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Periodicity and Correlations of the Emissivity of Liquid Metals

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A systematic analysis of the emissivity of liquid metals as a function of melting point, surface tension, and the position of elements in the Periodic Table has been carried out. Stable correlations between emissivity and thermodynamic characteristics, as well as the periodicity of emissivity variation within periods, have been established. The analysis suggests the existence of a common factor shaping both radiative and surface properties in the near-surface region of the melt. The obtained dependences can be applied to predict emissivity under conditions where direct measurements are difficult and can be used in modeling thermal radiation in high-temperature systems.

Keywords: emissivity, liquid metals, periodicity, surface tension, melting point, correlation.

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The emissivity of liquid metals is of interest both for fundamental thermal physics and for applied power engineering and metallurgy. Although a vast array of experimental data are available [1–3], systematic research aimed at identifying patterns of emissivity variation as a function of the position of elements in the Periodic Table (PT) has been limited to date. At the same time, the question of periodicity of individual thermophysical properties of metals (including characteristics related to the emissivity) was discussed in a number of fundamental studies [4,5], which formed the basis for generalizing approaches similar to the one implemented in [6].

In the present study, we perform a comprehensive analysis of the emissivity of liquid metals at temperatures close to the melting point taking into account the atomic number (Z), the melting point (T_m), and the molar surface tension (σ_m). The total emissivity was chosen for its critical role in calculations of the overall energy balance of high-temperature systems and with the aim of identifying general patterns that are less dependent on the individual spectral characteristics of elements.

A uniform data set compiled so as to ensure maximum consistency served as the basis for analysis. We used our own experimental results obtained under controlled conditions [6–10]. If direct measurement data for a certain element were lacking, the emissivity was calculated. This approach provides an opportunity to minimize systematic errors that arise inevitably when data from different sources are compared.

Data for high-purity metals were used to analyze correlations between the emissivity, T_m, and σ_m, since these properties are sensitive to impurities and surface state. The values of Z, T_m [4], σ_m [11], and normal total emissivity (ε_{tn}^m) of the liquid phase upon melting are listed in the table. Foote's approximation was used for elements with no experimental data available. It takes the form

$$\varepsilon_{tn} = 5.78\sqrt{\rho T} - 17.9\rho T + 44(\rho T)^{3/2}, \qquad (1)$$

where T is temperature [K] and ρ is resistivity [$\Omega \cdot \mathrm{m}$]. The values of ρ at T_m were taken from [12–14].

Figure 1 presents the dependence of ε_{tn}^m of metals on their atomic number. Data are grouped by PT periods and reveal a stable periodicity: within each period, ε_{tn}^m varies nonlinearly, following the patterns of electron shell filling.

The maximum values of ε_{tn}^m are observed in transition d metals (in particular, in elements with half-filled sublevels, such as V, Mo, or Re), which is consistent with the general trends in electron density and the degree of localization of electron states in the surface region of melt. Alkali and alkaline earth metals have minimum ε_{tn}^m, which reflects the predominantly delocalized nature of their electrons and the low density of states near the Fermi level [13]. The values of ε_{tn}^m for post-transition metals and semimetals decrease sharply, which is probably attributable to strengthening of covalent bonding components and a suppression of electron absorption efficiency. This pattern echoes the behavior of σ_m, which also features pronounced periodicity [11].

Analyzing the obtained sample, we come to the conclusion that the emissivity in the liquid state is not random, but varies in a regular pattern mediated by the structure of outer electron shells. The observed periodicity confirms that the radiative properties of metals remain tied to atomic characteristics even in the melt phase, where long-range order is lacking. This may suggest that correlations are preserved in the near-surface region.

Figure 2 shows the dependence of ε_{tn}^m on σ_m. The established dependence is approximated by equation

$$\varepsilon_{tn}^m = 3.1 \cdot 10^{-3}\sigma_m + 1.1 \cdot 10^{-3}. \qquad (2)$$

