

Semi-empirical predictive calculation of the thermal-physical properties of potassium-sodium melts based on their component data^{*}

Sergey V. Terekhov¹

¹ A.A. Galkin Donetsk Institute of Physics and Technology, 72 Rosa Luxemburg St., 283048 Donetsk, DPR, Russia

Corresponding author: Sergey V. Terekhov (svlter@yandex.ru)

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Abstract

Use of liquid-metal coolants in nuclear power plants has been the cause of unending interest in thermal-physical properties of metals and their alloys from both experimentalists and theorists. Power polynomials are used by experimentalists to approximate temperature changes in heat capacity, the coefficient of thermal linear (volume) expansion and other quantities. These polynomials have different form in different temperature intervals and need to be joined at the interval ends. This approach creates a number of difficulties in developing a unified methodology for calculating not only the thermal functions of metals, but also for predicting the behavior of their melts and alloys. The following was used in the paper to solve the problem at hand: the author's model of a two-phase local-equilibrium region (with different order parameters) and the modified rule of component mixing, taking into account the coordinated arrays of experimental data on the initial metals for calculating predictively the thermal-physical performance of potassium and sodium melts. It has been shown that using the model of a two-phase local-equilibrium region, new approximating functions and empirical formulas lead to a sufficiently adequate estimation of heat capacities, thermal linear expansion coefficients, densities, thermal conductivities and thermal diffusivities of melts. A discrepancy has been found between the mathematical description of the thermal conductivity of the $K_{56}Na_{44}$ melt and its experimental values.

Keywords

liquid-metal coolant, mixing rule, heat capacity, coefficient of thermal linear expansion, density, thermal conductivity, thermal diffusivity

Introduction

For 70 years, liquid metal systems based on potassium and sodium and their melts have been of unending interest to researchers, since they are used as the heat removal liquid in nuclear power plants (Novikov et al. 1956; Kirillov and Troianov 1958; Rudnev et al. 1962; Subbotin et al. 1970; Alekseev et al. 1972; Kirillov et al.

1990; Kirillov and Bogoslovskaya 2000; Rachkov et al. 2014; Fokin and Kulyamina 2021; Kuzina et al. 2022; Sorokin et al. 2022). The coolants used are the eutectic alloy $K_{78}Na_{22}$ (with the melting point $T_m = 260.5$ K and the weight fractions of components $n_1 = 0.78$ and $n_2 = 0.22$), and the compound KNa_2 ($K_{56}Na_{44}$ alloy with the melting point $T_m = 280$ K and the weight fractions of components $n_1 = 0.56$ and $n_2 = 0.44$); see the potassium-sodium state

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diagram in Lyakishev et al. 2001). The thermal properties of these systems are therefore very important in investigating the thermo- and hydrodynamic processes in power plant pipelines. Thermal characteristics of melts include heat capacity C [J/(mol·K)] (or specific heat capacity c [J/(kg·K)]), linear thermal (volume) expansion coefficient α_L (α_V) [1/K], density ρ [kg/m³], and heat and temperature conductivity λ [W/(m·K)] and a [m²/s] respectively.

The classical theory of metals (Livshits et al. 1980; Grimvall 1981) is known to be based on their representation in the form of a combination of free electrons and crystal lattice oscillations (phonons). This approach allows one to obtain only qualitative results (Grimvall 1981; Koshman 2014) rather than a quantitative description of experimental datasets. For example, the Einstein and Debye heat capacity models and their different modifications do not describe the growth in the heat capacity of most substances with a temperature growth. They cannot explain the presence of spikes (structural phase transformations), rounded (phase transitions of type I, e.g. crystal-liquid) or sharp (phase transitions of type II, e.g. ferromagnetic-paramagnetic) peaks and wells on the temperature dependence diagram for heat capacity. Thermal conductivity models just record the presence of a peak in the vicinity of absolute zero on the temperature curve and indicate that the presence of impurities in the metal is the cause for its occurrence (Livshits et al. 1980). This is likely to be due to the fact that the existing theoretical constructs do not take into account the presence of sub-systems of other quasiparticles in metals (Kaganov and Lifshits 1976; Brandt and Kulbachinsky 2005). In particular, the ordering processes in them with thermal effects being present affect the type of the temperature dependence of the thermal characteristic.

