Development of methods for calculating thermal conductivity of gas condensates-alternative motor fuels

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Abstract. The article is devoted to the development of methods for calculating the thermal conductivity of complex hydrocarbon mixtures such as gas condensates, which are widely used as components for motor and jet fuels. The work was carried out on the basis of the results of experimental studies of thermal conductivity of the eight most representative deposits in Central Asia in terms of group hydrocarbon composition. An analysis of existing equations for calculating the thermal conductivity of liquid petroleum products and the possibility of their use for gas condensates was carried out. A number of new equations are presented for calculating the thermal conductivity of gas condensates depending on the group hydrocarbon composition in a wide range of temperatures and pressures.

1 Introduction

A feature of the natural gases produced in Uzbekistan is the presence in them of natural mixtures of liquid hydrocarbons - gas condensates, which are valuable raw materials for the gas processing and petrochemical industries. Taking into account the size of gas production, as well as the gas condensate factor (which is within 20-30 cm3/m3), the total annual collection of gas condensate by 2025 could be ~ 1-1.5 million tons. [1]. Only in the depths of the Kokdumalak deposit there are about 10 million tons of condensates [2].

To carry out scientifically based calculations of heat transfer surfaces, technological processes and create modern large-scale production facilities for the chemical processing of gas condensates, it is necessary to have reliable values of thermophysical properties, in particular thermal conductivity λ. The role of thermophysical data in the design process is especially significant. The reliability of calculations of technological processes and equipment directly depends on the availability and accuracy of thermophysical information. The use of approximate data on the properties of substances in engineering calculations leads to a significant overestimation of the metal intensity of technological installations and a decrease in their technical and economic indicators.

In recent years, intensive research has been carried out on the use of gas condensates as components of motor and jet fuels. To analyze and gain a deeper understanding of the

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phenomena preceding combustion and to clarify the physical essence of the processes occurring in the engine cylinders, knowledge of the thermophysical properties of the fuels used is also necessary.

Work [3] presents the results of an experimental study of the thermal conductivity of gas condensates from eight fields in Central Asia (Shatlyk, Shakhpakhty, Gugurtli, North Mubarek, Naip, Achak, Gazli and Uchkyr) of various group hydrocarbon compositions in a wide range of temperatures and pressures. In the literature there are a number of semi-empirical and empirical formulas that make it possible to determine the thermal conductivity of liquids belonging to certain homologous series and groups with acceptable accuracy. Many of these equations are not applicable to the calculation of such complex mixtures as gas condensates, since they assume the availability of data on the structural and group hydrocarbon composition, critical parameters and molecular characteristics, etc., which, as a rule, for multicomponent mixtures or difficult to determine. or to a certain extent conditional.

Below we consider those of the proposed equations that can be used to calculate the thermal conductivity of mixtures of liquid hydrocarbons.

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$$\lambda = B\rho^{4/3}, \tag{1}$$

where

$$B = A \cdot C_p \cdot M^{1/3} = const. \tag{2}$$

The value of B can be determined from the experimental value of λ for any one temperature. A check of equation (1) carried out by Vargaftik showed that for the 12 liquids studied, the calculated values differ from the experimental ones by no more than 5%. Using experimental data on heat capacity and density, we calculated the thermal conductivity values of the studied gas condensates using equations (1) and (2). A comparison of the calculated thermal conductivity values with the experimental ones showed that for gas condensates good results are obtained when calculating thermal conductivity according to formula (1), the standard deviation of the calculated values from the experimental ones is ±1.8%. Lenoir [4] proposed an equation for determining the dependence of the thermal conductivity coefficient of liquids on density

$$\left(\frac{\lambda_p}{\lambda_{P_0}}\right)_t = \left(\frac{\rho_P}{\rho_{P_0}}\right)^z \tag{3}$$

