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Thermal Emission of Selected Refractory Metals

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In order to extend the existing temperature ranges, an experimental investigation of the normal total emissivity of Group V transition metals of the Periodic Table (vanadium, niobium, and tantalum) was carried out up to their pre-melting temperatures. Graphical illustrations of the experimental results are presented for each of the studied metals. The obtained emissivity data show a monotonic increase over the investigated temperature range. The results are discussed and compared with literature data from other authors. The emissivity of the studied metals was also calculated and analyzed using the Furt approximation.

Keywords: vanadium, niobium, tantalum, emissivity, solid phase, temperature dependence, Furt approximation.

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Introduction

It is impossible to design, produce and operate high-temperature machines and facilities without accurate knowledge of emission characteristics of used materials [1]. The emissivity of metals in a wide range of temperatures determines a nature of radiative heat exchange and makes it possible to perform calculations aimed at intensification and improved energy efficiency of technological processes [2]. In this regard, creation and population of a metal emissivity database with the wide temperature range becomes not only a relevant task, but a strategically important need. The investigations of this kind are a key tool for studying interparticle interaction and atomic motion in the metals, thereby establishing a relationship between their microscopic processes and macroscopic physical-chemical properties.

In order to investigate temperature dependences of individual properties of the metals with a close-packed crystal lattice, the present paper has measured normal total emissivity (ε_{tn}) of polished samples of the transition metals — vanadium, niobium and tantalum. The study has significantly widened the temperature range in comparison with data available in open sources for a solid polished phase. The investigated temperature range was from 400 to 2800 K and was limited by technical capabilities of an experimental installation.

Vanadium is widely employed in atomic power engineering for manufacturing shells of fuel element, liquid heat-carrier pipelines and critical heat-loaded parts [3–5]. Presently, niobium production and application is actively widened due to unique physical-chemical properties of this metal: a high melting point, getter capability, good cold-pressure processability and weldability. These properties make niobium demanded in space and nuclear industries [3–5]. Tantalum is an element with a high atomic density. Due to its properties, it is widely used in modern engineering and in space and nuclear industries [3–5]. The higher chemical resistance of tantalum to acids is used in production of precious metals such as gold and silver.

Table 1 shows the physical-chemical characteristics of the studied metal samples.

The authors of the present paper have analyzed literature data and showed a limited number of studies dedicated to investigation of ε_{tn} of the studied elements. The obtained results are discussed and compared with the available literature data in the subsequent sections.

1. Experimental conditions

The measurements were performed in the installation whose design is detailed in the publication [8]. The experimental method is similar to the method described by the authors in the paper [9]. The experimental cell with the sample was pre-depressurized to the pressure of 10^{-5} Pa and then filled with an inert gas of argon to the pressure of 1 atm. During the experiment, the cell pressure was constantly controlled and the excessive gas was throttled. The metal samples were of a rectangular form with the sizes 20×40 mm and the thickness of ~ 2 mm. Prior to the

Table 1. Physical-chemical characteristics of the samples

Metal	Chemical purity, %	T_{melt}, K	Roughness of the surface of the sample, μm
Vanadium	99.3	2220 [6]	0.01
Niobium	99.8	2745 [7]	
Tantalum		3280 [7]	

Table 2. Basic parameters of the experiment in the study [12]

Chemical purity of the sample, %	Error of the experiment, %	Methodof heating of the sample of the investigation, K	Temperature range
99.9	±5.5	Resistive	1270–1986

experiment, they were electromechanically polished. The samples were placed in the middle of a resistive heater with a pre-polished contact area. Mandatory isothermal exposure preceded each measurement of thermal radiation. The temperature field of the sample was controlled during the entire experiment by means of a set of tungsten-rhenium thermocouples punched in the sample.

The sample radiation was recorded in the temperature range from 400 to 2800 K — the limit temperature for the applied black body model made of tantalum. The model was a hollow cylinder with an internal set of diaphragms (an optical knife) [10]. The model was designed to produce equilibrium radiation, while using a corrugated bottom (a targeting heal) allowed getting the design value $\varepsilon_0 = 0.962$ [11].

It shall be noted that a thermal flux from the surface of the samples and the model was recorded by a direct vision radiometer without using focusing optics. Due to toxicity of vanadium all the preparatory operations with the sample of this metal were performed in a laboratory aspiration box.

