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Thermal Conductivity and Thermal Diffusivity of Pulse-Heated W–Re Alloys[1]

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Results are reported for the thermal conductivity and thermal diffusivity as a function of temperature for four W–Re alloys (4.0, 21.24, 24.07, and 31.09 mass% of Re) over a wide temperature range covering the solid and liquid states. The measurements allow the determination of specific heat and dependences among electrical resistivity, temperature, and density of the alloys into the liquid phase. The thermal conductivity is calculated using the Wiedeman–Franz law. Additionally, data for thermal conductivity and thermal diffusivity of the constituent elements, tungsten and rhenium, are presented for the first time. Both metals have been previously studied with the same experimental technique.

KEY WORDS: electrical resistivity; enthalpy; high temperature; liquid alloys; rhenium alloys; specific heat; thermal conductivity; thermal diffusivity; tungsten alloys.

1. INTRODUCTION

Pulse heating experiments are used for the quantitative analysis of the thermophysical properties of metals in both the solid and the liquid states [1, 2]. For this purpose the specimen is heated by a current pulse in a very short time (microseconds to milliseconds) into the liquid state, while the cylindrical geometry of the specimen is maintained.

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Systematic investigations on iron–nickel alloys [3] demonstrated that this method also can be used for the quantitative analysis of the thermophysical properties of simple alloy systems. Moreover, by comparisons with static methods (such as the laser-flash technique and differential scanning calorimetry [3]), it could be shown that the estimation of thermal conductivity and thermal diffusivity with the Wiedemann–Franz law [see Eq. (1)] gives reliable results for simple systems, that is, systems for which no phase transitions take place in the solid state [3].

These data are of interest in the field of refractory metal metallurgy, since such alloys are widely used as thermoelectrode materials for the purpose of high-temperature thermometry. Data for liquid alloys are also needed, e.g., for computer simulation of casting processes. This work is a continuation of previous investigations on W–Re alloys [4].

2. EXPERIMENTAL METHOD AND DATA EVALUATION

For the wire samples used in this study, values of the density at room temperature, the rhenium concentration, and the solidus and liquidus temperatures are given in Table I. The current through the sample is measured with the aid of a Pearson probe, and the voltage drop is measured between two voltage knife edges (the distance between the knives is measured with a cathetometer). The signals are reduced to a low voltage with ohmic voltage dividers and are recorded by means of fast data acquisition boards (10^6 samples per second). Under consideration of the division ratios, we can calculate the voltage drop between the two voltage knifes. The radiation intensity that is emitted by the heated wire surface is recorded with a fast pyrometer. Using Planck's law along with values of the intensity at the melting transition, the actual temperature of the sample can be calculated.

Table I. Parameters of Wire Samples[a]

Sample	Mass% Re	d (10^3 kg · m^{-3})	T_{solid} (K)	T_{liquid} (K)
N1	4.0	19.3627	3325	3395
N2	21.24	19.6377	3353	3493
N3	24.07	19.6835	3333	3463
N4	31.09	19.7983	3319	3421
W		19.300	3683	
Re		21.000	3453	

[a] Mass% Re measured by means of EDAX (energy-dispersive x-ray spectroscopy); d, density at room temperature, calculated from the density of constituent elements according to the mass fraction Re; T_{solid} and T_{liquid} from Ref. 5.

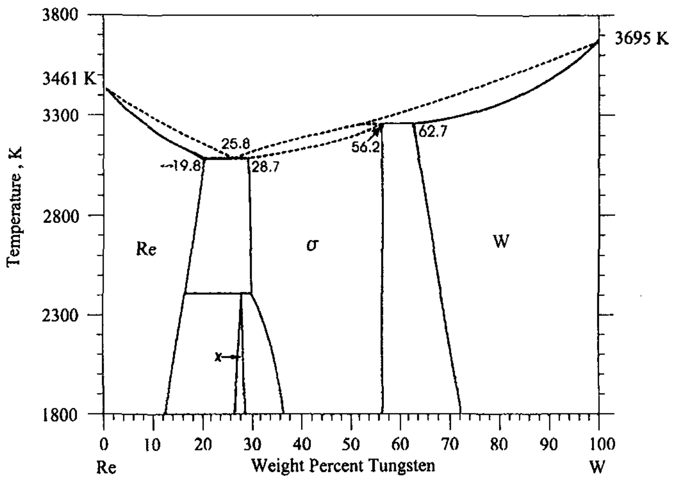

Fig. 1. The W–Re phase diagram from Ref. 5.

