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Influence of the surface condition on the normal total emissivity of titanium alloy

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Temperature dependence of the normal total emissivity (ε_{tn}) of titanium alloy VT6 was determined by a radiation-based method in the temperature range of 455 to 1710 K for two different surface conditions. For the clean surface, ε_{tn} increases monotonically. Oxidation leads to a twofold increase in ε_{tn} . The surface condition is the determining factor for ε_{tn} . The Foote approximation exhibits discrepancies with experimental data. Analysis of literature data and our own results has been systematized. The obtained results are necessary for correct specification of boundary conditions in thermal modeling.

Keywords: VT6 titanium alloy, normal total emissivity, temperature dependence, surface oxidation, Foote approximation.

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Due to a unique combination of high specific strength, corrosion resistance and thermal stability, titanium alloys are crucial structural materials in the aerospace and power engineering industries [1,2]. Accurate numerical simulation of high-temperature processes needs reliable values of thermophysical parameters, including emissivity which is critical for calculating radiant heat transfer and non-contact pyrometry [3,4].

Experimental determination of the titanium alloys' emissivity is complicated by their high chemical activity leading to rapid surface oxidation and modification of the surface condition under heating [5]. These surface effects often have a dominant influence which makes it difficult to reveal the emissivity dependence on bulk characteristics predicted by the classical theory [4]. As a result, literature data on emissivity are scattered, namely, its values vary with surface conditions and measurement technique [6,7].

Most of theoretical assessments of materials' emissivity are based on the Drude model. One of the practical consequences of that model is the Foote approximation which relates the normal total emissivity (ε_{tn}) to specific electrical resistance (ρ) and temperature (T) [8]. However, applicability of this approximation to real multicomponent alloys is limited, especially under high-temperature oxidation. The emerging oxide layers have different optical properties, which leads to a two— or three-fold increase in emissivity [6,7].

Modern fundamental research based on the electromagnetic levitation method has provided data on spectral emissivity of liquid-state alloy Ti–6Al–4V (analogue of VT6) [9–11]. However, the solid-state alloy exhibits in the operational temperature range (~ 300 – 1100 K) a lack of systematic and reproducible ε_{tn} data for controlled surface conditions.

Thus, the goal of this study was to experimentally determine the temperature dependence of solid VT6 alloy emissivity ε_{tn} for the samples with clean and oxidized metal surfaces. Additional tasks of the study include assessing applicability of the Foote approximation for predicting ε_{tn} of the system under consideration, comprehensive literature data analysis aimed at identifying factors determining the ε_{tn} spread, and formulating practical recommendations for engineering calculations.

Emissivity ε_{tn} was measured in the temperature range of 455–1710 K. This range covers not only operating temperatures but also a range of polymorphic transition $\alpha \leftrightarrow \beta$ (~ 1250 K), which allows assessing the emissivity behavior under phase transformation. We used in experiments a commercial alloy whose chemical composition matched GOST 19807–91 [12]. Prior to the experiment, the sample surfaces were subjected to mechanical grinding and polishing followed by degreasing.

