Physical properties of liquid gadolinium (to a temperature of 4000 k) at pulse current heating of thin foil

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Abstract. First the data were obtained for the enthalpy and heat capacity of liquid gadolinium with temperature measurement, ranging from 2000 K up to 4000 K. The heat capacity of gadolinium obtained under pulse current heating (5 µs), is consistent with the experimental data of stationary measurements near 2000 K (the magnitude of the heat capacity of liquid gadolinium equals ≈0.3 J/g·K for both cases). Above 3000 K, the heat capacity increases slightly, reaching values of 0.5 J/g·K for temperatures of 4000–4500 K (under a slightly elevated pressure).

1. Introduction

Gadolinium has the maximum absorption effect for thermal neutrons with an absorption cross section of 49,000 barns. Gadolinium alloys and compounds are used in a wide variety of applications due to their unique properties. It also has the highest isobaric heat capacity (37.07 J/mol·K) among metals under normal conditions, including lanthanides, which are characterized by increased heat capacity associated with the peculiarities of their electronic structure. Gadolinium melts at a temperature of 1591 K, boils at the temperature of 3553 K (under pressure of 1 bar). The heat capacity of gadolinium near melting was obtained in [1], back in 1974. Density and thermal expansion in the same state region are measured in [2].

The density dependence of the liquid gadolinium against temperature has no special features and for 1604 < T < 1850 K is described by an equation with a practically constant coefficient of volume expansion (6×10⁻⁵), 1/K [2]. The linearity of this dependence for the liquid state (in the range 1600–1700 K) makes it possible to extrapolate the results to higher temperatures for liquid gadolinium. As a result, for the maximum temperature in our measurements (4000 K), the density of liquid gadolinium (presumably) will be 4.8 g/cm³.

The authors [2] note that crystallization (as well as melting) of the sample did not occur at a constant temperature, but in a certain interval of ~18 K. The crystallization starts at a temperature T = 1604 ± 6 K. This value is 19 K higher than the standard value for the equilibrium melting point − 1585 K. Analysis of the authors [2] shows that this difference is not due to the methodological errors of the experiment, and contamination of samples with impurities of refractory metals, the content of which could increase during stationary experiments.

Pulse heating of conductors is usually carried out by a single pulse current of short duration. Registration of the current, voltage along the sample and the signal of the pyrometer depending on the
heating time allow calculating the heat dissipated in the sample (enthalpy), heat capacity and electrical resistance of the sample. The short heating time (~ 5 µs) makes it possible to neglect all types of heat losses. The method used to measure the thermal properties of conductors at high temperatures is described in detail in [3].

2. Specimens
The density of gadolinium samples was measured by weighing a rectangular gadolinium plate with dimensions of 0.084x31x46 mm. A value of 7.613 g/cm³ was obtained, which is lower than the density measured, for example, in [2] at a temperature of 293 K (7.880 g/cm³). Elemental analysis of 5 surface zones of this gadolinium sample (400x200 µm square each) showed that the studied gadolinium contained excess of carbon and oxygen. Average values for all 5 zones by weight % were: Gd – 93.48%; C – 3.76%; O – 2.75%. These results explain the lower specific density of the studied gadolinium.

Measuring the properties of gadolinium were performed on foil samples with dimensions of 0.04x3x15 mm, which was placed between two quartz plates with dimensions of 3.5x8x15 mm. The sample was glued to the bottom plate. For initial fixation, thin glass plates were glued to the side surfaces of the plates (figure 1). When the sample is heated in such a cell, the pressure in it increases due to its thermal expansion and inertia of the plates and, as estimates show, can reach ~ 30 bar (but not more than 100). This pressure does not affect the measurement results of the input energy (enthalpy) [4].

Thin gadolinium foil (about 40 microns) was placed between two plates (figure 1), and heated by a current pulse for 5 microseconds. Gadolinium ribbons about 3 mm wide and 18 mm long were clamped between the plates (quartz glass) to obtain cells of the 'sandwich' type (as shown in figure 1 and 2).