$$z = -2{,}94\ T/T_{кр} + 3.77, \quad (4)$$

where T_{cr} is the critical temperature, K In work [5], which presents experimental data on the thermal conductivity of a large group of liquid n-alkanes in the temperature range 20-200°C and pressures up to 49.0 MPa, it is shown that calculations of λ using formulas (3) and (4) lead to systematic underestimations, which at 180°C reach 8%. In this regard, an adjustment to the Lenoir equation has been proposed by changing the coefficient in equation (4) [5]. According to the author, when showing the degree

$$z^1 = -2{,}94\ T/T_{кр} + 4.55, \quad (5)$$

The deviation of the calculated values of k from the experimental ones for all liquid n-alkanes studied does not exceed 1.5%. The possibility of using the adjusted Lenoir equation for complex mixtures, in particular for calculating the thermal conductivity of narrow fractions of Ozeksuat oil, has been studied. The obtained calculated values of thermal conductivity differ from the experimental ones by no more than 1.5%. The good results obtained in this work for petroleum products prompted the use of the corrected Lenoir formula to determine the pressure dependence of the studied gas condensates. Critical temperatures of gas condensates were determined by equation [6, 7], based on the expansion coefficient of the liquid

$$T_{кр} = \frac{T_2 - T_1}{(\frac{\rho_1}{\rho_2})^{\frac{10}{3}} - 1} + T_2 + 6\ , \quad (6)$$

where T1 and T2 are the temperatures in K at which the liquid densities ρ1 and ρ2 are measured.

The critical temperatures calculated by equation (6) for some of the studied gas condensates have the following values: Shatlyk - 654 K, Shakhpakhty - 650 K, Gugurtli - 620 K, Naip -612 K, Gazli - 610 K. Comparison of the calculated thermal conductivity values of gas condensates using equations 3 and 4 in the temperature range 313–423 K and pressure 0.1–40.0 MPa with experimental results shows that the standard deviation is ±1.7%.

The main disadvantage of the considered methods for calculating thermal conductivity at elevated pressures is that it is necessary to know the temperature dependence of thermal conductivity and *P-V-T* data, the determination of which is associated with experimental difficulties. In this regard, the method for calculating the thermal conductivity of oils and petroleum products at various *P* and *T*, proposed by a group of employees of the Industrial Thermophysical Laboratory of the Grozny Petroleum Institute (OTFL GNI) [5], which is based on the principles of structural group analysis, that is $\lambda = f(\rho_4^{20}, n_D^{20}, M, T)$. The essence of the method is as follows: according to the data n_D^{20} (refractive index), ρ_4^{20} (relative density), *M* (molar mass), the thermal conductivity of the oil product at a fixed temperature value λ_{20}^P and the temperature coefficient are calculated α^P, then, using the well-known formula

$$\lambda_t^P = \lambda_{20}^P \left[1 - \alpha^P (t - 20)\right], \tag{7}$$

calculate the temperature dependence of thermal conductivity. Further, to calculate the thermal conductivity of petroleum products under pressure, a relationship similar in form to the Tate equation for specific volumes is recommended

$$\lambda_{P,t} = \frac{\lambda_t}{\left[1 - A \ln \frac{B(T) + P}{B(T) + P_0}\right]}, \tag{8}$$

where *A* and *B(T)* are some constants. The value of *A* is assumed to be the same for all petroleum products - 0.1988, *B(T)* is found by methods of similarity theory using the physicochemical characteristics (n_D^{20}, ρ_4^0) of the substances under study.

The results of calculating λ of gas condensates using this method and comparing them with experimental data showed that the standard deviation of the calculated values from the experimental ones is within ±4...5%, reaching a maximum of ±7...12%.