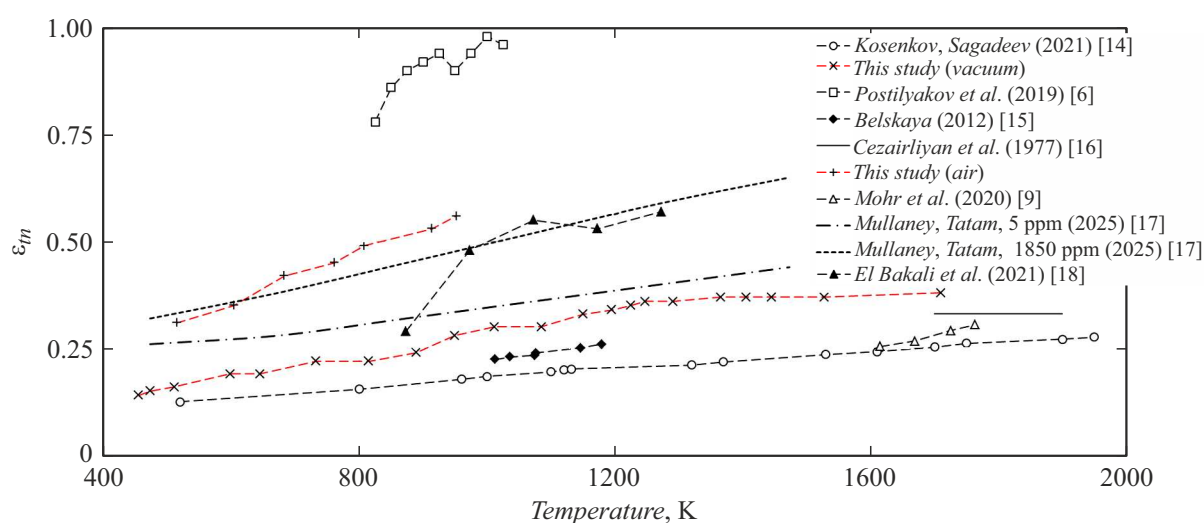
The measurements have been performed on a setup whose operating principle is based on the method of direct comparison with the absolute black body (ABB) [13]. The ABB model made of tantalum in the form of a cylindrical cup with a system of internal diaphragms had the emissivity of at least 0.96 over the entire range of operating temperatures. The sample and ABB model were heated by the resistive method. Temperature was controlled by four tungsten-rhenium thermocouples embedded into the sample's side surfaces. Thermal radiation was measured with a direct-vision radiometer with a thermoelement-based receiver whose signal (thermoelectric power) was recorded by voltmeters.

Experiments were conducted in two different modes.

1. Dynamic-vacuum measurements. The samples were heated under the residual pressure of $\sim 10^{-3}$ Pa with grad-

Experimental ε_{tn} values for alloy VT6

In dynamic vacuum						Upon oxidation in air	
T, K	ε_{tn}	T, K	ε_{tn}	T, K	ε_{tn}	T, K	ε_{tn}
455	0.14	889	0.24	1247	0.36	515	0.31
473	0.15	950	0.28	1291	0.36	604	0.35
511	0.16	1012	0.30	1366	0.37	683	0.42
598	0.19	1085	0.30	1405	0.37	761	0.45
645	0.19	1150	0.33	1445	0.37	808	0.49
732	0.22	1195	0.34	1528	0.37	914	0.53
815	0.22	1225	0.35	1710	0.38	952	0.56

**Figure 1.** Temperature dependence of the VT6 alloy ε_{tn} in comparison with literature data [6,9,14–18].

ual increase in temperature and isothermal time intervals for gas desorption from the surface. This mode ensured surface cleaning and made it possible to determine ε_{tn} characteristic of the clean metal surface of the alloy in the range of 455–1710 K.

2. Measurements after oxidation in air. To examine the oxide film influence, the samples were preheated in atmospheric air to ~ 900 K, then cooled and gradually heated with measuring the $\varepsilon_{tn}(T)$ dependence in the 515–952 K range. The upper limit was chosen so as to maintain the controllable oxide layer condition; at higher temperatures, the film structure is unstable, which prevents correct interpretation of the results. This regime simulated formation of the surface oxide layer in the mode close to real operational processes.

The error estimate accounted for the main sources of uncertainty: accuracy of maintaining and measuring the temperature and accuracy of determining thermoelectric power. The total relative error in determining ε_{tn} ranged from $\pm 3\%$ (at ~ 1700 K) to $\pm 5\%$ (at ~ 400 K).

Experimental temperature dependences of the alloy's ε_{tn} for two surface conditions are presented in the Table and Fig. 1 (for comparison, Fig. 1 presents the data from [14] for pure titanium VT1-00 in vacuum).

