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Thermoelectric properties of UB_4 from 300 to 850 K

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Abstract

Seebeck coefficient and the electrical conductivity for polycrystalline U^nB ($^nB_4$: natural boron) were measured in the temperature range from 300 to 850 K. Seebeck coefficient increased with temperature from 8 μVK^{-1} at 300 K to 40 μVK^{-1} at 850 K, and the electrical conductivity decreased with increasing temperature from 2.7×10^3 Scm^{-1} at 300 K to 2.3×10^3 Scm^{-1} at 850 K. The temperature dependence of the electrical conductivity suggested that UB_4 had semimetallic character. The power factor for U^nB_4 showed the value of 3.4×10^{-4} Wm^{-1} K^{-2} at 850 K. The power factor for isotopically enriched $U^{11}B_4$ showed the value of 7.2×10^{-4} Wm^{-1} K^{-2} at 850 K. It was suggested that the power factor for UB_4 was enhanced by the increase in carrier concentration with impurities.

Keywords: D. Electrical conductivity

1. Introduction

In the nuclear fuel cycle, a large amount of depleted uranium (99.8% ^{238}U) is discharged. The depleted uranium is not adequate for the re-use as nuclear fuel except the blanket in fast-breeder reactor. Therefore, the utilization of the depleted uranium is important to complete the nuclear fuel cycle.

The thermoelectric material directly converts heat into electricity, and vice versa. The thermoelectric figure of merit (Z), $Z=(\alpha^2\cdot\sigma/\kappa)$ where α is Seebeck coefficient, σ is electrical conductivity and κ is thermal conductivity, is proportional to $m^{2/3}(\mu_c/\kappa_{ph})$ [1], where m is the effective mass of the charge carrier, μ_c is the charge carrier mobility and κ_{ph} is the thermal conductivity by phonons. In general, materials having large average atomic number tend to exhibit large μ_c/κ_{ph}, and f-electrons lead to large effective mass of the charge carrier. Since uranium has the large atomic number (92) and 5f-electrons, the thermoelectric properties of materials including U are of interest. Furthermore, κ_{ph} is able to controlled by isotopic composition.

We have investigated thermoelectric properties of URu_2Si_2, $U_2Ru_3Si_5$ [2] and UB_4 [3]. The power factor for URu_2Si_2 and $U_2Ru_3Si_5$ showed the maximum value of 1.2×10^{-4}, 5.4×10^{-5} Wm^{-1} K^{-2} at 1100 K, respectively. The thermoelectric properties for UB_4 were only measured in the temperature range from 80 to 300 K. Power factor and Z increased with temperature and showed the maximum value of 1.3×10^{-5} Wm^{-1} K^{-2} and 4.2×10^{-3} K^{-1} at 300 K, respectively. Many compounds including boron such as β-boron [4], boron carbide [5–7] and α-AlB_{12} [4] indicate high thermoelectric properties at high temperature. Therefore, UB_4 is expected to exhibit high thermoelectric properties at high temperature. The thermoelectric properties for UB_4, however, were not reported except our previous study at low temperatures [3].

In this study, Seebeck coefficient and the electrical conductivity for polycrystalline U^nB_4 (^nB: natural boron) were measured in the temperature range from 300 to 850 K to investigate the thermoelectric properties for UB_4. The thermoelectric properties for $U^{10}B_4$ and $U^{11}B_4$, which were prepared from ^{10}B or ^{11}B enriched boron powder and depleted uranium were also reported to confirm the isotope effect for electrical properties.

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2. Experimental

Each specimen was prepared by the arc-melt method in an argon atmosphere using ^{n}B powder (natural boron, Koch Chemicals Ltd), isotopically enriched ^{10}B and ^{11}B powders (TRACE Co., Ltd) and depleted-U metal (Power Reactor and Nuclear Fuel Development Corp.). The isotopic compositions for boron powders and U metal are shown in Table 1. A presence of a single phase for each sample was confirmed by X-ray diffraction analysis.

Seebeck coefficient and the electrical conductivity were measured in vacuum in the temperature range from 300 to 850 K. Seebeck coefficient was calculated within $\pm 10\%$ errors from the difference of temperature and electromotive force between the both ends of sample. The electrical conductivity was measured by the direct current four-probe method.

3. Results and discussion

UB_4 crystallizes in the ThB_4-type structure (space group *P4/mbm*) [8–10]. The lattice parameters of $U^{n}B_4$, $U^{10}B_4$ and $U^{11}B_4$ are shown in Table 1 together with a literature value of $U^{n}B_4$ [10]. From the isotopic composition dependence of the lattice parameters for Ge [11] and C [12], the lattice parameters are considered to depend on the isotopic composition as a consequence of the zero-point motion. The differences in the lattice parameters among three samples, however, could not be detected.