On the other hand, discrepancies between experimental data on the thermal-physical properties of melts and their components ((Novikov et al. 1956; Kirillov and Troianov 1958; Rudnev et al. 1962; Novitskiy and Kozhevnikov 1975; Drits et al. 1985; Larikov and Yurchenko 1985; Zinoviev 1989; Kirillov et al. 1990; Ryabukhin 1999; Kirillov and Bogoslovskaya 2000; Kirillov and Deniskina 2000; Fokin and Kulyamina 2021) contained in different databases (Belov et al. 2020; Sechenov and Rybenko 2022) are caused by the whole number of factors. These include preliminary preparation of the sample (surface treatment, thermal annealing, degassing, and others), type and accuracy of the measuring device used, type of atmosphere in the device's inner chamber, investigation technique, recording of kinetic processes in the substance, and so on. In this connection, to describe the experimental data arrays, it is necessary to select consistent measurement results. The mathematical approximation of experimental temperature dependencies for thermal-physical performance using power functions should also be considered unsatisfactory. They are not fit to describe curves and peculiarities on these across the entire interval in which the condensed state of the substance exists. It is therefore required to describe new physical models and

use new approximate mathematical functions to describe the thermal properties of liquid metal melts used in nuclear and thermonuclear power plants.

One of the issues unresolved in modern metallurgy is predicting the properties of melts using data on similar characteristics of their components. As a result, the purpose of the study is to use a model of a two-phase local-equilibrium region (Terekhov 2018, 2023a, 2023b, 2023c, 2023d, 2024), modify the rule of mixing components (Kingery 1967) (or, in chemistry terms, rules for obtaining a compound with a given composition), take into account the experimental databases of the type described in "Database on thermophysical properties of liquid-metallic coolants of advanced nuclear reactors. Thermophysical properties of liquid eutectic alloy of sodium with potassium 56%K+44%Na", and introduce new approximating functions for calculating thermal-physical properties of potassium melts with sodium and their components.

Modified mixing rule. Consistent calculation of thermal-physical properties of potassium and sodium and their melts

Modification of the component mixing rule

To obtain a system composed of specified components with characteristics A_i and mole fractions n_i ($i = 1, 2$), rule Kingery 1967 is used:

$$A_s(T) = n_1 A_1(T) + n_2 A_2(T), \quad (1)$$

where $A_s(T)$ is the property of the system as a whole. Formula (1) produces rather approximate values of the system quantities, so it is proposed to modify it as

$$A(T) = g(T_0)[A_s(T) + f(T)], \quad (2)$$

where $g(T_0) = A_c(T_0)/A_s(T_0)$ is the adjustment factor equal to the ratio of the melt characteristic measured at the temperature T_0 to the characteristic of the system as a whole calculated at the same temperature based on the mixing rule (1); and $f(T)$ is the approximating function. In future, we will calculate the heat capacity, C , (or the specific heat capacity, c) and the linear thermal expansion coefficient, α_L , (TLEC) based on the two-phase region model formulas (Terekhov 2023a, 2023b, 2023c, 2023d, 2024), and other characteristics using known ratios, approximating functions and formulas (2).

To estimate thermal-physical properties of potassium and sodium and their respective alloys, $K_{78}Na_{22}$ and $K_{56}Na_{44}$, mutually consistent experimental data arrays were selected. There is often a major spread in experimental values obtained by different authors caused by the whole range of reasons. These include preliminary

preparation of the sample, presence of volatile impurities in same, selection of the crucible and atmosphere materials in the measuring chamber, type and accuracy class of the equipment used, etc. The use of self-consistent data made it possible to reduce the calculation errors to a level of less than 5% (except for the temperature conductivity of the $K_{56}Na_{44}$ alloy at temperatures of over 1000 K (Fig. 5).

Heat capacity

To calculate the heat capacities of potassium and sodium, we shall use the ratios provided in Terekhov 2023a, 2023b, 2023c, 2023d, 2024. The calculation formula has the form

$$C = C_b + C_k \quad (3)$$

where the first term describes the heat capacity baseline, as well as potential jumps on same (structural transformations) and is defined by the expression

$$C_b = k_1 T + \sum_j k_{2j} x_{1j} \quad (4)$$

k_1 and k_{2j} are the constant coefficients; T [K] is the temperature; and x_{1j} is the volume fraction of the ordering phase in subsystem j , which is equal to

$$x_{1j} = \frac{1}{2} \left[1 - \text{th} \left(a_j \frac{T_{xj} - T}{T} \right) \right] \quad (5)$$