The work [3] presents the results of an experimental study of the thermal conductivity of gas condensates, carried out by the author, in a wide range of temperatures and pressures. By jointly processing thermal conductivity with the physicochemical characteristics of the studied objects, we obtained equations that describe the temperature and pressure dependences

$$\lambda_t = a_0 - (a_1 - a_2 T - a_3 T \rho_4^{20}) \Pi + (a_4 - a_5 \rho_4^{20}) T, \tag{9}$$

where a_0 = 0,1796; a_1=4.917·10^{-4}; a_2=5.973·10^{-4} ; a_3=8.432·10^{-4}; a_4=0.083·10^{-4}; a_5=3.139·10^{-4}; ρ_4^{20} -

relative density; *П* – proportion of paraffin hydrocarbons in gas condensate or fraction The equation was tested on 57 gas condensate systems. The standard deviation of the thermal conductivity values calculated using equation (9) from the experimental ones is within 2...5%, the maximum deviations reach 12%.

The pressure dependence of the thermal conductivity of gas condensates and fractions is described by the equation

$$\lambda_{P,T} = \lambda_t + (a_0 - a_1 T_k + a_2 T)(P - P_0), \tag{10}$$

where a_0 = 3,263·10^{-4}; a_1=4.225·10^{-7}; a_2=7.381·10^{-7}; λ_t – temperature dependence of thermal conductivity, *W/(m•K); T_k* volumetric boiling point, *K*; *T*—temperature, *K*; *P* - test pressure, *MPa*; P_0—atmospheric pressure, *MPa.* The calculation error using the equation reaches 5...15%.

Thus, analysis of the results of calculating the thermal conductivity of petroleum products using existing equations and comparisons with experimental values show that most of the considered equations do not reflect experimental data with the required accuracy and only some of them can be recommended for calculating the thermal conductivity of gas condensates. In particular, for calculating the temperature dependence of the thermal conductivity of gas condensates, the Vargaftik formula (1) gives satisfactory results, and at elevated pressures, the Lenoir equation with corrected exponents (3), (4). However, to use them, it is necessary to have initial experimental thermophysical

information. Using the GNI OTFL technique to calculate **λ** of gas condensates generally gives satisfactory results, but in some cases the calculation error reaches ±7.0%.

The obtained experimental material and the established features of the dependence of **λ** of the studied products on physical and chemical characteristics make it possible to obtain generalizing dependencies for calculating **λ** of gas condensates in a wide range of temperatures and pressures without initial experimental information with higher accuracy. The research results are presented below.

2 Recommended methods for calculating the thermal conductivity of gas condensates

It is shown in [6] that a generalization of experimental data on the transfer coefficients of petroleum products and gas condensates can be carried out on the basis of the theory of the corresponding states. The theory of corresponding states is usually applied to petroleum products in two ways:

1) the volume-average boiling point is taken as the characteristic temperature;

2) the characteristic temperature is determined by two density values of a given oil product. We investigated the possibility of using both methods to generalize experimental data on the thermal conductivity of gas condensates.

The law of corresponding states for the thermal conductivity of liquids at pressures close to atmospheric is represented in the form

$$\lambda_\tau / \lambda_0 = \varphi(\tau) , \tag{11}$$

where λ_τ —is the thermal conductivity coefficient at temperature T, λ_θ - is a constant value for each liquid and determined by the method of setting the characteristic parameters; $\varphi(\tau)$ - is a universal function for thermodynamically similar substances; τ - reduced temperature.

Let us consider the case when the volume-average boiling point of the studied gas condensates is taken as the characteristic temperature. Then formula (11) takes the form

$$\lambda^* = \varphi\ (T/T_к), \tag{12}$$

where $\lambda^* = \lambda_\tau / \lambda_{\tau=0,8}$ − reduced thermal conductivity; T_k - is the volumetric boiling point of gas condensate, *K*.

