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Boron isotope effects on the thermoelectric properties of UB_4 at low temperatures

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Abstract

The thermoelectric power and electrical resistivity of UB_4 with three different boron isotope enrichments were measured in the temperature range from 80 to 300 K. The thermoelectric power of each sample decreased with decreasing temperature, but a slight saturation of the decrease due to phonon-drag effect was observed below 120 K for both boron isotope enriched samples. The resistivity decreased with decreasing temperature and there existed no large difference among the resistivities of natural boron, boron-10 enriched and boron-11 enriched samples. The figure of merit for thermoelectricity of UB_4 was lower by three orders of magnitude than that of well-known thermoelectric material $(Bi, Sb)_2(Te, Se)_3$. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

In the nuclear fuel cycle a large amount of depleted uranium is discharged. The depleted uranium (99.8% U-238) cannot be used for nuclear fuel. The utilization of the depleted uranium is important in the nuclear engineering. Since uranium has 5f-electrons and the largest atomic number of all naturally occurring elements, uranium compounds will exhibit interesting properties. We have been investigating new possibilities of some uranium compounds such as URu_2Si_2 and $U_2Ru_3Si_5$ as thermoelectric materials [1]. The compound of UB_4 is expected to be a novel thermoelectric material as follows. In general, materials having large average atomic number are thought to have enhanced μ_c/κ_{ph} (μ_c: charge carrier mobility, κ_{ph}: thermal conductivity due to phonons) [2] as well as a large thermoelectric figure of merit Z. The thermoelectric figure of merit can be written as

$$Z = S^2/\rho\kappa, \qquad (1)$$

where S represents the thermoelectric power, ρ is the electrical resistivity and κ is the thermal conductivity. Furthermore the layer structure of UB_4 [3] probably causes the decrease in the thermal conductivity. Due to the presence of 5f-electrons, some uranium compounds are known as heavy fermion compounds in which charge carrier has a large effective mass at low-temperature. The large effective mass can enhance the optimum thermoelectric figure of merit and lead to particularly large values of the thermoelectric power [4]. Although UB_4 is not a heavy fermion compound, the effective mass of its electrons is expected to be larger than that of regular alloys [5]. It was observed that the Seebeck coefficient increases with decreasing impurity content [6]. An isotope with different mass can be regarded as an impurity. In this study, the thermoelectric properties of three kinds of UB_4 samples, which consist of boron-10, boron-11 and natural boron, were measured to clarify the applicability of this material for use as a thermoelectric material.

2. Experimental

The starting materials, 99.999% pure natural B powder and 99.98% pure U metal, were mixed and arc-melted together several times in purified argon

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Table 1
The lattice parameters of samples

Sample	*a* (nm)	*c* (nm)
$U''B_4$	0.70771(8)	0.39813(6)
$U^{10}B_4$	0.70769(8)	0.39810(6)
$U^{11}B_4$	0.70778(10)	0.39819(9)
$U''B_4$ [3]	0.70773(2)	0.39816(2)

atmosphere. Isotopic compositions used in this study were 99.75% boron-10, 99.52% boron-11 and natural boron. The arc-melted samples were shaped into cylindrical rods of about 4 mm in diameter and 20 mm in length. X-ray diffraction (XRD) indicated the presence of a single phase for each sample. The lattice parameters are shown in Table 1 comparing with the other data [3]. Thermoelectric power, S, and electrical resistivity, ρ, were measured from 80 to 300 K in vacuum by using the standard 4-wire method [7].