Alloys have a melting region instead of a single melting temperature. As suggested in Ref. 4, the present work again adopted for each alloy an averaged value of solidus and liquidus temperatures from the phase diagram in Ref. 5 as the reference temperature. More details concerning the temperature measurements are discussed in Ref. 4.

From these signals the thermal conductivity and thermal diffusivity are estimated, depending on the temperature. The specific electrical resistivity and heat capacity used for these calculations have been presented in Ref. 4.

Figure 1 shows the W–Re phase diagram [5]. For the case of three materials examined in the solid state, no phase transitions occur. During pulse heating the alloys in the solid state will be close to thermodynamic equilibrium. For alloy N4, a phase transition occurs at a temperature of about 2200 K. Phase transitions in the solid state can lead to deviations from thermodynamic equilibrium in fast pulse heating experiments [3]; however, during melting, thermodynamic equilibrium conditions will be reestablished. Therefore, the values for liquid alloy N4 again will be close to the thermodynamic equilibrium values, which can be observed in Figs. 2 and 3 showing the thermal conductivity and thermal diffusivity, respectively, since alloy N4 does not differ significantly from alloy N2 or N3.

The thermal conductivity can be estimated from the specific electrical resistivity using the Wiedemann–Franz law:

$$\lambda(T) = \frac{LT}{\rho(T)} \tag{1}$$

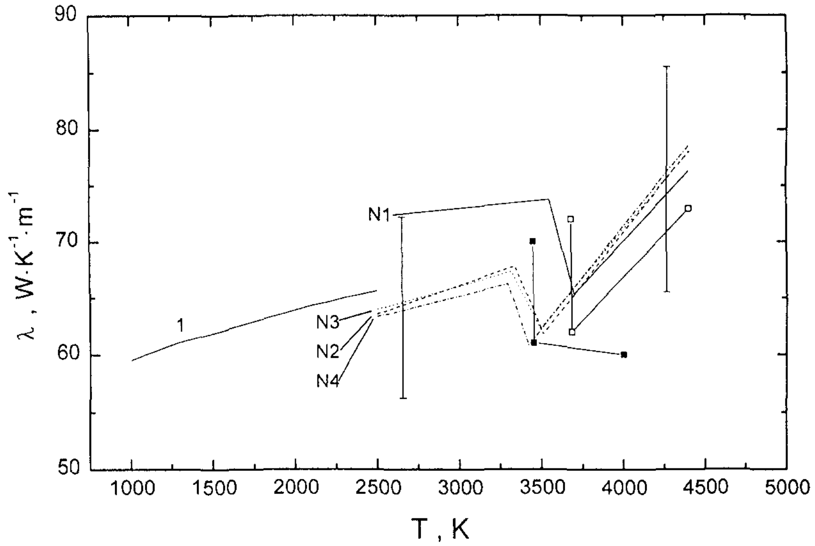

Fig. 2. Thermal conductivity of alloys N1, N2, N3, and N4 versus temperature. Open squares, values for W calculated with Eq. (1) from Ref. 7; filled squares, values for Re calculated with Eq. (1) from Ref. 7, and 1: values for Re from Ref. 8.