The volume-average boiling point of gas condensate was determined according to the method given in [7]. The values of $\lambda\tau = 0.8$ were found at the reduced temperature value corresponding to $\tau = 0.8$ in order to process experimental data without extrapolation. As a result of graphical processing of the dependence of the reduced thermal conductivity of gas condensates on the reduced temperature in the temperature range 313-423 K, the following equation was obtained

$$\lambda^* = -0{,}837\ \tau + 1{,}671. \tag{13}$$

The standard deviation of the thermal conductivity values calculated by equation (13) from the experimental ones is ±1.0%. In the case when the characteristic temperature is determined from the temperature dependence of the density of the oil product ($\tau = \beta \cdot T$), formula (11) can be presented in the form

$$\lambda^{**} = \lambda / \lambda_{\tau=0,29} = \varphi(\beta T) \tag{14}$$

where $\lambda_{\tau=0.29}$ is the reference value of thermal conductivity at a temperature corresponding to $\tau = 0.29$ (the value $\tau = 0.29$ is taken from the condition of describing experimental data without extrapolation); β - is the temperature coefficient, determined from two density values according to the formula

$$\beta = \frac{\rho_1 - \rho_2}{\rho_1 T_2 - \rho_2 T_1}, \tag{15}$$

where ρ_1 and ρ_2 are the density of the oil product at temperatures T_1 and T_2 The accuracy of determining β depends on the selected temperature range and the error in determining ρ_1 and ρ_2, which should be quite high, on the order of ±0.05–0.1% [5]. To process experimental data on the thermal conductivity of gas condensates using the method under consideration, experimental data on the density of gas condensates in Central Asia [8-26], which were measured according to the author's estimate with a maximum error of ±0.1%, were used. The proposed empirical equations for calculating λ of heavy petroleum products, obtained on the basis of the correlation between thermal conductivity and complexes and in the temperature range −20...150°C, give an error of no more than 8%. We have attempted to extend these correlations to gas condensates. As calculations have shown, for the studied gas condensates, satisfactory results are obtained when using the complex $C_P^{30}\rho_t^{4/3}$. The dependence of thermal conductivity λt on the complex $C_P^{30}\rho_t^{4/3}$ within the temperature range of 40–150°C is described by the equation

$$\lambda_t = 0{,}098 C_P^{30} (\rho_t \cdot 10^{-3})^{4/3} - 0{,}0277, \tag{16}$$

where C_P^{30} - is the heat capacity of gas condensate at a temperature of 30°C, *kJ/kg·K*; ρ_t—density of gas condensate at temperature *t°C, kg/m3*. The application of formula (16) to calculate the thermal conductivity of the studied gas condensates showed that the root-mean-square error is ±1.0%, the maximum is 4.2%. Unlike equations (1), (13), the calculation of thermal conductivity using equation (16) does not require experimental values of λ, however, it is necessary to have data on the heat capacity (at 30°C) and the temperature dependence of density. As was shown above, the use of the law of corresponding states to calculate the thermal conductivity of gas condensates gives good results, but to use the resulting equations it is necessary to have at least one experimental value of the thermal conductivity of the gas condensate under study, which reduces their practical value. By processing our experimental data in λ_t - τ coordinates with subsequent correlation by molar mass *M*, we obtained a dependence that allows us to calculate the thermal conductivity of gas condensates without initial experimental information

$$\lambda_t = 0{,}12\ (1{,}693 - 0{,}833\ \tau - M^*), \tag{17}$$

where $\tau = T/T_k$; $M^* = M / \overline{M}$; $T_к$ - volume average boiling point, *K*; $\overline{M}$ - average molar mass equal to 140.

Using equation (17), the thermal conductivity of all studied gas condensates was calculated (in the temperature range 313–433 K). The root mean square deviation for 106 points was about ±2.5%, the maximum did not exceed ±5.0%. The obtained experimental material on thermal conductivity at high pressures made it possible to establish that for the studied gas condensates, within the experimental error, thermal conductivity increases linearly with increasing pressure. Therefore, the thermal conductivity values of the studied gas condensates within the pressure range of 0.1–40.0 MPa can be calculated using the

equation

$$\lambda_{P,T} = \lambda_T + (\frac{d\lambda}{dP})_T (P - P_0), \quad (18)$$

where $\lambda_{P,T}$, λ_T—thermal conductivity at pressure P, atmospheric pressure and temperature T, $W/m\cdot K$; $(d\lambda/dP)_T$—a value characterizing the effect of pressure on thermal conductivity; λt - can be determined from equations (1), (13), (16), (17); $(d\lambda/dP_T)$ - calculated from experimental data for each gas condensate.