In vacuum, ε_{tn} increases monotonically from 0.14 to 0.38, which is typical for a clean, mirror-like metal surface. Oxidation in air leads to a sharp increase in ε_{tn} to 0.56 at 952 K. This increase indicates that the alloy's ε_{tn} is primarily a function of its surface condition rather than of the material bulk property.

To compare our results with literature data obtained for hemispherical emissivity (ε_{th}) [9,15,16], recalculation to normal conditions was performed. There is no rigorous analytical expression interrelating ε_{tn} and ε_{th} . According to [4], ratio $\varepsilon_{tn}/\varepsilon_{th}$ for polished metals is approximately 0.83 to 1. In this paper, a common coefficient of 0.85 is used for all the data. This value was chosen as a representative estimate accounting for the roughness of real samples. Emphasize that this correction is of estimative character and its purpose is qualitative comparison aimed at revealing general regularities. The error of this assumption does

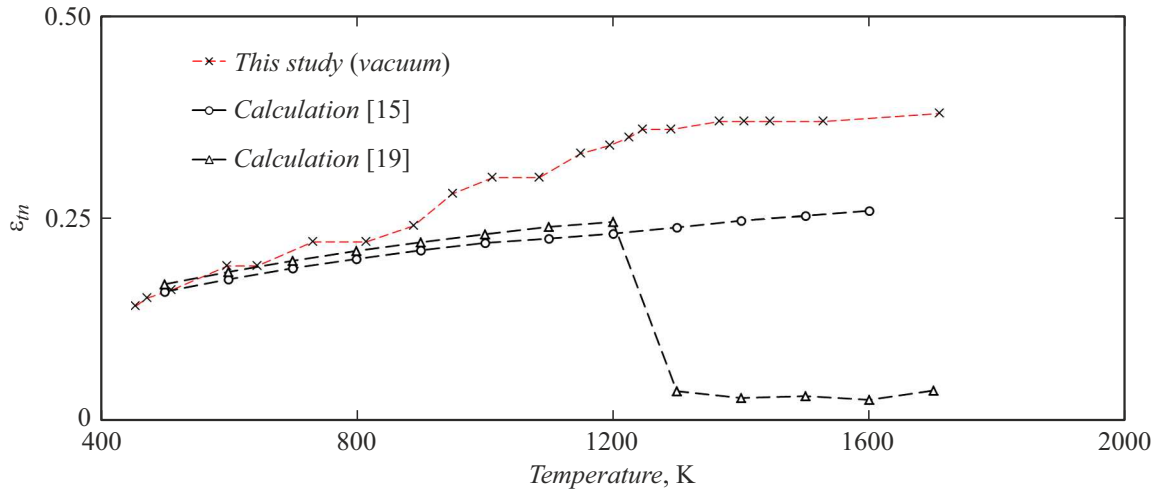


Figure 2. Comparison of experimental data on ε_m of alloy VT6 with calculations via the Foote approximation [15,19].

not exceed the spread of compared experimental data themselves and does not affect the reliability of conclusions.

It is reasonable to compare our results with data obtained under conditions simulating the process of wire-arc surfacing [17]. In the cited study, the samples were prepared in argon environments with different residual oxygen contents (< 5 to 1850 ppm), and their ε_m was determined from measurements of the room-temperature spectral reflectance. Data for the clean surface exhibit qualitative agreement. The ε_m values belong to a comparable range and demonstrate a monotonous increase with temperature. Somewhat lower absolute values observed in our case may be caused by a higher surface cleanliness provided by dynamic vacuum. For the oxidized surface, comparison with the sample [17] also revealed the expected pattern: oxidation induces a considerable increase in ε_m . This comparison clearly demonstrates that the decisive factor for ε_m of titanium alloys is the surface condition which, in turn, directly depends on the oxygen content in environment during heating.

Fig. 2 illustrates the comparison of our data with calculated curves obtained via the Foote approximation [8] which, within the Drude model for solids, relates ε_m to specific electrical resistance ρ [$\Omega \cdot \text{cm}$] and temperature T [K]:

$$\varepsilon_m = 5.78\sqrt{\rho T} - 17.9\rho T + 44(\rho T)^{3/2}.$$

For the Ti–Al–V alloys, we used experimental data on $\rho(T)$ from [15] (averaging over samples doped with Al and V); for the Ti–6Al–4V alloy, data from [19] were used.