3. Results and discussion

The temperature dependences of thermoelectric power for the three UB_4 samples are shown in Fig. 1. The values of thermoelectric power decrease with decreasing temperature. From the slopes of the temperature dependence of thermoelectric power ($\Delta\alpha$), the Fermi energy (E_F) was deduced using Eq. (2) for each sample.

$$E_F = L/\Delta\alpha, \tag{2}$$

where L is the Lorenz number [8]. The Fermi energy of each UB_4 sample was almost the same −1.04 eV. The carrier electron concentration obtained from the Fermi energy was 4.82×10^{27} m^{-3} and indicated metallic property. Below 120 K, the thermoelectric power for boron-isotope enriched samples seems to be approximately constant, suggesting the presence of phonon-drag thermoelectric power [9,10] caused by the strong interaction between phonons and electrons. In the case of UB_4 sample prepared from natural B, the slope change due to phonon-drag was not clearly observed in Fig. 1. It has been observed for some materials that the phonon-drag thermoelectric power decreases with increasing impurity content, e.g., the phonon-drag effect of Au doped with 2.2 mol% Au is one-third that of pure Au [6]. Natural boron can be considered as boron-11 doped with 20 mol% boron-10. Therefore the magnitude of the phonon-drag thermoelectric power was small and was not clearly observed.

The electrical resistivities of UB_4 are shown in Fig. 2 as well as those reported by Matterson and Jones [11] for UB_4 and Wang and Lee [12] for U metal. The difference in the electrical resistivity among three UB_4 samples seemed to be small. The resistivity of UB_4 at room temperature in this study was about an order of magnitude larger than that by Matterson and Jones [11]. The small temperature dependence of electrical resistivity also indicated that these samples were metallic.

The thermoelectric figures of merit Z of UB_4 expressed by Eq. (1) were calculated from the thermoelectric power, electrical resistivity and thermal conductivity, which was calculated by the Wiedemann–Franz relation, in this study and the results are shown in Fig. 3. The thermoelectric figures of merit of three UB_4 samples are lower by an order of magnitude than that of

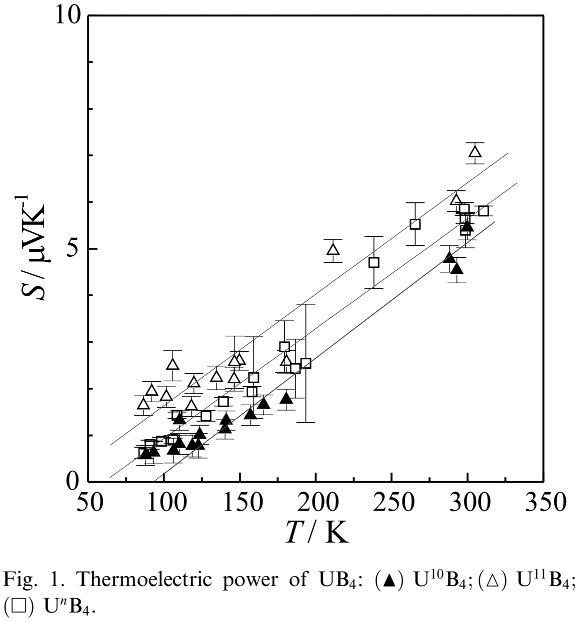

Fig. 1. Thermoelectric power of UB_4: (▲) $U^{10}B_4$; (△) $U^{11}B_4$; (□) $U''B_4$.

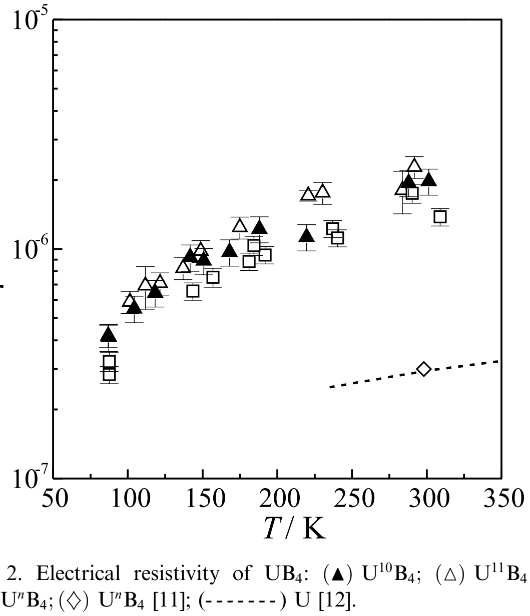

Fig. 2. Electrical resistivity of UB_4: (▲) $U^{10}B_4$; (△) $U^{11}B_4$; (□) $U''B_4$; (◇) $U''B_4$ [11]; (-------) U [12].