Of practical interest is the establishment of an analytical connection between thermophysical properties and the group hydrocarbon composition of the studied products, which would make it possible to calculate the properties of gas condensates from unexplored fields. Previously, the influence of group hydrocarbon composition on the thermal conductivity of the study objects was noted. The content of a relatively large amount of paraffin hydrocarbons in gas condensate causes an increase in thermal conductivity, and the presence of cyclic (aromatic and naphthenic) hydrocarbons reduces it. Analysis of experimental data on the thermal conductivity of the studied products and their group hydrocarbon composition made it possible to establish a relationship between thermal conductivity, density and content of paraffin hydrocarbons in gas condensate. The basis for this was equation (1). When processing experimental data in the high-pressure region, the dependences of the values of B and the exponent on temperature were taken into account. In addition, a connection was established between the value of B and the group hydrocarbon composition of gas condensates and the dependence was obtained

$$\lambda_{P,T} = B(\tau, \Pi)\rho_{P,t}^{n(\tau)} \cdot 10^{-3}, \quad (19)$$

where $B = 0{,}3086 + 0.0347\Pi + 0{,}11\tau$; $n = 4{,}7—2{,}57\ \tau$; $\tau = T / \overline{T}$; $\rho_{P,T}$ – density of gas condensates at different P and T, $kg/m3$; - average boiling point equal to 450 K; Π is the proportion of paraffin hydrocarbons in the gas condensate.

Equation (19) does not require experimental information about the thermal conductivity of the substance under study, which distinguishes it favorably from others. To calculate the density in the high-pressure region in the absence of data, you can use the technique developed in the industrial thermophysical laboratory of the State Research Institute based on the Tate equations, which, as indicated in [5], allows you to calculate the density of gas condensates with a maximum error of ±1.0%. Using equation (19), the thermal conductivity of all studied gas condensates was calculated in the temperature range of 313–473 K and pressures of 0.1–40.0 MPa. The density values of gas condensates are borrowed from [8]. The standard deviation for 96 points is ±1.8%, the maximum does not exceed ±4.0...4.5%.

The results obtained and a comparison of calculated and experimental values of λ allow us to recommend equation (19) for calculating the thermal conductivity of gas condensates containing paraffin hydrocarbons from 30 to 80% in the temperature range of 313-473 K and pressures of 0.1-40, 0 MPa.

In [8], an equation is proposed for calculating the density of gas condensates at various temperatures and pressures

$$\rho = (at + b) \cdot 10^{-3}, \quad (20)$$

where $a = (-7{,}83 + 4{,}79\text{-}10^{-2} P + 0{,}504\cdot 10^{-4} \cdot P^2) \cdot 10^{-4}$, $b = 0{,}793 + 0{,}641\cdot 10^{-3}$, $p—3{,}234 - 10^6 P^2$.

According to the author, equation (20) describes the experimental data with an error of no more than ±1.0 + 2.0% and is valid for gas condensates, the relative density of which is

in the range ρ_4^{20} = 0.76-0.79. The gas condensates of most fields in Uzbekistan are within the same limits ρ_4^{20}. In this regard, of practical interest is the transformation of equation (19) taking into account (20) to the form

$$\lambda_{P,T} = B(\tau, \Pi)\left[a(T-273)+b\right]^{n(\tau)}, \tag{21}$$

To use equation (21), knowledge of the group hydrocarbon composition of gas condensate is sufficient. The equation gives satisfactory results for gas condensates with a content of paraffin hydrocarbons in the range of 30…80% and a relative density ρ_4^{20} = 0.76-0.79 in the temperature range 313-473 K and pressures 0.1-40.0 MPa. Calculation of the thermal conductivity of the studied gas condensates and comparison with experimental data show that the standard deviation for 102 points is equal to ±2.2%, the maximum does not exceed ±5.0%. As a result of processing the thermal conductivity of gas condensates from all studied fields in Central Asia, together with physicochemical characteristics, the equation was obtained

$$\lambda_{P,T} = a_1 \cdot \rho_4^{20} - \alpha\,(T - 293{,}15)\cdot 10^{-3} - a_4\,(P - P_0), \tag{22}$$
$$\alpha = a_2 - a_3\, n_D^{20}$$

where a_1 = 0,1540; a_2 = - 0,0004; a_3 = - 0,1236; a_4 = 0,0004; ρ_4^{20} - relative density; n_D^{20} — refractive index: P_0 - atmospheric pressure, MPa.