Figure 1. Cells with gadolinium samples (thickness 40 microns) that were prepared for pulse current heating: 1 – the tip of the gadolinium foil; 2 – thin plates were glued for holding the two thick glass plates; 3 – upper silica glass plates: size 8x15 mm, thickness 5 mm.

Figure 2. Gadolinium cell, clamped in the supply current electrodes: 1 – gadolinium foil is clamped between two quartz plates; 2 – clamp, tightly pressing the tips of the gadolinium plate to the supply electrodes through copper gaskets; 3 – supply current electrodes.

Heating was carried out in air, considering that in a short heating time of ~5 µs the oxidation of the sample can be neglected. In addition, the sandwich-type cell provided both the absence of vaporization during rapid heating and the absence of discharge on the gadolinium surface (pulse voltage on the sample is several kilovolts).

A detailed discussion of the temperature measurements during pulsed heating was discussed in [5]. The temperature of the sample was measured using a high-speed brightness pyrometer at a wavelength
of 856 nm [5], calibrated by a temperature tungsten lamp. The lower limit of measurement is 2000 K. To calculate the temperature of the samples, the data on the reflectivity of liquid gadolinium depending on the photon energy obtained in [6] were used. These data are obtained for gadolinium temperature of 1623 K, for the angle of incidence close to normal - 82°. In our case, the wavelength of the pyrometer corresponds to the energy of the photons is set at 1.448 eV, which according to [6] corresponds to the reflectivity \( R = 0.62 \). The value of the emissivity \( \varepsilon \) was found using the ratio \( \varepsilon = 1 - R = 0.38 \).

The high-speed pyrometer recorded the temperature starting from 2000 K, i.e. for gadolinium already in the liquid state. The experiment allowed us to obtain the thermal properties of liquid gadolinium: enthalpy and heat capacity, up to 4000 K. A satisfactory comparison with the equilibrium values [7] was obtained for the heat capacity near 2000 K.

3. Experimental results

For figure 3 the dependence of gadolinium enthalpy against temperature (up to 6000 K) is presented. The boiling point of gadolinium on this curve is not determined, since at high input energy and at high temperatures, the pulse pressure increases, due to inertial properties of thick quartz plates (above \( \approx 4000 \) K it is no longer possible to talk about measuring the heat capacity at a constant pressure \( C_P \)). Therefore, we give the heat capacity of liquid gadolinium \( C_P \) only up to 4000 K (figure 4). Measurement error for \( C_P \) – 15 %.

![Figure 3. Specific input energy H (enthalpy) of liquid gadolinium against temperature.](image)

At 2100 K, the obtained data on \( C_P (0.3 \text{ J/g-K}) \) within the measurement error are consistent with the data [7]. The equilibrium boiling point for gadolinium – 3553 K. There is no sign of boiling at this point (figure 4), since the cell pressure is slightly increased ( gadolinium is clamped in the silica glass plates), and the boiling point is shifted to a higher temperature. We believe that this pressure (about a few dozen bars) has a weak effect on the measured enthalpy value.

4. Conclusions

Thermophysical properties for specimens of gadolinium were obtained under fast heating by electrical pulse current. The data were obtained for the enthalpy and heat capacity of liquid gadolinium with temperature measurement, ranging from 2000 K up to 4000 K. The heat capacity of gadolinium obtained under pulse current heating (5 µs), is consistent with the experimental data of stationary measurements near 2000 K (the magnitude of the heat capacity of liquid gadolinium equals \( \approx 0.3 \text{ J/g-K} \) for both cases).
Above 3000 K, the heat capacity increases slightly, reaching values of 0.5 J/g-K for temperatures of 4000–4500 K.

Figure 4. The dependence of the heat capacity of the liquid Gd against temperature. A linear approximation of the data is shown: $C_p(T) = 0.131 + 8 \times 10^{-5} T$, J/(g·K) for the temperature range 2250–4000 K. For 2250 K, a heat capacity of 0.3 J/g-K is observed; it grows weakly with rising temperature.

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