a_j is the parameter related to the change in the chemical potential of phase 1 in subsystem j ; T_{xj} is the coexistence temperature for two phases with equal volume fractions. The second term in (3) is related to the implementation of phase transitions and is determined by the equality

$$C_k = T \sum_j k_{3j} u_{1j} \quad (6)$$

where k_{3j} are constants, and $u_j = \partial x_{1j} / \partial T$

Table 1 presents the model parameters for potassium and sodium, and Fig. 1 presents temperature dependencies of their specific heat capacities, and also shows the results of the predictive estimates, based on formula (2), for changes in the temperature of specific heat capacities for the $K_{78}Na_{22}$ and $K_{56}Na_{44}$ alloys. For the $K_{78}Na_{22}$ melt, the fitting factor is equal to $g(T_0) = 1$, and function $f(T) = 0$.

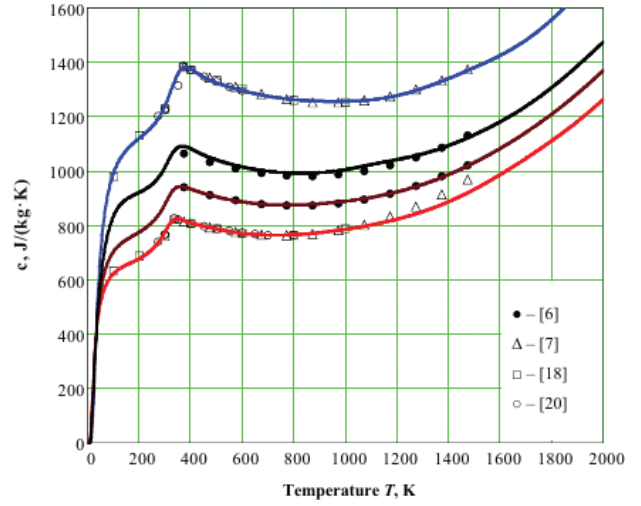


Figure 1. Temperature dependences of specific heat capacities for potassium K (red line) and sodium Na (blue line), and estimated variations in specific heat capacities for the $K_{78}Na_{22}$ and $K_{56}Na_{44}$ melts (respectively brown line and black line) with temperature based on the modified mixing rule (2).

For the $K_{56}Na_{44}$ melt, parameter $g(T_0) = 1$, function $f(T) = T [k_{31} u_1 + k_{32} u_2] - 7.6$, $k_{31} = 200$, $u_1 = dx_{11}/dT$, $x_{11}: T_{x1} = 418$, $a_1 = 0.3$, $k_{32} = 5.8$, $u_2 = dx_{12}/dT$, $x_{12}: T_{x1} = 1170$, $a_2 = 6$. It can be seen from Fig. 1 that formula (2) produces a reasonably good agreement with the experimental data.

Thermal linear expansion coefficients (TLEC)

The Grüneisen second rule (Stølen and Grande 2004, p. 14) establishes the relationship between the heat capacity (3) and the thermal linear expansion coefficient (TLEC), a_L , which makes it possible to calculate it using the formula

$$a_L \cdot 10^6 = q_1 T + q_2 x + q_3 T u, \quad (7)$$

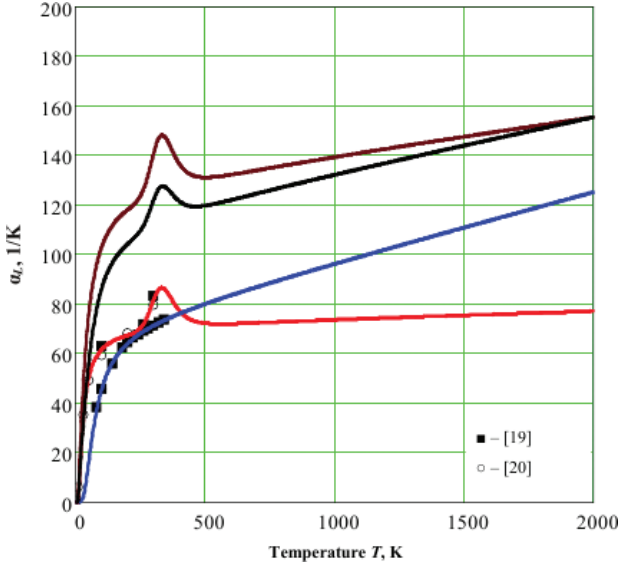
where q_1 , q_2 , and q_3 are constant coefficients. Table 2 presents the parameters of the theoretical construction for potassium and sodium, and Fig. 2 shows the dependence of their respective TLECs on temperature. The fit for the sodium TLEC calculation was based on the sodium density data. Fig. 2 also shows the results of calculating, using formula (7), the temperature changes for the $K_{78}Na_{22}$ and