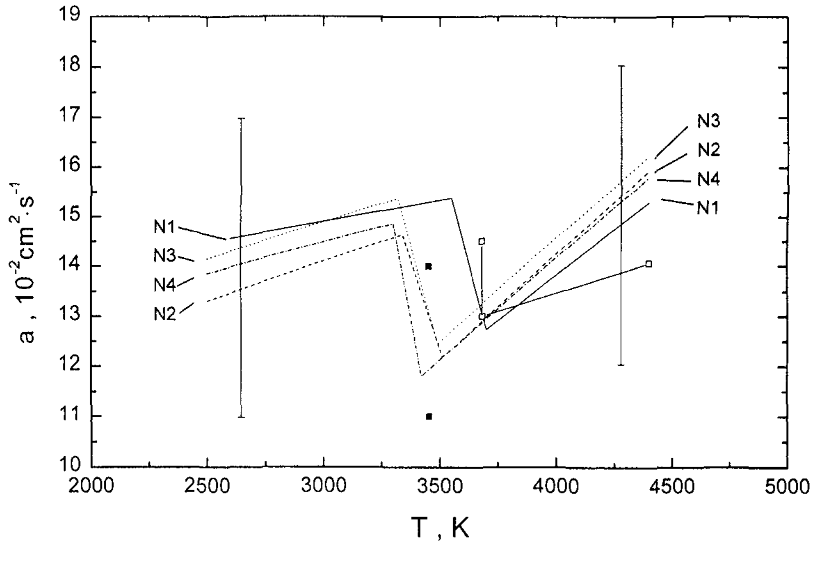

Fig. 3. Thermal diffusivity of alloys N1, N2, N3, and N4 versus temperature. Open squares, values for W [7]; filled squares, values for Re [12].

where λ is the thermal conductivity, L is the Lorenz number ($2.45 \times 10^{-8}\ \mathrm{V^2 \cdot K^{-2}}$), T is the temperature, and ρ is the specific electrical resistivity. The thermal diffusivity a can be determined from the following relation:

$$a(T) = \frac{\lambda(T)}{c_\mathrm{p}(T)\, d(T)} \tag{2}$$

where c_p is the specific heat at constant pressure and d is the density.

For the case of the specific electrical resistivity, one has to consider that a pulse heated wire expands in the radial direction and this cross-section enlargement has to be included. The expansion of the sample is measured using a high-speed CCD camera [6].

3. RESULTS

The temperature dependence of the specific electrical resistivity, enthalpy, and specific heat capacity have been reported in Ref. 4. Here, the temperature dependence of the thermal conductivity λ and the thermal diffusivity a is discussed. In the following figures, the corresponding least-square fits (see Table II) (obtained from single experiment series) for the different materials are plotted.

Table II. Least-Squares Coefficients for Thermal Conductivity λ and Thermal Diffusivity a in the Solid and Liquid States[a]

	Solid		Liquid	
	u	v	u	v
N1				
λ	68.69	1.421×10^{-3}	7.214	1.57×10^{-2}
a	12.27	8.755×10^{-4}	−0.8984	3.682×10^{-3}
N2				
λ	51.03	5.017×10^{-3}	−2.39	1.83×10^{-2}
a	9.34	1.587×10^{-3}	−2.64	4.222×10^{-3}
N3				
λ	53.82	4.067×10^{-3}	1.91	1.73×10^{-2}
a	10.41	1.493×10^{-3}	2.02	4.149×10^{-3}
N4				
λ	54.29	3.619×10^{-3}	−1.51	1.82×10^{-2}
a	10.71	1.258×10^{-3}	−2.19	4.09×10^{-3}

[a] The temperature range is from 2500 K to T_s for the solid state and from T_l to 4400 K for the liquid state. λ (or a) $= u + vT$, λ in $\mathrm{W \cdot K^{-1} \cdot m^{-1}}$, a in $10^{-2}\ \mathrm{cm^2 \cdot s^{-1}}$, and T in K.

Figure 2 shows the thermal conductivity in the solid and in the liquid temperature range for all four alloys. The values for tungsten and rhenium were estimated by means of Eq. (1) using values from Ref. 7. Data for solid rhenium from Ref. 8 are also plotted in Fig. 3. The alloys N2, N3, and N4, at concentrations of 21.24, 24.07, and 31.09 mass% Re, respectively, show more or less the same behavior. For alloy N4, which undergoes a phase transformation at about 2200 K within experimental uncertainty, no major differences compared to N2 and N3 are observed. When impurities are added to a pure metal, the impure metal will obey the standard Wiedeman–Franz law over an even wider temperature range [9].

