ELSEVIER

Journal of Nuclear Materials 294 (2001) 206–208

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Thermoelectric properties of URu_2Si_2 and $U_2Ru_3Si_5$

Yuji Arita *, Kenji Terao, Satoshi Mitsuda, Yoshimasa Nishi, Tsuneo Matsui, Takanori Nagasaki

Department of Quantum Engineering, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

Abstract

The new possibility of uranium noble metal silicides for thermoelectric materials was investigated by the analogy with transition-metal silicides having good thermoelectric power. The Seebeck coefficient and electrical resistivity of URu_2Si_2 and $U_2Ru_3Si_5$ were measured in the temperature range from 300 to 1100 K and Hall coefficient at room temperature. The Seebeck coefficients of URu_2Si_2 and $U_2Ru_3Si_5$ were obtained to be −48.9 and −32.8 μV K^{-1} at 1100 K, respectively. The temperature dependences of the electrical resistivities for URu_2Si_2 and $U_2Ru_3Si_5$ were small, which are similar to that of uranium metal rather than that of silicon semiconductor. As thermoelectric materials for high temperatures, URu_2Si_2 and $U_2Ru_3Si_5$ were thought to be not promising.

1. Introduction

In the nuclear fuel cycle a large amount of depleted uranium and some fission-produced noble metals, such as Ru, Pd and Rh, are discharged. The noble metals are valuable and important in many areas. The number of nuclear power plants is large, and consequently the amount of fission-produced noble metals is large and becomes comparable to that of national resources of the world. Although some of these reactor-produced noble metals have radioactivity, they will become non-radioactive after about 50 years. The depleted uranium cannot be used for nuclear fuel, but uranium is an interesting element with 5f-electrons and the largest atomic number of the naturally occurring elements. In general, compounds with large average atomic number are thought to have enhanced μ_c/κ_{ph} (μ_c: charge carrier mobility, κ_{ph}: thermal conductivity due to phonon) [1] as well as high dimensionless thermoelectric figures of merit ZT ($ZT = S^2T/\rho\kappa$, S: Seebeck coefficient, T: absolute temperature, ρ: electrical resistivity, κ: thermal conductivity). Average atomic numbers of URu_2Si_2 and $U_2Ru_3Si_5$ are 99.27 and 91.97, respectively. From the presence 5f-electrons, some uranium compounds are known as the heavy fermion compounds which have a large effective mass of electron or hole at low temperature. The large effective mass of charge carriers can also give large Seebeck coefficient values [2] and thus large optimum thermoelectric figures of merit. The effective masses of charge carriers in URu_2Si_2 and $U_2Ru_3Si_5$ are 25–140 times larger than those in non-heavy fermion compounds [3–5]. The Seebeck coefficients and electrical resistivities of URu_2Si_2 and $U_2Ru_3Si_5$ have been only measured at low temperatures [5–10]. The compounds URu_2Si_2 and $U_2Ru_3Si_5$ are thought to be similar to the transition-metal silicides which have high melting points and good electric properties. In this study, the thermoelectric properties of URu_2Si_2 and $U_2Ru_3Si_5$ were measured to clarify their applicability for use as thermoelectric materials.

2. Experimental

The starting materials, Si powder (purity: 99.999%) and Ru powder (purity: 99.9%), were mixed in the desired molar ratio and cold-pressed into a pellet at

* Corresponding author. Tel.: +81-52 789 5310; fax: +81-52 789 4399.

E-mail address: y-arita@nucl.nagoya-u.ac.jp (Y. Arita).

0022-3115/01/$ - see front matter
PII: S0022-3115(01)00476-7

400 kg cm^{-2}. This pellet and a uranium piece (purity: 99.98%) were arc-melted together several times in purified argon atmosphere, and then annealed in an evacuated quartz tube at 1173 K for one week (URu_2Si_2) and at 1073 K for two weeks ($U_2Ru_3Si_5$). The arc-melted samples were shaped into a cylindrical rod of about 5 mm in diameter and 25 mm in length. X-ray diffraction (XRD) indicated the presence of a single phase for each sample. The Seebeck coefficient, S, and the electrical resistivity, ρ, were measured in the range from 300 to 1100 K at a pressure of 10^{-4} Pa of air. A Pt/Rh–Pt thermocouple was attached on each end of the sample rod, and another two Pt wire electrodes were placed between them in the standard 4-wire arrangement. The Hall effects were measured at room temperature in a magnetic field up to 3 T.