Table 1. Theoretical construction parameters for calculation of heat capacities for potassium K and sodium Na

Metal	Parameters									
K	T_{x1}	a_1	$k_1 \cdot 10^3$	k_2	T_{x2}	a_2	k_{32}	T_{x3}	a_3	k_{33}
	35	0.82	2.5	32.3	336.53	7.8	0.43	435	2.0	3.6
	T_{x4}	a_4	k_{34}	T_{x5}	a_5	k_{35}	T_{x6}	a_6	k_{36}	T_{x7}
	1570	1.9	1.3	2180	2.8	7.6	2540	5.1	6.5	2800
	a_7	k_{37}								
	5.1	3.8								
Na	T_{x1}	a_1	$k_1 \cdot 10^3$	k_2	T_{x2}	a_2	k_{32}	T_{x3}	a_3	k_{33}
	53	0.78	0.7	33.6	370.94	5.8	0.8	510	1.8	3.1
	T_{x4}	a_4	k_{34}	T_{x5}	a_5	k_{35}				
	1618	4.1	1.0	2320	4.1	6.3				

Table 2. Model parameters for the potassium and sodium TLEC calculation

Metal	Parameters						
	T_{x1}	a_1	$q_1 \cdot 10^3$	q_2	T_{x2}	a_2	q_3
K	49	0.48	3	99	336.53	5.6	6
Na	98	0.81	28	84	1156.1	1.2	24.0

**Figure 2.** Temperature dependences of potassium, K (red line) and sodium Na (blue line), and estimated temperature conductivities for the $K_{78}Na_{22}$ melt (brown line) and the $K_{56}Na_{44}$ melt (black line) according to the modified mixing rule.

$K_{56}Na_{44}$ TLECs. When computing the TLECs for the melts to calculate the fitting coefficient $g(T_0)$ ($f(t) = 0$), formula by Ryabukhin 1999 was used that relates the TLECs to the melting point, T_m . Therefore, the melt TLEC predictive estimates are obtained using formula (2) with $f(T) = 0$: $g(260.5) = 1.78$ for $K_{78}Na_{22}$ and $g(280) = 1.6$ for $K_{56}Na_{44}$. The obtained results can be confirmed by measurements in the temperature intervals not explored.

Density

The temperature dependence of the substance density is determined by the formula

$$\rho(T) = \rho_0(T_0)[1 + \alpha_T(T)(T_0 - T)], \quad (8)$$

where the bulk coefficients for the potassium and sodium TLECs were computed from the following equalities ($T_0 = 293$ K)

$$\begin{aligned} \text{K: } \alpha_{T1}(T) &= 3.9 \cdot \alpha_{L1}(T), \quad \rho_{01}(T_0) = 845; \\ \text{Na: } \alpha_{v2}(T) &= 2.07 \cdot \alpha_{L2}(T), \quad \rho_{02}(T_0) = 931 \end{aligned} \quad (9)$$

Table 3. Model parameters for calculating thermal conductivities of potassium and sodium

Metal	Parameters of approximating functions							
	p	s	T_x	a_1	k_2	$T_{x1}T_{x2}$	$a_{21}a_{22}$	$k_{31}k_{32}$
K	-0.026	105.2	336.0	2000	-45.3	4.0-	1.9-	1889.4-
Na	-0.047	152.0	371.01	2000	-50.0	5.964.0	2.01.2	5444.9-124.0

Fig. 3 shows the temperature curves of the potassium and sodium densities calculated by formula (8) and the temperature changes in the densities of the $K_{78}Na_{22}$ and $K_{56}Na_{44}$ melts. The $K_{56}Na_{44}$ melt density line was plotted with fitting values $g(T_0) = 1, f(T) = 14$ based on formula (2).