Material N1, with only 4 mass% Re, shows a significantly different behavior. In the solid it has much higher λ values; in the liquid the corresponding λ values are less than those for N2, N3, and N4 but still higher than those for liquid W. The strange behavior of the thermal conductivity of rhenium in the liquid phase also has to be noted. References 10, 11, and 18 report decreasing uncorrected resistivity (ρ_o) values for liquid rhenium. For volume-corrected resistivity, only a slight increase with temperature can be found, which results in decreasing λ values for the liquid phase. To

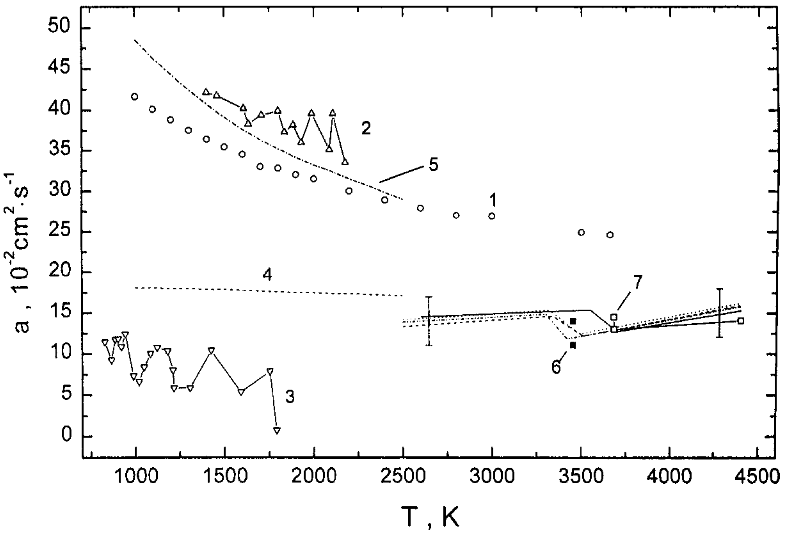

Fig. 4. Comparison of Fig. 3 to literature data. (1) Open Circles: recommended values from Ref. 13 for W. (2) Open triangles: highest literature values for W [15]. (3) Open inverse triangles: lowest literature values for W [14]. (4) Dashed line, values from Ref. 8 for Re. (5) Dashed-dotted line: values for W from Ref. 8. (6) Filled squares, recommended values for Re at the melting transition [12]. (7) Open squares: values for melting transition of W [16].

Table III. Selected Values for λ ($W \cdot K^{-1} \cdot m^{-1}$) and a ($10^{-2} cm^2 \cdot s^{-1}$) for All Alloys at Different Temperatures (T)

	N1		N2		N3		N4	
T(K)	λ	a	λ	a	λ	a	λ	a
2500	72.2	14.5	63.6	13.3	64.0	14.1	63.3	13.9
3000	73.0	14.9	66.1	14.1	66.0	14.9	65.2	14.5
T_s	73.4	15.2	67.9	14.7	67.4	15.4	66.3	14.9
T_l	60.5	11.6	61.5	12.1	61.8	12.4	60.8	11.8
3500	62.3	12.0	61.7	12.1	62.5	12.5	62.2	12.1
4000	70.0	13.8	70.8	14.3	71.1	14.6	71.3	14.2
4500	78.0	15.7	80.0	16.4	79.8	16.7	80.4	16.2

the best of our knowledge, only rhenium has this behavior, and an increase in the liquid phase, as can be seen, e.g., for W, should be the normal behavior.

Figure 3 shows the thermal diffusivity versus temperature for N1, N2, N3, and N4. In the liquid phase the values for the thermal diffusivity of N2 and N4 are very close, N3 is somewhat higher, and N1 is somewhat lower. In the solid phase there is slightly more difference in the individual plots, but here, an extreme enlargement of the *a* scale is given. Considering the large error bar in this figure, all the different alloys lie within the estimated maximum uncertainty. Also, the values for Re [12] and W [7] are given. In Fig. 4 a comparison of N1, N2, N3, and N4 to literature values for the thermal diffusivity of W and of Re is given [8, 12–16]. To include all values, the *a* axis had to be reduced; therefore, the values for N1, N2, N3, and N4 from this work are plotted in Fig. 3 on a more detailed scale.