Reliability degree of the presented results was determined in accordance with recommendations of GOST R 8.736-2011, which is applied to evaluate measurement accuracy in fundamental and applied investigations.

The evaluation of metal emissivity was described by a function of the following magnitudes:

$$\varepsilon_{tn} = f(\varepsilon_0, \alpha_{p1}, \alpha_{p2}), \quad (1)$$

where ε_0 — emissivity of the model; α_{p1} — TEMF developed by a radiometer thermoelement when measuring model radiation $[V]$, α_{p2} — TEMF developed by the radiometer thermoelement when measuring sample radiation $[V]$.

Due to complexity of calculation of the measurement error, the authors provide final results of the calculations. The maximum measurement error was 8 % at the temperature of 405 K, while the minimum one was 5 % at the temperature of 2759 K.

2. Results and discussion

The emissivity ε_{tn} of vanadium was measured within the temperature interval from 405 to 2200 K. It results in obtaining a gently sloping, monotonically increasing curve for intensity change depending on the temperature (Fig. 1). The experimental data obtained herein have been compared with results of the study [12] to show coincidence at the qualitative level only. The experimental values of [12] are approximated by an expression of the following kind:

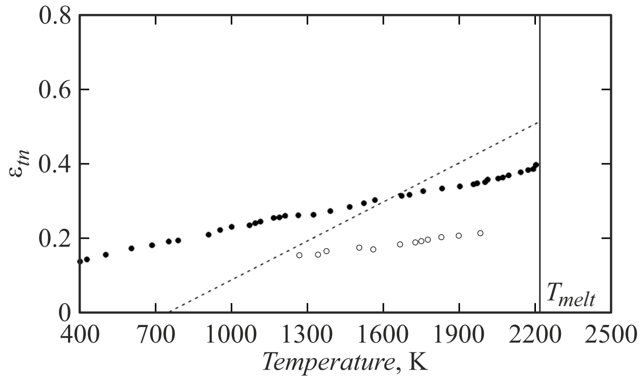

Figure 1. Dependence of ε_{tn} of vanadium on the temperature: • — the measurements by the authors; ○ — [12]; the solid line — calculation by means of the Furt approximation.

$$\varepsilon_{tn} = 0.107 + 0.005(T/1000) + 0.025(T/1000)^2. \quad (2)$$

Discrepancy between the results of the present investigation and the data of the study [12] is due to differences in the experimental approach including a method, purity of the sample and a state of their surface. The basic parameters of the experiment described in the study [12] are shown in Table 2.

Fig. 1 also shows results of the calculations by the authors using the Furt approximation [13]. The approximation is a functional dependence ε_{tn} on the resistivity $r, [\Omega \cdot \mathrm{m}]$ and the temperature $T, [\mathrm{K}]$ and takes the following form

$$\varepsilon_{tn} = 5.78\sqrt{rT} - 17.9rT + 44(rT)^{3/2}. \quad (3)$$

For calculation, the resistivity values have been taken from the study [14], wherein they are obtained by linear approximation of the experimental data presented in a large number of investigation papers. The design value ε_{tn} intersects the experimental data at the temperature of 1670 K and demonstrates overestimation up to the temperature which precedes melting.

Analysis of the literature data about thermophysical properties of vanadium by the authors has disclosed investigations dedicated to total hemispherical emissivity (ε_{th}) in the solid state. Fig. 2 shows graphic comparison of the results [17–20] with the results of recalculation of experimental values of ε_{tn} into ε_{th} by the method [2]. The

Table 3. Basic parameters of the experimental investigations of ε_{th} of vanadium by various authors

Paper	Chemical purity of the sample, %	Error of the experiment, %	Sample heating method	Temperature range of the investigation, K
[15]	99.82	±4–6.5	Resistive	1100–1750
[16]	99.7	Not specified	Electrostatic	1350–2180
[17]	99.9	±5	Pulsed	1880–2050
[18]	99.7	±3	Electrostatic	2220