The measured electrical conductivity (σ) is plotted as a function of temperature in Fig. 1 together with the electrical conductivity for $U^{n}B_4$ below 300 K. The electrical conductivity for $U^{n}B_4$ decreased with increasing temperature from 2.7×10^{3} Scm^{-1} at 300 K to 2.3×10^{3} Scm^{-1} at 850 K. The electrical conductivity of $U^{n}B_4$ at 300 K obtained in this study is in good agreement with that obtained in our previous study [3]. Although, the electrical conductivity of metals decreases inversely proportional to the temperature, the decreasing rate of the electrical conductivity for UB_4 was 40% of that for metals. Moreover, the magnitude of the electrical conductivity is smaller than that for metals. These behaviors suggest that UB_4 is semimetal.

As is seen in Fig. 1, the electrical conductivities for $U^{10}B_4$ and $U^{11}B_4$ were larger than that for $U^{n}B_4$. According to our previous study [13], the thermal conductivity for isotopically enriched boron carbide ($^{10}B_4^{12}C$, $^{11}B_4^{12}C$, $^{10}B_4^{13}C$ and $^{11}B_4^{12}C$) was smaller than that for natural boron carbide $^{n}B_4^{n}C$. In general, the thermal conductivity of isotopically enriched materials are enhanced by the reduction of the probability of the phonon-isotope scattering. The reduction of the thermal conductivity for isotopically enriched boron carbide indicated that the content of impurities in isotopically enriched powders was larger than that in natural powder. Therefore, it was thought that the electrical conductivity for $U^{10}B_4$ and $U^{11}B_4$, which were made from the same isotopically enriched boron powder as in the previous study [13], were larger than that for $U^{n}B_4$ due to the impurities.

Seebeck coefficient (α) is plotted as a function of temperature in Fig. 2 together with that for $U^{n}B_4$ below 300 K [3]. Seebeck coefficient for $U^{n}B_4$ was positive and increased with temperature from 8 μVK^{-1} at 300 K to

Table 1
The lattice parameters and isotope abundances

Sample	Lattice parameter		Isotopic composition	
	a (nm)	*c* (nm)	^{10}B (at.%)	^{11}B (at.%)
$U^{n}B_4$	0.70771(8)	0.39813(6)	19.8	80.2
$U^{10}B_4$	0.70769(8)	0.39810(6)	99.75	0.25
$U^{11}B_4$	0.70778(10)	0.39819(9)	0.48	99.52
$U^{n}B_4$ [10]	0.70773(2)	0.39816(2)	19.8	80.2

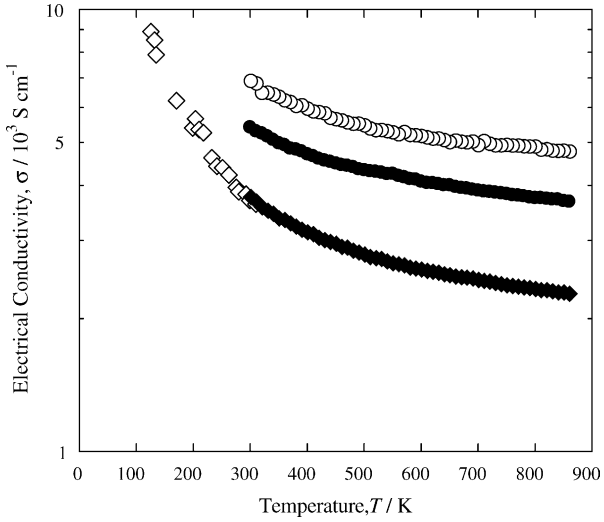

Fig. 1. Temperature dependence of the electrical conductivity for UB_4. (♦) $U^{n}B_4$, (●) $U^{10}B_4$, (○) $U^{11}B_4$, (◇) $U^{n}B_4$ [3].