Numerical parameter values

Metal	Z	T_m, K	ε_{tn}^m	σ_m, kJ/mol	Metal	Z	T_m, K	ε_{tn}^m	σ_m, kJ/mol
Li	3	454	0.060	21.27	Mo	42	2888	0.291*	96.19
Be	4	1560	0.173	37.51	Te	52	723	0.048*	–
Na	11	371	0.046	15.34	Ru	44	2607	0.287*	90.77
Mg	12	923	0.093	33.7	Rh	45	2233	0.240*	81.76
Al	13	934	0.110	42.4	Pd	46	1825	0.204	62.78
K	19	337	0.035	12.33	Ag	47	1234	0.108	43.54
Ca	20	1112	0.105	31.89	Cd	48	594	0.094	33.95
Sc	21	1812	0.202	57.3	In	49	430	0.079	33.77
Ti	22	1941	0.259*	73.68	Sn	50	505	0.098	33.64
V	23	2178	0.266*	77.11	Sb	51	904	0.065*	24.42
Cr	24	2148	0.237*	61.11	Cs	55	302	0.058*	11.16
Mn	25	1517	0.132*	45.96	Ba	56	983	0.130*	30.8
Fe	26	1807	0.243*	68.14	Hf	72	2495	0.280	88.08
Co	27	1768	0.250	67.76	Ta	73	3270	0.317*	102.51
Ni	28	1728	0.200	61.82	W	74	3655	0.328*	106.3
Cu	29	1356	0.138	48.62	Re	75	3453	0.336*	113.61
Zn	30	693	0.108	33.08	Os	76	3300	0.300*	93.51
Ga	31	303	0.083*	33.76	Ir	77	2716	0.270*	91.73
Ge	32	1070	0.077*	30.52	Pt	78	2042	0.210	75.41
Rb	37	312	0.054*	11.93	Au	79	1336	0.125	55.24
Sr	38	1041	0.132*	31.2	Hg	80	234	0.080*	27.77
Y	39	1775	0.214	60.04	Tl	81	577	0.075*	29.86
Zr	40	2123	0.265	82.55	Pb	82	600	0.074	29.96
Nb	41	2770	0.271*	90.95	Bi	83	544	0.069*	–

*Calculated using Foote's approximation.

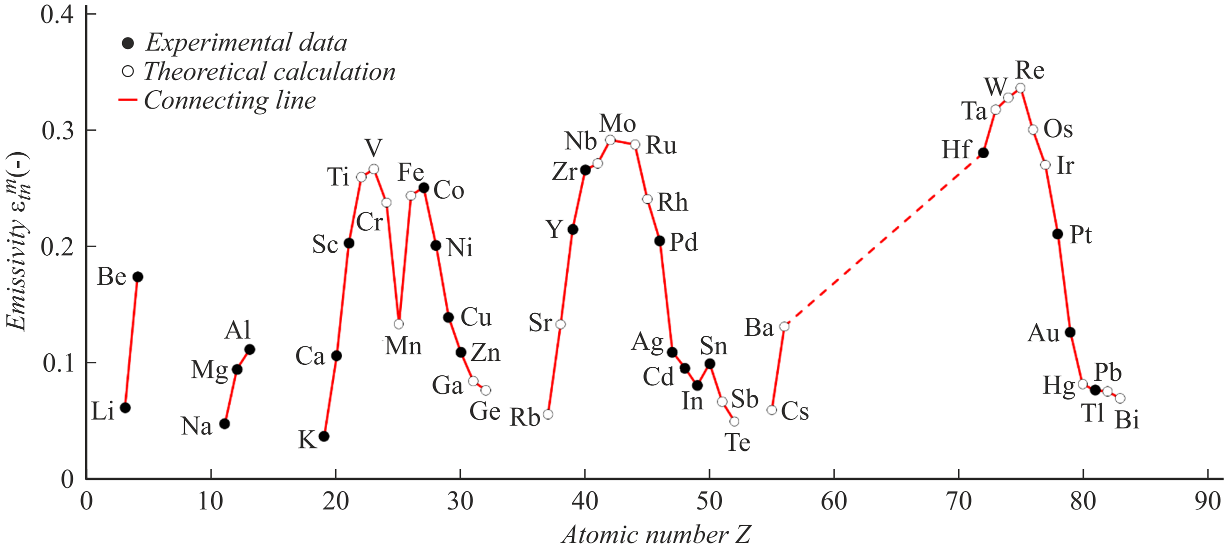

Figure 1. Periodicity of variation of ε_{tn}^m of metals.

The high coefficient of determination ($R^2 = 0.94$) and visual analysis of the scatter of points suggest that this dependence is linear in nature. The determined correlation supports the assumption of a physical connection between ε_{tn}^m and σ_m. The dependence illustrates a general linear trend. The data for alkali metals (σ_m values below 40) and d-metals (σ_m values greater than 80) are grouped in the region of low and high ε_{tn}^m, respectively. This clustered behavior is consistent with fundamental differences in the electronic structure and interatomic interactions in these groups of elements [15].

