value of p-type 2C.I.-V-Al made from cast-iron scrap chips was comparable to those previously reported<sup>22,23)</sup> and have  $1604 \mu\text{Wm}^{-1}\text{K}^{-2}$  at 200°C, the highest  $PF$  value at a certain temperature. Here, too, cast-iron scrap chips were used as the starting material. Additionally, the  $PF$  values of undoped 2C.I.-V-Al prepared using cast-iron scrap chips showed an approximate two-fold improvement ( $967 \mu\text{Wm}^{-1}\text{K}^{-2}$  at 200°C, the highest  $PF$  value at a certain temperature) compared to those previously reported.<sup>24,25)</sup> It is interesting to note that the performance of undoped and p-type 2C.I.-V-Al made from cast-iron scrap chips is comparable to that of Fe<sub>2</sub>VAl prepared using pure Fe, especially at temperatures higher than 200°C, which has been reported in the literature. Unfortunately, in this study, the n-type 2C.I.-V-Al specimen made from cast-iron scrap chips could not be fabricated due to the effect of impurities in the cast-iron scrap chips, such as O, Si, Mn and C.

Thermoelectric performance is often discussed along with the figure of merit,  $Z = \alpha^2\sigma/k$ , requiring not only an increase in the power factor,  $PF$ , but also a decrease in thermal conductivity,  $k$ . Figure 8 shows the variations in the dimensionless figure of merit,  $ZT$ , with the measuring temperature of the annealed Fe<sub>2</sub>VAl specimens prepared using pure Fe and cast-iron scrap chips. As a result, the  $ZT$  (dimensionless figure of merit) values of undoped 2C.I.-V-Al prepared using cast-iron scrap chips showed an approximate two-fold improvement, while those of the p-type 2C.I.-V-Al prepared using cast-iron scrap chips were approximately 10% smaller than those previously reported.<sup>22-25)</sup> Thus, undoped and p-type 2C.I.-V-Al made from cast-iron scrap chips showed positive results in thermoelectric performance, where, again, cast-iron scrap chips were used as the starting material.

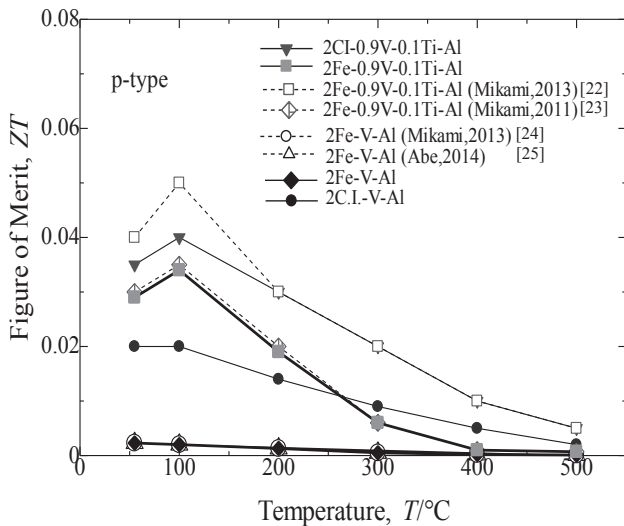


Fig. 8 Variation in the dimensional figure of merit,  $ZT$ , with the measuring temperature of the annealed Fe<sub>2</sub>VAl samples at 900°C for 2 days and 450°C for 6 hours.

#### 4. Conclusion

The upgraded recycling of cast-iron scrap chips was discussed as an eco-friendly and cost-effective production process to fabricate Fe<sub>2</sub>VAl thermoelectric materials. In this study, the undoped and p-type 2C.I.-V-Al specimens prepared using cast-iron scrap chips were successfully fabricated, even though they used cast-iron scrap chips as the starting material. The maximum power factor, the  $PF$  value of p-type 2C.I.-V-Al, made from cast-iron scrap chips, was comparable to values previously reported,<sup>22,23</sup> with  $1604 \mu\text{Wm}^{-1}\text{K}^{-2}$  at 200°C, the highest  $PF$  value at a particular temperature. Meanwhile, the undoped 2C.I.-V-Al prepared using cast-iron scrap chips showed an approximate two-fold improvement in the power factor value, with  $967 \mu\text{Wm}^{-1}\text{K}^{-2}$  at 200°C, the highest  $PF$  value at a particular temperature compared with those previously reported.<sup>24–26</sup> Undoped and p-type 2C.I.-V-Al fabricated from cast-iron scrap chips delivered positive results in thermoelectric performance where cast-iron scrap chips were used as the starting material. Nevertheless, in this study, the n-type 2C.I.-V-Al specimen made from cast-iron scrap chips could not be obtained because impurities present in cast-iron scrap chips, such as O, Mn, C and Si, may affect the conduction of the 2C.I.-V-0.9Al-0.1Si specimen prepared using cast-iron scrap chips, from n-type to p-type. Hence, further investigation on the effect of impurities in cast-iron scrap chips for the development of an n-type 2C.I.-V-Al alloy is

necessary, as are additional improvements in thermoelectric performance. In this regard, a suggestion would be to experimentally prepare the n-type Fe<sub>2</sub>VAl sample made from pure Fe and dope it with C, Mn or Si. Thus, we could understand the effects of each impurity on the conduction type.

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