Heat transfer coefficients of gadolinium in the liquid state

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Abstract. The thermal conductivity and the thermal diffusivity of gadolinium (Gd) have been measured by the laser flash method in the temperature range from 1610 to 1740 K of the liquid state. The measurement error of the heat transfer coefficients was 4–6%. The fitting equations for investigated properties have been received. It has been established, that temperature dependences of transport properties of liquid gadolinium have linear character. The table of reference data for thermal conductivity and thermal diffusivity of gadolinium have been developed for scientific and practical use.

1. Introduction
An active study of the rare earth metals (REM) properties has begun since the middle of the 20-th century, when methods for their purification were developed and the metals with a relatively low content of impurities were obtained. The increased interest in the studying their properties is triggered by prospects for the use of lanthanides in various branches of science and technology, as well as the possibility of solving one of the most important problems, namely determination the relationship of the substance electronic composition with its structure and physical properties in condensed state. However, many properties of REM have not been experimentally studied, or have not been investigated in sufficient detail to date.

The thermal conductivity and the thermal diffusivity of REM have been most thoroughly studied in the low- and moderate temperature regions (near 293 K). At high temperatures corresponding to the solid phase, the data on the thermophysical properties of REM are fragmentary and inconsistent in some cases. Regarding the liquid state, the data on heat transfer coefficients of light REM are scarce and uninformative; they do not reflect changes in these coefficients with temperature variations, and discrepancy of the measurement results can be significantly greater than their total errors. The thermal conductivity and the thermal diffusivity of gadolinium in the liquid state were studied only in works [1–3]. Moreover, in [3] the thermal diffusivity of single-crystal gadolinium samples was studied in the solid state. The measurements were taken along the [1120] direction perpendicular to the hexagonal axis. More experimental information on the heat transfer coefficients of liquid gadolinium could not be found. The search for literature data and experiments on measuring the gadolinium transport properties in a wide range of temperatures of the solid state by the standard method of laser flash on “free” samples were already performed by us earlier in [4]. Based on the foresaid, the aim of this work was an experimental investigation of gadolinium thermal conductivity and thermal diffusivity in the liquid state.
2. Experimental technique

The laser flash method was used to determine the thermal conductivity ($\lambda$) and the thermal diffusivity ($a$) of liquid metal. The main experiments were performed on the LFA–427 setup [5]. The gadolinium sample of the GdM-1 brand was cut from the same metal ingot, whereof a sample was taken for measuring $a$ in solid state [4].

In the liquid state, gadolinium, like other rare earth metals, interacts with almost all materials. The most resistant to the melt of this metal is tantalum. In this regard, experiments were performed on Gd sample fused into sealed tantalum cell, the design and geometric dimensions of which were similar [6]. This approach has been repeatedly used in the study of melts and has been well tested in experiments with liquid Nd [7], Sm [8], Pr [9] and Ce [10]. The sample was formed between the bottom of the crucible and the insert and was a flat layer with about 2.4 mm thickness. The values of heat capacity ($c_p$), density ($\rho$), $a$ and thermal expansion of tantalum, required for data processing, were taken from [11–14]. Moreover, tantalum thermal diffusivity [14] was obtained on samples from the same metal, from which measuring cell was turned out.

The prepurified Gd sample and Ta cell were separately annealed in vacuum at ~$2 \times 10^{-5}$ mbar for 4 hours at temperatures of 1400 K and 1500 K, respectively. Further procedures for sealing the cell with the test sample are described in detail in [7–10].

To create good contact between the sample and parts of the cell, the measurements began from a temperature substantially exceeding the melting temperature of Gd. Data on $\rho$ [15] and $c_p$ [11] of gadolinium, as well as the calculation model, described in detail in [6], were used to obtain values of the thermal conductivity and the thermal diffusivity. The measurement error of $\lambda$ and $a$, estimated by the method of [6], was 4–6%.

3. Results and discussion

Figure 1 presents the measurement results on the thermal conductivity of Gd, which were obtained in heating-cooling cycles. It can be seen that our data are well reproduced. The temperature dependence of $\lambda$ is linear, and no anomalies can be found on it. A similar behavior of $\lambda(T)$ in liquid state was also observed in our experiments with other REM, with the exception of samarium, in which the electronic phase transition occurred, and the heat transfer coefficients of the melt nonlinearly increased with the temperature.

Results of the thermal conductivity measurements were fitted by the least squares method:

$$\lambda(T) = -6.4 + 1.393 \times 10^{-2} \, T,$$

where $T$ is the temperature in K, $\lambda$ is in W (m K)$^{-1}$.

The thermal diffusivity was calculated, using the relationship between the heat transfer coefficients ($\lambda = a \rho c_p$) according to $\lambda$, $\rho$ [15] and $c_p$ [11] measurement results. By approximating the results of the calculation by the least squares method, the following equation was obtained:

$$a(T) = -4.6 + 0.854 \times 10^{-2} \, T,$$

where $a$ is in $10^{-6}$ m$^2$ s$^{-1}$. The standard deviations of the experimental points from the approximation dependences (1) and (2) do not exceed 1.5%. The recommended $\lambda$ and $a$ values, obtained using equations (1) and (2), as well as published $\rho$ and $c_p$ data of gadolinium are presented in the table.
Figure 1. The measured values of the thermal conductivity of Gd melt. 
1 – experimental data, 2 – equation (1).

A comparison of Gd thermal diffusivity results with the data of [1–3] is shown in figure 2. Since unsteady-state methods were used in [1–3], and \( a \) was the measured value, then this quantity was compared. Authors of [1, 2] and [3] used the method of radial heat waves and dynamic method of plane temperature waves, respectively. The measurement error of \( a \) was equal to 2–3% in [1, 2] and 5% in [3]. It can be seen from figure 2, that the maximum deviation between our recommended data (equation 2) and experimental points of [1–3] does not exceed the total measurement errors.

Figure 2. Comparison of the experimental results on the thermal diffusivity of liquid Gd. 
1 – [1], 2 – [2], 3 – [3], 4 – our recommended values.
Table. The recommended data of liquid gadolinium thermophysical properties.

| $T$, K  | $\lambda$, W (m K)$^{-1}$ | $a$, $10^{-6}$ m$^2$ s$^{-1}$ | $\rho$, kg m$^{-3}$ [15] | $c_p$, J (g K)$^{-1}$ [11] |
|---------|--------------------------|---------------------------|-----------------|-----------------|
| 1600    | 15.89                    | 9.06                      | 7414            | 0.236           |
| 1625    | 16.24                    | 9.28                      | 7403            | 0.236           |
| 1650    | 16.58                    | 9.49                      | 7392            | 0.236           |
| 1675    | 16.93                    | 9.70                      | 7380            | 0.236           |
| 1700    | 17.28                    | 9.92                      | 7369            | 0.236           |
| 1725    | 17.63                    | 10.13                     | 7358            | 0.236           |
| 1750    | 17.98                    | 10.35                     | 7347            | 0.236           |
| 1775    | 18.33                    | 10.56                     | 7336            | 0.236           |
| 1800    | 18.67                    | 10.77                     | 7325            | 0.236           |

Conclusion
New reliable experimental data on the thermal conductivity and the thermal diffusivity of gadolinium melt have been received in the temperature range of 1610–1740 K. The linear character of change in the transport properties of liquid gadolinium with temperature has been established. The measurement results of this work have been compared with known literature data.

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