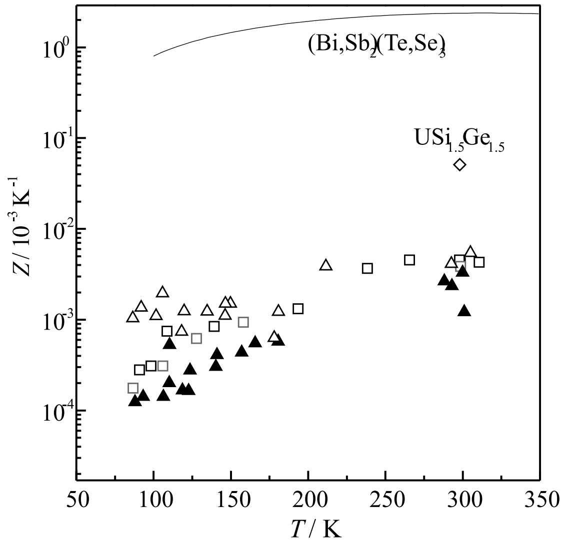

Fig. 3. Thermoelectric figure of merit of UB$_4$: (▲) U^{10}B$_4$; (△) U^{11}B$_4$; (□) U^{n}B$_4$; (◇) USi$_{1.5}$Ge$_{1.5}$ [13]; (——) (Bi, Sb)$_2$ (Te, Se)$_3$ [14].

other uranium compound, e.g., USi$_{1.5}$Ge$_{1.5}$ [13] and three orders of magnitude lower than those of well-known thermoelectric materials (Bi, Sb)$_2$(Te, Se)$_3$ [14]. These poor thermoelectric properties may come from the metallic properties such as the small thermoelectric power and the large thermal conductivity.

4. Conclusion

The thermoelectric powers of three samples of UB$_4$ with different isotopic compositions are all about 4–6 μV/K at room temperature and decreased with decreasing temperature. Both boron-isotope-enriched samples showed a small phonon-drag thermoelectric power below 120 K. It is concluded that uranium tetraboride is a poor thermoelectric material due to metallic properties such as low thermoelectric power and large electrical resistivity.

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References

[1] Y. Arita, K. Terao, T. Matsui, Proc. Int. Conf. Thermoelectrics 14 (1998) 540.
[2] G.A. Slack, Solid State Phys. 34 (1979) 1.
[3] R. Chipaux, G. Ceilia, M. Beauvy, R. Troc, J. Less-Common Met. 121 (1986) 347.
[4] B. Louie, R. Radebaugh, in: Proceedings of First National Thermoelectric Cooler Conference, 1992, p. 140.
[5] A. Menovsky, J.J.M. Franse, J.C.P. Klaasse, J. Cryst. Growth 70 (1984) 519.
[6] R.S. Crisp, J. Rungis, Philos. Mag. 22 (1970) 217.
[7] R.J. Buist, in: D.M. Rowe (Ed.), CRC Handbook of Thermoelectrics, CRC, London, 1995, p. 189.
[8] C.M. Bhandari, in: D.M. Rowe (Ed.), CRC Handbook of Thermoelectrics, CRC, London, 1995, p. 27.
[9] J. Gurevich, Phys. USSR 9 (1945) 447.
[10] M. Bailyn, Philos. Mag. 5 (1960) 1059.
[11] K.J. Matterson, H. Jones, Trans. Br. Ceram. Soc. 60 (1961) 475.
[12] G.C.Y. Wang, C.V. Lee, in: Y.S. Touloukian (Ed.), Thermophysical Properties of High Temperature Solid Materials 1, Macmillan, New York, 1967, p. 1049.
[13] L.H. Brixner, J. Inorg. Nucl. Chem. 25 (1963) 783.
[14] H. Submann, K. Stecher, W. Eichler, S. Langhammer, N.H. Yuen, N.W. Sung, N.W. Voung, S.S. Hoau, V.M. Trishenikov, M.B. Cherbina-Samoilova, Sault-6-Sojus, Izd. Nauka, Moscow, 1985, p. 37.