Equation (22) is applicable for calculating gas condensates and fractions with a paraffin hydrocarbon content of 10 - 85%. The deviation of the calculated values from the experimental ones does not exceed 4-5% (root mean square) and 5-7% (maximum). An equation of the form (22) with a dependence has the same qualitative results. Equation (22) for the temperature dependence of thermal conductivity (P = P_0) with the corresponding coefficients a1...a3 improves the approximation of calculations and experiment. The maximum deviation does not exceed 3...4%. Based on a comparative analysis of existing equations and methods for calculating the thermal conductivity of complex mixtures of liquid hydrocarbons, as well as on the results of a generalization of experimental data on the thermal conductivity of 8 gas condensate deposits of significant reserves and characteristic composition in Central Asia, recommendations are proposed for calculating λ gas condensates at various temperatures and pressures (Table. 1). The recommendations allow, depending on the initial information about the required limits of temperature and pressure, the required accuracy of thermal conductivity calculations, to select the appropriate method or equation.

Table 1. Recommendations for calculating the thermal conductivity of gas condensates.

Original information	Equation or calculation method λ	Source	Limits of application	Deviation from experiment	
				δ_{max}, %	σ, %
At atmospheric pressure $\lambda\tau$					
λ_i , ρ_T	The equation (1)	[4]	273–433 K	± 4,0	± 2,0
λ_i , T_K	The equation (14)	Автор	0, 6 < τ < 1,1	± 2,0	± 1,0
T_K ,M	The equation (17)	Автор	233…433 K	± 5,0	± 2,5
At elevated pressures $\lambda_{P,T}$					
λ_T , $\rho_{P,T}$	Equation (4) with adjusted exponent (5)	[5]	313 – 493 K; 0,1 – 40,0 MPa	± 5,0	± 2,0

n_D^{20}, ρ_4^{20}, M,T_K	Methodology OTFL GNI	[5]	313 – 493 K; 0,1 – 40,0 МПа	± 7,0	± 3,0
Group hydrocarbon composition	The equation (19)	Author	313 – 493 K; 0,1 – 40,0 MPa П= 0,3 …0,8	± 5,0	± 2,0
Group hydrocarbon composition	The equation (21)	Author	313 – 493 K; 0,1 – 40,0 MPa П= 0,3 …0,8 ρ_4^{20} = 0,76…0,79	± 5,0	± 2,5

Here δ_{max} is the maximum deviation, σ is the standard deviation

3 Conclusion

The possibilities of using existing methods for calculating the thermal conductivity of petroleum products for gas condensates are considered. Analysis of the results of calculating the thermal conductivity of petroleum products using existing equations and comparisons with experimental data show that most of the considered equations do not reflect experimental data with the required accuracy and only some of them can be recommended for calculating the thermal conductivity of gas condensates. To calculate the temperature dependence of the thermal conductivity of gas condensates, equations are obtained that do not require initial experimental information. Root mean square calculation error λ ± 2.5% and ± 1.5%, maximum - ± 5.0% and ± 3.0%. Equations were obtained for calculating the thermal conductivity of gas condensates at elevated pressures and various temperatures depending on their group hydrocarbon composition. The standard deviation of the calculated values from the experimental ones is ± 2.0 ... 2.5%. Recommendations have been developed for calculating the thermal conductivity of gas condensates from new, previously unexplored fields, depending on the availability of initial information, the required temperature range, pressure and the required accuracy.

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