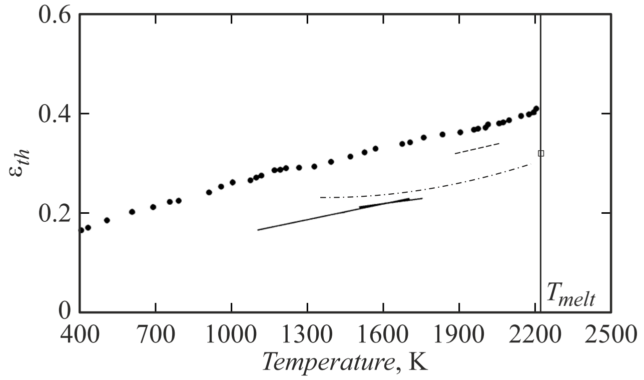

Figure 2. Dependence of ε_{th} of vanadium on the temperature: • — recalculation of the measurements by the authors; the solid lines — [15]; the dashed dotted line — [16]; the dashed line — [17]; □ — [18].

recalculation formula is as follows:

$$\varepsilon_{th} = \frac{\varepsilon_{tn}}{0.755 + 0.533 \cdot \varepsilon_{tn}}. \qquad (4)$$

The recalculation by the formula (4) is approximate estimation which allows performing qualitative comparison of the results only and imposes essential limitation on data convergence.

The study [15] has experimentally obtained values of ε_{th} of vanadium approximated by two straight lines with intersection in the point of 1600 K. Based on these data, the expressions (5) and (6) are proposed, wherein specified average deviation of the experimental points from the approximating straight lines is $\pm 0.14\,\%$.

$$\varepsilon_{th} = 0.052 + 1.05 \cdot 10^{-4} T, \ (1100-1600\,\mathrm{K}), \qquad (5)$$

$$\varepsilon_{th} = 0.113 + 0.67 \cdot 10^{-4} T, \ (1600-1750\,\mathrm{K}). \qquad (6)$$

Based on analysis of an array of the experimental data, the authors of the paper [16] have proposed an approximating expression of the following kind that is recommended for use:

$$\varepsilon_{th} = 0.38 - 2.52 \cdot 10^{-4} T + 9.9 \cdot 10^{-8} (T^2/K^2), \quad (1350-2180\,\mathrm{K}), \qquad (7)$$

where K is a coefficient of proportionality of an interval of investigation temperatures. The author of the paper [17] recommends to use the expression obtained by linear approximation:

$$\varepsilon_{th} = -5.413 \cdot 10^{-2} + 1.93 \cdot 10^{-4} T, \quad (1880-2050\,\mathrm{K}). \qquad (8)$$

The paper [18] specifies a unity value of intensity of radiation of vanadium in the melting point. Fig. 2 shows only qualitative coincidence of growth of the values of ε_{th} obtained in the papers [15–18]. It can be related to differences in technical and methodical approaches to experiment performance as well as to various chemical purity of the sample, which is shown in Table 3.

When investigating ε_{tn} of niobium within the temperature interval 410–2689 K, it results in obtaining a gently sloping, monotonically increasing dependence of intensity on the temperature (Fig. 3). The results of the authors' experiment well agree with the data recommended in the study [13], starting from the temperature of 2000 K and up to the temperature which precedes metal melting. The observed growth of ε_{tn} of niobium may be explained by formation of disordered areas in the crystal lattice of the metal in the solid phase during heating.

The same figure shows results of calculation of ε_{tn} of niobium by means of the Furt approximation. It has been calculated using an averaged array of resistivity values which was taken from the studies [19–21]. The observed behavior of ε_{tn} well agrees to a general temperature dependence.

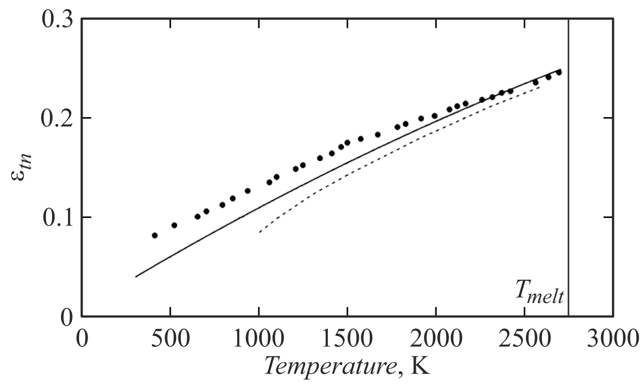

Figure 3. Dependence of ε_{tn} of niobium on the temperature: • — the measurements by the authors; the dashed line — [13]; the solid line — calculation by means of the Furt approximation.