Thermal conductivity

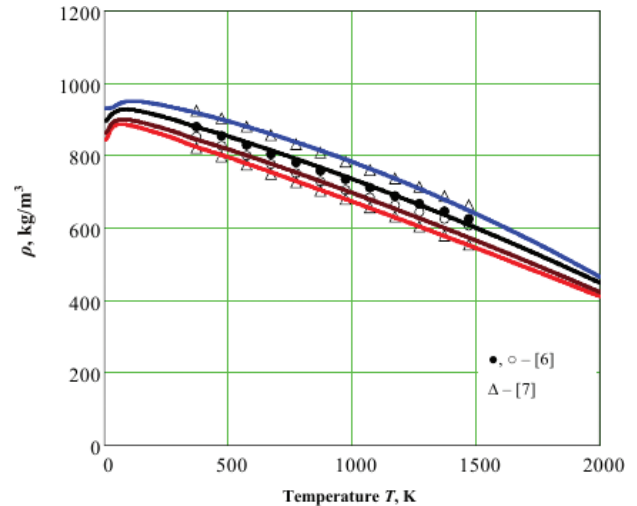
The spreading of heat in the local equilibrium two-phase region is accompanied by its thermal expansion. Temperature changes in thermal conductivities of potassium and sodium are described by the following relationships:

$$\lambda(T) = \lambda_L(T) + k_2 x_1(T) + [k_{31} u_1(T) + k_{32} u_2(T)] T \quad (10)$$

where the approximating function $X_L(T)$ has the form

$$\lambda_L(T) = pT + s. \quad (11)$$

The model parameters in formula (10) are shown in Table 3. Fig. 4 presents the temperature behavior of the potassium and sodium thermal conductivities calculated using formulas (10) and (11), and the temperature-caused variations in the thermal conductivities of $K_{78}Na_{22}$ and $K_{56}Na_{44}$, are also shown, compared with experimental data (Zinoviev 1989; Kirillov and Deniskina 2000; Novitskiy and Kozhevnikov 1975; Database on thermophysical properties of liquid-metallic coolants of advanced nuclear reactors. Thermophysical properties of liquid eutectic alloy of sodium with potassium 56%K+44%Na; Physical quantities. Guide 1991).

**Figure 3.** Temperature-dependent changes in the densities of potassium K (red line) and sodium Na (blue line), and estimated density-temperature dependences for the $K_{78}Na_{22}$ melt (brown line) and the $K_{56}Na_{44}$ melt (black line) according to the modified mixing rule.

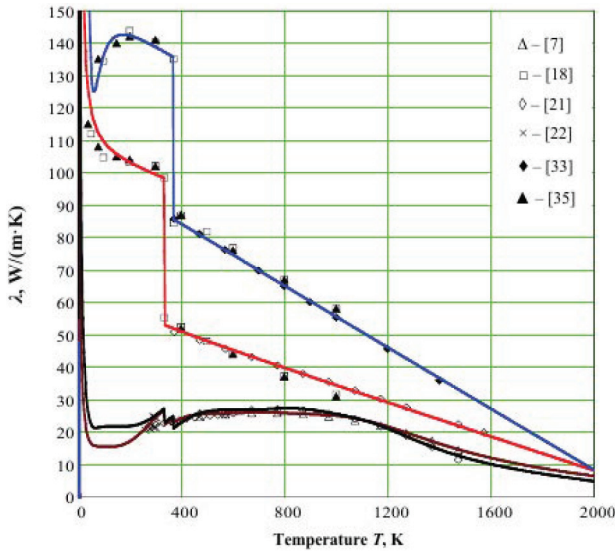


Figure 4. Temperature behavior of thermal conductivities for potassium K (red line) and sodium Na (blue line), and estimated variations in thermal conductivities of the $K_{78}Na_{22}$ melt (brown line) and the $K_{56}Na_{44}$ melt (black line) with temperature based on the modified mixing rule.

According to formula (2), the fitting coefficient for the $K_{78}Na_{22}$ melt is $g(293) = 0.133$, and the function $f(T)$ was calculated using a formula of the form (6) with the parameter values taken from Table 4. The thermal conductivity for the liquid $K_{56}Na_{44}$ melt was calculated using data from “Database on thermophysical properties of liquid-metallic coolants of advanced nuclear reactors. Thermophysical properties of liquid eutectic alloy of sodium with potassium 56%K+44%Na: the fitting coefficient is $g(293) = 0.135$, and the function $f(T)$ was calculated using a formula of the form (6) with the parameter values taken from Table 4.

