Research of thermoradiation properties and expansion of tantalum carbide at high temperatures

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Abstract. The article describes an experimental method for studying of the thermophysical properties of refractory carbides, in particular for studying thermal expansion and radiation spectra of the sample during millisecond pulse electrical heating. As a result emission spectra of tantalum carbide were obtained in the temperature range 2400-3000 K. Also the data obtained make it possible to determine the coefficient of thermal expansion and thermoradiation properties of high refractory carbides, in particular for tantalum carbide.

1. Introduction
Carbides of refractory metals are widely used in high-temperature engineering, in particular in the nuclear industry, as well as in aviation and space technology. At the same time, the thermophysical properties of these materials at the temperatures close to the melting point of about 4000 °C for tantalum carbide, such as thermal expansion coefficient (CTE) and emissivity, are not well investigated.

The preferred method for studying the thermophysical properties of refractory carbides at given temperatures is the method of pulse electrical heating. One of the advantages of this method is high measurement accuracy (up to 2 % for CTE measurements). In the case of tantalum carbide, this approach has proven itself in the study of materials made by both sintering and spraying [1].

2. Experimental setup and experimental procedure
The description of experimental setup and characteristics of its components are considered in detail, for example, in [2]. In this article, we restrict ourselves to providing the scheme of experimental setup for electrical pulse heating of refractory electrically conductive materials, see Figure 1.

Some advances were made to the diagnostics for measuring linear expansion of the material. A detailed description of the expansion measurement technique is given below.

The expansion measurement technique used in previous papers [2-3] was modified, which led to a significant increase in accuracy. The measurements of the relative expansion of the sample using the data for cross section of the specimen had an error of the order of 10 %. New approach allows measuring of the relative elongation of the heated sample with an accuracy of 1.5-2 %, which is better for almost the order of previous magnitude.
Figure 1. Scheme of the experimental setup.

The main principle of the method used is the measurement of the absolute linear elongation of a heated sample by determining the changing position of one of the collets with which the sample is attached, while the second collet remains stationary. To determine the position of the collet, a pointer is mounted on it, and a laser with a wavelength of 532 nm and a power of up to 0.5 W is used for illumination. The scheme of the experimental technique is presented in Figure 2.

It is important to note that due to the use of the backlight of the pointer allocated to the heated sample, the possibility of optical distortions due to heating of the dense gas layer near the sample (and a change in the refractive index of the gas) during heating is excluded. As was shown earlier [4], these distortions can lead to errors in the measurement of thermal expansion comparable with the magnitude of the expansion itself. Previously we used small protective glass plate for excluding this negative influence of refraction, but presence of the plate drastically (significantly) reduced number of good experiments [4]. Beside that, measurements errors were about 10 % for refractory metals.

Using new elongation measurement technique based on absolute measurement elongation of the heated sample (for the carbide samples, it can be of the order of 0.5 mm near melting temperature), allows effectively reduce the measurement errors in comparison with the method based on the analysis of thermal images of the cross section of the sample that was used in previous works [2-3]. In addition, the use of continuous laser illumination makes it possible to carry out measurements starting immediately from the moment the heating starts using the minimum possible exposure, which also leads to an increase in the number of shooting frames in a single experiment.
3. Material and its preparation

For the experiments we used material obtained by sintering from stoichiometric tantalum carbide under pressure. The realized sintering parameters were: maximum temperature - 1950 °C, heating rate - 100 °C / min, pressure - 60 MPa, holding time - 15 minutes, matrix diameter - 15.4 mm.

The initial blanks had the size of \( \varnothing \) 15.2 x 1.1 mm and the density of 98 % of the theoretical density, the microhardness was 15.7 GPa. It should be noted that in order to avoid destruction of the material during rapid heating, it was important to achieve the maximum possible sample uniformity and material density close to theoretical. After the initial blanks were made, experimental specimens with the form of a parallelepiped with dimensions of about 1.1 x 1.5 x (9-15) mm were made using a low-speed saw.