3. Results and discussion

The temperature dependences of the Seebeck coefficients for URu_2Si_2 and $U_2Ru_3Si_5$ are shown in Fig. 1, indicating that both samples are n-type semiconductors. The values of the Seebeck coefficients of URu_2Si_2 and $U_2Ru_3Si_5$ increase with temperature and reach −48.9 and −32.8 V K^{-1} at 1100 K, respectively. The Seebeck coefficient of URu_2Si_2 is larger than that of $U_2Ru_3Si_5$ at all temperatures. The Fermi energies of URu_2Si_2 and $U_2Ru_3Si_5$ were calculated from the slope of the temperature dependence of the Seebeck coefficient. The Fermi energy of URu_2Si_2 thus obtained was 0.79 eV and at least 10 times larger than that obtained previously for URu_2Si_2 at low temperatures [11]. The measured Fermi energy of $U_2Ru_3Si_5$ was 0.71 eV. These large Fermi energies show that the effective mass is not large. Since the large effective mass is caused by an enormous electronic density of states at the Fermi level due to strong electronic interactions, the enhancement of the electronic interactions in URu_2Si_2 and $U_2Ru_3Si_5$ seems not to appear from the present result. The electrical resistivities of URu_2Si_2 and $U_2Ru_3Si_5$ are shown in Fig. 2 together with those of the Si and U [12]. The electrical resistivities of URu_2Si_2 and $U_2Ru_3Si_5$ have almost the same value, about 2.0×10^{-5} Ω m regardless of temperatures. This small temperature dependence is similar to that of uranium rather than silicon semiconductor. The small temperature dependence of the electrical resistivity and the low Seebeck coefficient of URu_2Si_2 and $U_2Ru_3Si_5$ indicate that these samples are metallic. The thermal conductivity κ was calculated by the following Wiedemann–Franz relation:

$$\kappa = LT/\rho, \quad (1)$$

where L is the Lorenz number.

The values of the dimensionless thermoelectric figure of merit, ZT of URu_2Si_2 and $U_2Ru_3Si_5$ were 0.098 and 0.043 at 1100 K, respectively, based on the Seebeck coefficient S in Fig. 1, electrical resistivity in Fig. 2 and thermal conductivity calculated from Eq. (1). The values of ZT for URu_2Si_2 and $U_2Ru_3Si_5$ are shown in Fig. 3 as a function of temperature, and these are small compared to other thermoelectric materials.

From the measurement of the Hall coefficient by Onuki et al. [8], the carrier concentration of URu_2Si_2 at room temperature was estimated to be 1×10^{28} m^{-3}. The Hall mobility μ of URu_2Si_2 was also calculated to be 1.9×10^{-5} m^2 V^{-1} s^{-1} from the present electrical resistivity data and the relation $\mu = R_H/\rho$ [8], where R_H is the Hall coefficient. The Hall coefficient and the carrier

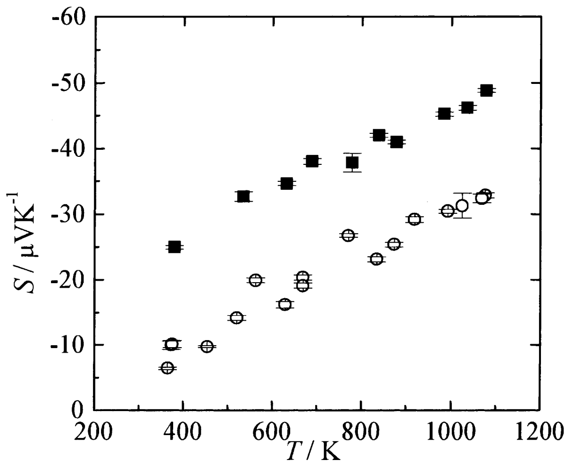

Fig. 1. Seebeck coefficients of URu_2Si_2 and $U_2Ru_3Si_5$: (■) $UR_{u2}Si_2$; (○) $UR_2Ru_3Si_5$.

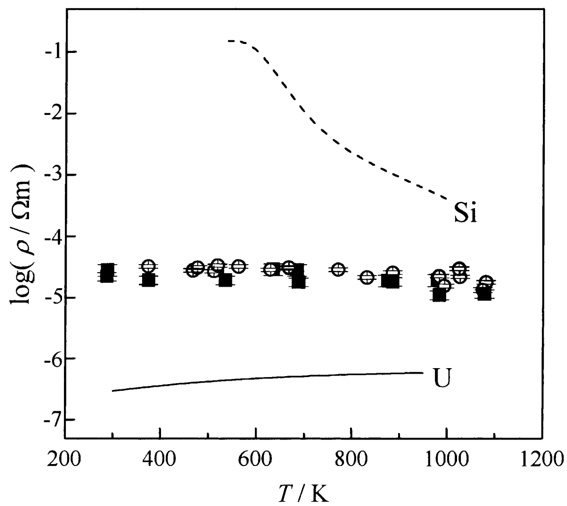

Fig. 2. Electrical resistivities of URu_2Si_2 and $U_2Ru_3Si_5$: (■) URu_2Si_2; (○) $U_2Ru_3Si_5$; (———) U [12]; (-----) Si [12].