Temperature conductivity

This thermal-physical characteristic defines the transport of internal energy and is calculated using formula by Sheludyak et al. 1992, p. 8):

$$a = \lambda / (c \cdot \rho),$$

Table 4. Model parameters for calculating the approximating function

Melt	Parameters											
	T_{x1}	a_1	k_{31}	T_{x2}	a_2	k_{32}	T_{x3}	a_3	k_{33}	T_{x4}	a_4	k_{34}
$K_{78}Na_{22}$	620	0.9	50	690	1.0	175	810	3.1	22	1200	2.8	48
$K_{56}Na_{44}$	640	1.4	100	980	2.9	30	1000	1.2	20	1200	5.0	8

Table 5. Changes in the thermal properties of the $K_{78}Na_{22}$ melt (top line) and the $K_{56}Na_{44}$ melt (bottom line) in the vicinity of the solid-liquid phase transition

Property	Temperature, K					
	340	350	360	370	380	390
c [J/(kg·K)]	936.11078	940.81086	942.11090	941.11090.7	938.81088.9	935.91085.5
$\alpha_L \cdot 10^6$ [1/K]	148.2127.9	147.127.4	145.1126.5	143.125.4	140.9124.3	138.9123.4
ρ [kg/m ³]	852886.4	849.6884.3	847.4882.2	845.2880.2	843.1878.3	841.1876.4
λ [W/(m·K)]	22.523.1	23.223.7	23.824.4	24.325	23.321.7	23.822.3
$a \cdot 10^6$ [m ² /s]	31.124.1	31.924.7	32.825.4	33.626.1	32.422.7	33.223.5

where λ is thermal conductivity; $c = C/M_a$ is specific heat capacity; $M_a, 10^{-3}$ [kg/mol] is the atomic (molecular) weight of metal (melt), ρ – density.

Fig. 5 demonstrates the temperature curves of thermal conductivities for potassium ($M_{a1} = 39.098 \cdot 10^{-3}$), sodium ($M_{a2} = 22.989 \cdot 10^{-3}$) and their melts calculated using formula (12). Table 5 shows the values of the thermal-physical characteristics for the melts in the vicinity of the solid-liquid phase transition.

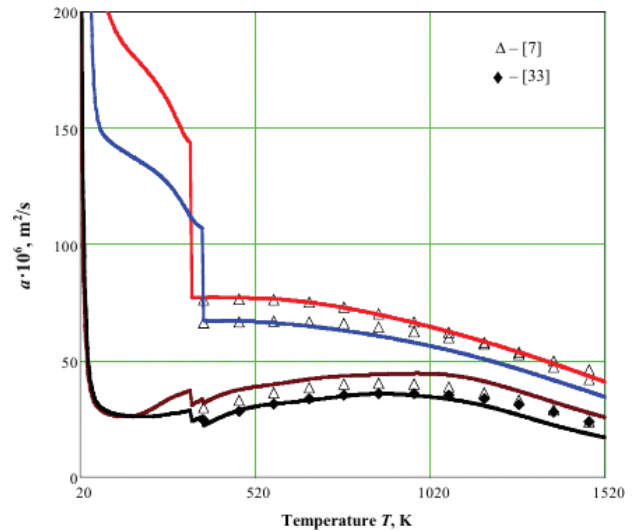


Figure 5. Temperature dependences of thermal conductivities for potassium K (red line) and sodium Na (blue line) and estimated variations in temperature conductivities of the $K_{78}Na_{22}$ melt (brown) and the $K_{56}Na_{44}$ melt (black line).

Conclusions

Knowing the only experimental value of a thermal-physical characteristic, one can use a model of a two-phase region, a modified mixing rule and new approximating functions to undertake a practically consistent calculation of heat capacities, thermal linear expansion coefficients, densities, and heat and temperature conductivities for potassium-sodium melts and their components, the

temperature range being from absolute zero temperature to evaporation temperature. Adjustment of theoretical results using a modified rule for obtaining a system with a given composition leads to the development of a sequential program for calculating the thermal-physical properties of the melt and its components. This fact is confirmed by a fairly adequate description of the thermal conductivities of potassium, sodium and their melts, obtained using

adjusted values of the thermal-physical quantities of components. Differences between theoretical and experimental data for the temperature conductivity of the $K_{56}Na_{44}$ melt at temperatures of above 1000 K may be caused by the kinetic processes that form the liquid-gas phase transition. Therefore, there is a need for precision studies into the temperature conductivity of the $K_{56}Na_{44}$ melt for temperatures of above 1000 K.

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