The observed significant discrepancy between the calculated curves and experimental data, as well as between the calculated curves themselves, may be explained by a number of factors that limit the Foote approximation applicability to alloys.

1. Dopants significantly modify the electronic structure and scattering mechanisms, thus violating the simple correlation inherent in the approximation.

2. Electrical resistivity is sensitive to the phase composition, grain size and thermal history of the sample. In the cited studies, differences in the sample microstructures resulted in differences in ρ values and, hence, in calculated $\varepsilon(T)$.

3. A significant effect may be produced by interstitial impurities (oxygen, hydrogen, carbon) whose concentration may vary [20].

Thus, simple semi-empirical relationships like the Foote approximation turn out to be insufficient for quantitatively predicting ε_m of multicomponent titanium alloys; this confirms the importance of direct experimental data.

Comparison with literature data makes it possible to reveal the regularity (Fig. 1). Data obtained by the method of pulsed (subsecond) heating in an inert environment [16] exhibit low ε_m values corresponding to a clean metal surface, since the oxide film has no time to emerge in a fraction of a second. Our measurements in vacuum confirm the revealed regularity. On the contrary, static methods implying long exposure to air [6], as well as our measurements in air, provide high ε_m values, which matches the conditions for formation of an oxide layer whose intrinsic optical properties become decisive. Hence, the main source of the literature data scatter is just the difference in surface conditions (metal/oxide) caused by the measurement techniques.

Work [20] has revealed the following important aspect: emissivity variations at the temperatures of ~ 1250 K may be associated not with the phase transition (the region of polymorphic transformation), but with the kinetics of interaction with the material of gases adsorbed on the surface under the crystal lattice restructuring. In the case of rapid heating, a temporary increase in emissivity is possible due to the gas diffusion into the surface layer; then, during prolonged exposure, the emissivity decreases as a result of diffusion into the bulk. This means that neglecting such surface kinetic processes may lead to incorrect interpretation of data as a manifestation of the bulk

phase transition. Our experimental curve (vacuum) in this temperature range exhibits a weak variation in ε_{in} , which is consistent with the above-described mechanism and requires more comprehensive study.

The analysis performed allows constructing the following hierarchy of factors determining emissivity ε_{in} of titanium alloys with respect to degree of their influence.

1. The environment and time of high-temperature exposure are the key parameters controlling the growth of oxide film whose thickness and composition become the main factors in the ε_{in} increase [6,17,18].

2. The measurement technique (dynamic/static) determines which surface condition (metal or oxide) will be measured. Comparison of data obtained by different techniques without taking into account this factor is incorrect.

3. The alloy's composition and initial surface treatment play a secondary role compared to the effect of oxide layer [7,15].

In this study we succeeded in establishing key regularities determining ε_{in} of solid-state titanium alloy VT6. The alloy's ε_{in} has been experimentally shown to be governed primarily by the surface condition that depends on the environment and heating thermokinetics. In vacuum, a clean metal surface exhibits a monotonic ε_{in} increase with temperature, whereas the oxide layer formation in air leads to a sharp increase in ε_{in} . It has been found that the Foote approximation does not provide a satisfactory quantitative prediction of $\varepsilon_{in}(T)$ because of being affected by doping, microstructure and impurities; this confirms the necessity of direct experimental data. The main reason for the literature data scatter is the difference in the states of surface under different experimental conditions. While interpreting data in the region of polymorphic transformation, it is necessary to take into account the possible effect of the surface processes' kinetics. In practical thermal calculations, emissivity ε_{in} for alloy VT6 should be considered as a function of specific atmospheric conditions and temperature-time mode, but not as a constant value.

Conflict of interests

The authors declare that they have no conflict of interests.

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