All plots of measurements are given as linear fits. The coefficients are listed in Table II. The polynomial is of the form, physical property (λ or a) = $u + vT$.

4. ESTIMATE OF UNCERTAINTIES

For temperature measurements the uncertainty is ±5% in the melting region, increasing to about ±10% in the final liquid phase. The error in the uncorrected electrical resistivity is estimated to be ±4%, increasing to ±7% for the corrected electrical resistivity. The estimated uncertainty of the specific heat should not exceed ±12%, and that of the thermal conductivity ±15%. Thermal diffusivity values, therefore, will be uncertain by up to 20%.

5. DISCUSSION

As discussed in Ref. 12, the principal carriers for thermal conduction in solid metals are electrons and lattice waves. However, for temperatures close to the melting point of pure metals, electronic conduction is the predominant mechanism and lattice conduction is negligible. The Lorenz number L for most pure metals is close to its theoretical value ($2.45 \times 10^{-8}\ \mathrm{V}^2 \cdot \mathrm{K}^{-2}$), as discussed in Ref. 12. Therefore, Eq. (1) is a good tool to obtain thermal conductivity in the vicinity of the melting point. As discussed in Ref. 9, impurities up to 10% will not lead to drastic changes. For an alloy concentration higher than 10%, measurements on copper alloys [17] indicate that the normal Lorenz number relationship is still valid.

Contributions of convection may lead to large uncertainties in measured values for thermal conductivity, which is the major problem when using static methods on liquid samples. Due to the fast heating rate of pulsed experiments, convection is surely suppressed at the beginning of the liquid phase.

Furthermore, it is also possible to evaluate the thermal diffusivity a from heat capacity and electrical resistivity using Eq. (2). Because of the additional parameter, c_p, the specific heat capacity, with an uncertainty of about 12%, the uncertainty of the thermal diffusivity will be higher than those of the thermal conductivity obtained by pulse heating. However, for pulse heating experiments, the volume increase cancels in Eq. (2), as the same but opposite correction value is used in the corrected values for resistivity and for the actual density. Therefore, the uncertainty of the volume expansion does not have to be considered; the calculation can be performed with uncorrected (ρ_0) values and the density at room temperature. As all values are available from the measurements, the temperature dependence of the thermal diffusivity is given here, although for numerical modeling of casting processes, the thermal conductivity is the important input parameter.

As can be seen from Fig. 2, the temperature dependence of the thermal conductivity for all four alloys is strongly dominated by tungsten; in the liquid phase, the λ values of all four alloys are somewhat higher than those of W. The influence of the behavior of rhenium already discussed is negligible here. The values for W and Re at the melting transition (λ_s–λ_l) show good agreement with those of the four alloys, and extrapolation of the λ values from Ref. 8 for Re to higher temperatues shows good agreement with our estimation of λ_s for pure rhenium.

As can be seen from Fig. 4, there is a wide spread in the literature values for the thermal diffusivity of pure tungsten. The recommended values from Ref. 12 for Re agree quite well with our data; the highest values for W in the solid phase [15] are indicated by 2, the lowest values for W in the solid phase are indicated by 3 [14]. The recommended values from Ref. 13 for W are indicated by 1 in Fig. 4 and are somewhat higher than those of the W–Re alloys in this work. Values for *a* at the beginning of melting and at the end of melting for both W [16] and Re [12] show very good agreement with those of the four alloys investigated here. To explain the discrepancy of our values for all four alloys with the recommended *a* values of Ref. 13 for pure W, the contribution of Re might be the reason, as the *a* values for pure Re are much lower than those of W, as indicated in Fig. 4 by the Re values from Ref. 8. As discussed in Section 4, the maximum uncertainty of about 20% in the *a* values must always be considered during this discussion.

6. CONCLUSION

In this paper the thermal conductivity and thermal diffusivity in the temperature range from 2500 to 4400 K for four W–Re alloys (see Table I) were investigated. In a previous paper [4] on the same materials the temperature dependences of enthalpy, specific heat capacity, specific electrical resistivity, and density in the temperature range from 2500 to 4400 K were reported.

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