Particle size for the initial powder was about 0.8-1.5 μm. Initial chemical composition of the material before the sintering is presented in table 1.

Table 1. Chemical composition of initial powder, weight %.

| Total C | Free C | O   | N   | Al  | Ca  | Fe  | Ti  | Na | Nb | Si | S  |
|---------|--------|-----|-----|-----|-----|-----|-----|----|----|----|----|
| 6.23    | 0.1    | 0.12| 0.025| 0.004| 0.003| 0.034| 0.002| 0.005| 0.08| 0.005| 0.005|

To study the chemical composition of the material under question, an X-ray analysis of the phase composition and structural characteristics of the starting powder used for sintering of the samples was performed. The diffraction spectra were recorded to determine the phase composition of the samples and assess their structural state by a Bruker D8 Discover diffractometer using CuK\( \alpha \) radiation and a LynxEye position-sensitive detector. The records were carried out with the increments of 0.01° along the Bragg angle 2\( \theta \) and accumulation time of 1 s per detector strip, which in total gives about 170 s for each point in the angle. The phases were identified using the BrukerAXS DIFFRAC.EVA v.4.2 software and the ICDD PDF-2 international database; TOPAS was used to determine the structural characteristics. The X-ray reflection profile was adjusted using the data for the LaB6 reference sample (NIST SRM 660b).

In figure 3 the state diagram of the Ta-C system is depicted. As one can see, tantalum carbides have a wide homogeneity region, which amounts to 41.5–49.5 atomic percent carbon (corresponds to TaC\(_{0.73}\) and TaC\(_{0.96}\) compounds) [5]. Figure 4 shows obtained diffraction spectrum. The obtained values of the crystal lattice parameter indicate a slight deviation of the studied sample from the stoichiometric composition of TaC, for which it equals to 445.6 pm.
Figure 3. The state diagram of Ta-C systems [6].

Figure 4. Diffraction spectrum of the TaC studied specimens.

**4. Experimental results**

A typical view of images captured by a high-speed camera is shown in figure 5.

The sequence of frames for clarity is located from left to right, left frames correspond to the initial stage of the experiment. Measurement of the position of the pointer relative to the vertical axis allows calculate the linear thermal expansion of the investigated material, and, accordingly, the CTE of this material.

During the experiment, emission spectra of tantalum carbide samples were obtained at high temperatures. One of the obtained spectra at a temperature close to 2400 K is shown in figure 6.
Figure 5. Typical strip photograph showing the pointer shifting while TaC specimen expands during heating.

Figure 6. TaC emission spectrum at a temperature of 2437±25 K in the wavelength range of 250-800 nm.

The obtained spectra made it possible to calculate the true temperature of the samples by the method described in detail in [7]. For this, the obtained spectral dependences were recalculated in the so-called Wien coordinates. In this case, the slope coefficient of the straight line approximating the obtained values corresponds to the reciprocal temperature. A graph of one of the spectra obtained at a temperature of approximately 3000 K, plotted in these coordinates, can be seen in figure 7.
Figure 7. The TaC radiation spectrum at a temperature of 300±30 K, plotted in the Wien coordinates.

5. Conclusion
The experiments performed demonstrated the applicability of the described method for measuring thermal elongation and radiation spectra of refractory materials during pulse electrical heating. As a result of the experiments, emission spectra of tantalum carbide were obtained in the temperature range 2400-3000 K. The data obtained make it possible to calculate the coefficient of thermal expansion and thermoradiation properties (such as spectral emissivity) of refractory carbides, in particular, tantalum carbide.

The data obtained may be of interest for high-temperature technology, since thermal expansion coefficients are an important parameter in the calculation of heat-shielding structures, and tantalum carbide (along with tantalum-hafnium carbide) is one of the promising materials for use in this field. In addition, with the help of CTE, the temperature dependence of the specific volume of tantalum carbide can be obtained, which is used to construct wide-range equations of state of substances at high temperatures.

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