Based on this dependence, we pose the following hypothesis: in a thin near-surface melt layer ($\sim 2{-}3$ interatomic distances), both radiative and surface properties

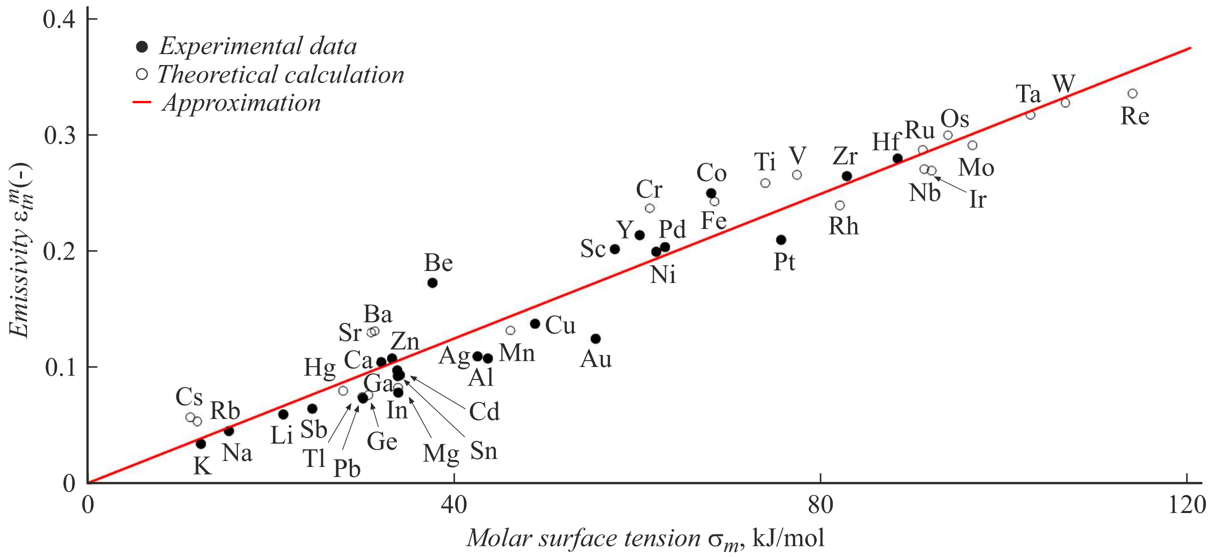

Figure 2. Correlation of ε_{tn}^m of metals with σ_m.

are formed simultaneously, shaped by the electron gas density, the degree of localization of states, and the nature of interatomic interactions. In the case of *d* metals with high electron densities, localized states may enhance both surface tension and the probability of transitions involved in thermal emission. This explains the observed growth of ε_{tn}^m with increasing σ_m. The correlation in metals with low σ_m, such as alkali metals, is weaker; delocalized electron clouds reduce the contribution to emissivity in these systems. Thus, the very structure of the near-surface layer, which changes upon transition to liquid, becomes the key to understanding both characteristics.

Figure 3 shows the dependence of ε_{tn}^m on T_m, where a strong linear correlation was also found ($R^2 = 0.92$):

$$\varepsilon_{tn}^m = 9.0 \cdot 10^{-5} T_m + 27 \cdot 10^{-3}. \tag{3}$$

The growth of ε_{tn}^m with increasing T_m may be driven by several physical mechanisms. First, the destruction of crystal lattice during melting is accompanied by partial delocalization of electrons and redistribution of the density of states (especially in the Fermi level region). This raises the probability of free transitions and reduces the surface reflectivity, promoting an increase in emissivity. Second, an increase in temperature leads to an increase in amplitude of atomic vibrations and the emergence of dynamic inhomogeneities in the structure of melt [15]. These factors alter the depth and structure of the surface layer, affecting the emissivity. Metals with high T_m tend to have strong metallic bonds and a high contribution of *d* electrons, which ensure a high density of states and, consequently, high emissivity.

Although the overall statistical significance of the revealed correlations is verified by high coefficients of determination, the scatter of experimental points relative to the approximating lines allows one to identify elements with significant deviations. In the context of dependence on σ_m, the greatest discrepancies are found for metals with anomalies in the electronic structure, which underlie the specific nature of interatomic interactions in the surface layer. These include Be, which has a small atomic radius and a significant covalent bonding component [5] that lead to the formation of a highly durable and refractory oxide film and increase sharply the emissivity in experiments [16]. Another example is Au. The anomalies of its properties are associated with relativistic effects (compression and stabilization of electron orbitals), which affect the Fermi energy and radiative characteristics of the surface [5].