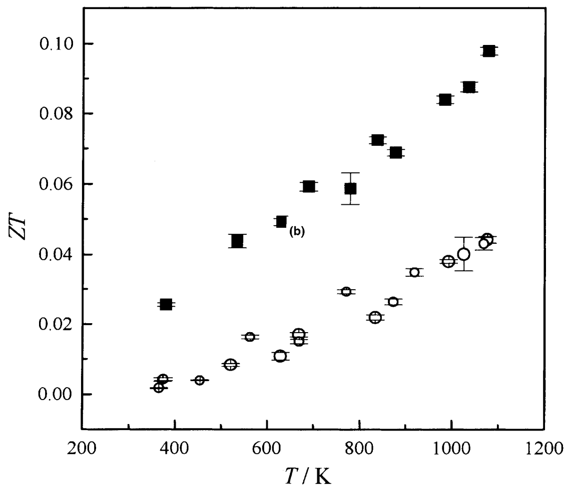

Fig. 3. Dimensionless figure of merit: (■) URu_2Si_2; (○) $U_2Ru_3Si_5$.

concentration of $U_2Ru_3Si_5$ can be expected to be similar to those of URu_2Si_2 because of the similar values and tendencies of electrical resistivities of the two materials, seen in Fig. 2. The carrier concentrations of URu_2Si_2 and $U_2Ru_3Si_5$ obtained in this study were so large that the Seebeck coefficient was small.

4. Conclusion

The thermoelectrical properties of uranium–ruthenium–silicon compounds, URu_2Si_2 and $U_2Ru_3Si_5$, were studied. The Seebeck coefficient of URu_2Si_2 which was about -24 $\mu V\ K^{-1}$ and decreased to about -49 $\mu V\ K^{-1}$, was larger than that of $U_2Ru_3Si_5$, -6 $\mu V\ K^{-1}$ at room temperature and -33 $\mu V\ K^{-1}$ at 1100 K. The electrical resistivities of URu_2Si_2 and $U_2Ru_3Si_5$ were similar and exhibited a metallic behavior. The values of dimensionless thermoelectric figure of merit of URu_2Si_2 and $U_2Ru_3Si_5$ were 0.098 and 0.043 at 1100 K, respectively. The uranium–ruthenium–silicon compounds, such as URu_2Si_2 and $U_2Ru_3Si_5$, showed poor thermoelectric properties above room temperature.

Acknowledgements

The present work was partially supported by the REIMEI Research Resources of Japan Atomic Energy Research Institute, Grant-in-Aid for Encouragement of Young Scientists, No. 11780373, 1999 and Research Foundation for the Electrotechnolorgy of Chubu No. R-10211.

References

[1] G.A. Slack, Solid State Phys. 34 (1979) 1.
[2] B. Louie, R. Radebaugh, in: Proceedings of the First National Thermoelectric Cooler Conference, 1992, p. 140.
[3] T.T.M. Palstra, A.A. Menovsky, J. van den Berg, A.J. Dirkmaat, P.H. Kes, G. Nieuwenhuis, J. Mydosh, Phys. Rev. Lett. 55 (1985) 2727.
[4] J. Sakurai, H. Hasegawa, A.A. Menovsky, J. Schweiser, Solid State Commun. 97 (1996) 689.
[5] H.F. Hundley, L.C. Bourne, A. Zettl, C. Rossel, M.B. Maple, Solid State Commun. 62 (1987) 603.
[6] L. Piraux, E. Grivei, B. Chevalier, P. Dordor, E. Marquestaut, J. Etourneau, J. Magn. Magn. Mater. 128 (1993) 313.
[7] F.R. de Boer, J.J.M. Franse, E. Louis, A.A. Menovsky, J.A. Mydosh, T.T.M. Palstra, U. Rauchschwalbe, W. Schlabitz, F. Steglich, A. de Visser, Physica B 138 (1986) 1.
[8] E. Hickey, B. Chevalier, P. Gravereau, J. Etourneau, J. Magn. Magn. Mater. 90&91 (1990) 501.
[9] Y. Onuki, T. Yamazaki, I. Ukon, T. Omi, K. Shibutani, T. Komatsubara, I. Sakamoto, Y. Sugiyama, R. Onodera, K. Yonemitsu, A. Umezawa, W.K. Kwok, G.W. Crabtree, D.G. Hinks, Physica B 148 (1987) 29.
[10] J. Sakurai, H. Hasegawa, A.A. Menovsky, J. Schweizer, J. Magn. Magn. Mater. 7 (1990) 689.
[11] U. Rauchschwalbe, Physica B 147 (1987) 1.
[12] Y.S. Touloukian (Ed.), Thermophysical Properties of High Temperature Solid Materials, vol. 1, Macmillan, New York, 1967.