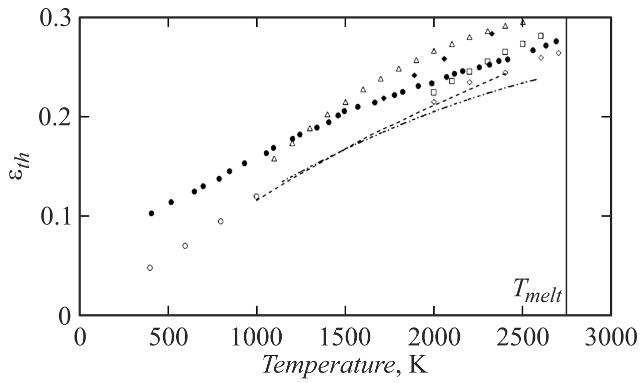

Figure 4. Dependence of ε_{th} of niobium on the temperature: • — recalculation of the measurements by the authors; the dashed line — [13]; △ — [20]; the dashed dotted line — [22]; ○ — [23]; □ — [24]; ◊ — [25].

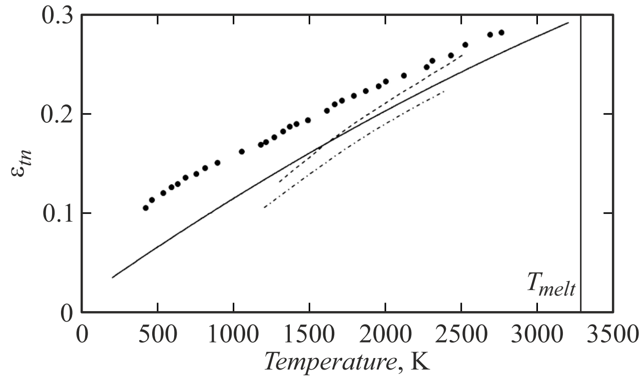

Figure 5. Dependence of ε_{tn} of tantalum on the temperature: • — the measurements by the authors; the dashed line — [13]; the dashed dotted line — [14]; the solid line — calculation by means of the Furt approximation.

Fig. 4 shows the result of similar recalculation of ε_{tn} into ε_{th} using the formula (4). There is clearly observed coincidence of the nature of variation of ε_{th} with the results obtained by the other authors.

The authors of the study [24] have presented results as a solid curve described by the quadratic function:

$$\varepsilon_{th} = -0.118 + 2.305 \cdot 10^{-4} - 2.936 \cdot 10^{-8}T^2. \quad (9)$$

The authors of the study [25] have approximated the experimental data within the entire investigated range using a polynomial of the following form:

$$\varepsilon_t h = 6.93 \cdot 10^{-2} + 7.93 \cdot 10^{-2}T. \quad (10)$$

A summary graph of dependence of intensity of ε_{th} shows that within the different temperature intervals there is a clear general trend of variation of emissivity of niobium depending on the temperature. The authors explain the available discrepancies by differences in experiment performance such as roughness degree of the samples in the solid phase, possible presence of oxide films on the surface, percent content of the basic element as well as differences in investigation technique, including sample heating procedures and instrument arrangement for recording thermal radiation from the metal surface. Table 4 generalizes the discussed parameters of the experiments done for niobium by various researchers.

When investigating ε_{tn} of tantalum within the temperature interval from 420 to 2759 K, it results in obtaining a gently sloping, monotonically increasing dependence of radiation intensity on the temperature. The results are shown on Fig. 5 to demonstrate good compliance with reliable reference data [13,22]. The same figure shows results of calculation of ε_{tn} of tantalum by means of the Furt approximation. It has been calculated using an averaged array of resistivity values which was taken from the studies [28–31].

Fig. 6 shows the results of recalculation of ε_{tn} into ε_{th} using the formula (4). The values of ε_{th} have been compared with the literature data to show averaged dependence of tantalum radiation intensity on the temperature.