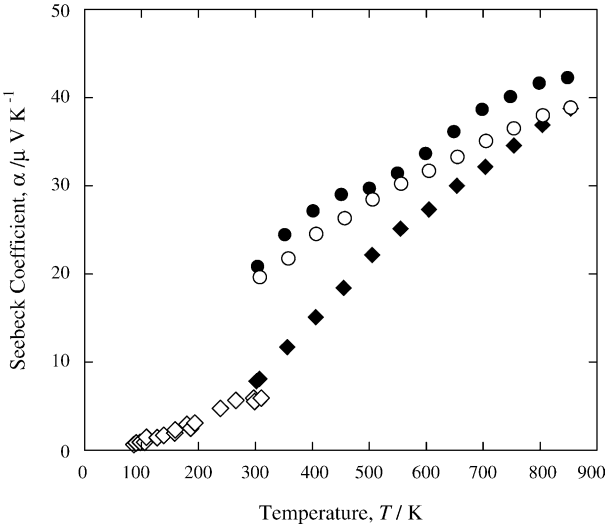

Fig. 2. Temperature dependence of Seebeck coefficient for UB_4. (♦) $U^{n}B_4$, (●) $U^{10}B_4$, (○) $U^{11}B_4$, (◇) $U^{n}B_4$ [3].

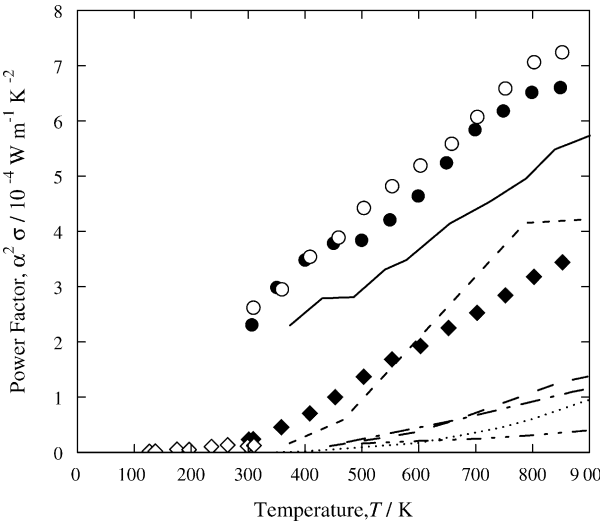

Fig. 3. Temperature dependence of the power factor for UB_4. (♦) U^nB_4, (●) $U^{10}B_4$, (○) $U^{11}B_4$, (◇) U^nB_4 [3]. (—) U [14,15], (– – –) B_4C [5], (-·-·-) B_4C [6]. (-··-) B_4C [7], (- - - -) β-boron [4], (·····) α-AlB_{12} [4].

40 μVK^{-1} at 850 K. The Seebeck coefficient of U^nB_4 at 300 K obtained in this study is also in good agreement with that obtained in our previous study [3]. The magnitude of Seebeck coefficient was larger than that for metals, which corresponded to semimetallic behavior of the electrical conductivity. Seebeck coefficients for $U^{10}B_4$ and $U^{11}B_4$ were larger than that for U^nB_4 below 600 K, and the deference tended to be small with increasing temperature above 600 K.

The power factor ($\alpha^2\cdot\sigma$), which is the electrical output of the thermoelectric devices, is shown in Fig. 3. The power factor for UB_4 increased monotonically with temperature and showed the value of 3.4×10^{-4} Wm^{-1} K^{-2} for U^nB_4, 6.6×10^{-4} Wm^{-1} K^{-2} for $U^{10}B_4$ and 7.2×10^{-4} Wm^{-1} K^{-2} for $U^{11}B_4$ at 850 K. Although the power factor for U^nB_4 was smaller than that of β-boron [4] and U metal [14,15], the power factor for $U^{10}B_4$ and $U^{11}B_4$ up to 850 K was larger than that for U metal [14,15], β-boron [4], boron carbide [5–7] and α-AlB_{12} [4]. These relatively high power factors for $U^{10}B_4$ and $U^{11}B_4$ are ascribed to large carrier concentrations introduced by non-isotopic impurities.

4. Conclusion

The electrical conductivity decreased with increasing temperature and Seebeck coefficient was larger than that for metals, which suggested that UB_4 indicated semimetallic behavior.

The power factor for U^nB_4 showed the maximum value of 3.4×10^{-4} Wm^{-1} K^{-2} at 850 K. The power factor of $U^{10}B_4$ and $U^{11}B_4$ were larger than that for U^nB_4, indicating that the power factor was enhanced by the impurities. The power factor of $U^{11}B_4$ showed the value of 7.2×10^{-4} Wm^{-1} K^{-2} at 850 K which was close to a standard value for practical use, $\alpha^2\cdot\sigma=1\times10^{-3}$ Wm^{-1} K^{-2}. Therefore, UB_4 is a promising thermoelectric material. Moreover, improvement of the power factor is expected by the control of the charge carrier density.

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