As for the correlation with T_m, systematic deviations (close to, e.g., 32% for Te), are apparently related to the nature of elements themselves. In the case of semimetals, the melting point ceases to be an unambiguous indicator of the strength of metallic bonding in melt, since their properties are largely determined by the covalent component and the complex electronic structure that is characteristic of liquid semiconductors [17].

The indicated deviations do not negate the established general patterns, but are indicative of the presence of two types of physical limitations for the proposed models. The first limitation pertains to peculiar electronic structures leading to anomalous surface properties that cannot be characterized fully by parameter σ_m. The second one is related to fundamental differences in the nature of chemical bonding with melting point T_m ceasing to be a unique measure of melt metallicity. These factors need to be taken into account in order to refine predictive models of emissivity of liquid metals further.

The obtained data confirm that Z, σ_m, and T_m may be regarded not only as fundamental parameters, but also as indicators of state of the near-surface layer of melt, which determines the emissivity. The proposed

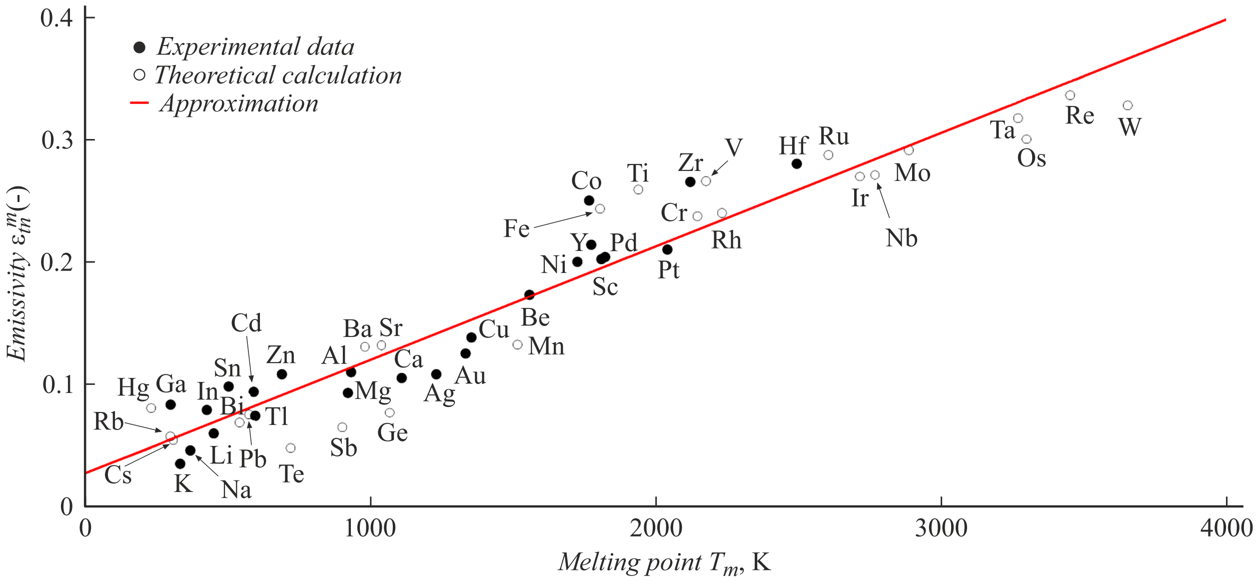

Figure 3. Correlation of ε_{tn}^m of metals with T_m.

hypothesis, which relates radiative and surface properties through their common near-surface nature, may form the basis for a new approach to interpreting the thermophysical characteristics of liquid metals. Naturally, this hypothesis requires independent verification. It may be verified by examining the surface structure of liquid metals via X-ray reflectometry [18], comparing the obtained data with spectral emissivity dependences found in literature [2,3], and performing computer modeling of the phase boundary, which allows for direct study of the structure of the near-surface layer [19].

Thus, the above analysis revealed the characteristic dependences of emissivity of liquid metals on their thermodynamic and surface characteristics. It was found that ε_{tn}^m of metals in the liquid state correlates with their atomic number, surface tension, and melting point, which is confirmed by the high coefficient of determination of the corresponding approximations. The observed periodicity of emissivity variation over the PT periods is consistent with the electronic configuration of elements, which is especially true for transition metals, where ε_{tn}^m reaches its maximum for elements with half-filled *d* sublevels.

The revealed relation between emissivity and σ_m suggests the existence of a common near-surface mechanism that shapes both radiative and surface properties of metallic melts. The obtained results are a crucial step toward a deeper understanding of the relations between thermodynamic and radiative properties of liquid metals and may find application in thermal physics and metallurgy.

Conflict of interest

The authors declare that they have no conflict of interest.

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