In authors' opinion, the results of the study [26] demonstrate overestimated values in the temperature range 1700–2300 K. It should be noted that the data obtained by the authors of the present study also exceed the recommended values [13,22], which is probable related to the previously mentioned causes.

For calculation of the emissivity, the authors of the study [32] recommend to use a functional expression of the following type:

$$\varepsilon_{th} = -3.739 \cdot 10^{-2} + 1.644 \cdot 10^{-4}T - 1.598 \cdot 10^{-8}T^2. \quad (11)$$

The author of the study [34] describes his experimental results for emissivity of tantalum by the six-degree polynomial.

The quantitative discrepancies for the results of ε_{th} are due to the following factor: non-ideal optical smoothness of the sample surfaces, deviations of characteristics of the used black body models as well as use of the samples with a different chemical composition and content of the basic element [35–37]. It follows from the above listed that only strict adherence to the said conditions provides correct and definite obtaining of the results. In Table 5 the authors have partly generalized the discussed parameters of the experiments done for tantalum by various researchers.

Also note satisfactory performance of the theoretical approach based on the Furt approximation for calculating ε_{tn} of the solid polished phase of the metals. In all the cases, the performed calculations demonstrate qualitative coincidence of the theoretical results with the experimental data. Since reliability of the theoretical calculations directly depends on accuracy of the used values of resistivity (r) of the metals, the authors have closely selected the array of the data used in the calculations.

Table 4. Basic parameters of the experimental investigations of ε_{th} of niobium by various authors

Paper	Chemical purity of the sample, %	Error of the experiment, %	Sample heating method	Temperature range of the investigation, K
[20]	99.99	±2	Pulsed	1500–2600
[21]	99.9	±3	Pulsed	1100–2500
[23]	99.9	Not specified	Electromagnetic	300–1000
[24]	99.999	±2	Pulsed	2000–2600
[25]	99.9	±3–6	Pulsed	2000–2700

Table 5. Basic parameters of the experimental investigations of ε_{th} of tantalum by various authors

Paper	Chemical purity of the sample, %	Error of the experiment, %	Sample heating method	Temperature range of the investigation, K
[26]	99.9	±5	Pulsed	1100–2300
[28]	99.99	±2	Pulsed	1900–3000
[32]	99.9	±2	Pulsed	2000–2800
[33]	99.95	Not specified	Resistive	2400–2800
[34,35]	99.98	±2	Pulsed	1500–3200

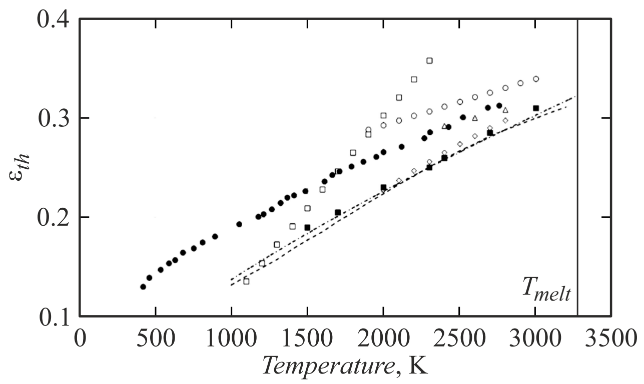

Figure 6. Dependence of ε_{th} of tantalum on the temperature: • — recalculation of the measurements by the authors; the dashed line — [13]; the dashed dotted line — [22]; □ — [26]; ○ — [28]; ◊ — [32]; △ — [33]; ■ — [34].

Conclusion

The aim of the present investigation was to study normal total emissivity (ε_{tn}) of the polished samples of vanadium, niobium and tantalum with essential expansion of the temperature ranges in comparison with the studies of other authors. The new temperature dependences of ε_{tn} have been obtained for the said metal within the interval from 400 to 2800 K. There is observed the monotonic increase of the values of ε_{tn} with an increase of the temperature. The limit temperature of the investigation was limited by technical capabilities of the experimental installation. The obtained data require further refinement as well as extending the temperature range into an area of the liquid state.

The experience of the previous investigations has shown that during study of emissivity of the metals it is necessary to strictly take into account such factors as a chemical composition of the sample, cleanliness of its surface and thermal conditions of experiment performance.

Conflict of interest

The authors declare that they have no conflict of interest.

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