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Predicting the flare temperature of binary mixtures according to data on activity coefficients

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Abstract

A method for calculating the flash point from the results of simulating liquid–solid equilibrium at constant pressure using the Gibbs equation is discussed. A model is used to predict the flash point of the mixture based on the modified Le Chatelier equation, the Antoine equation and a model for estimating the activity coefficient.

The flammability hazard of liquids is primarily characterized by their flash point. The flash point is defined as the temperature at which a liquid evaporates and forms a flammable mixture with air. To measure the flash point, closed and open type devices are used. In closed-type devices, the state of equilibrium between the liquid and vapor components of the mixture is studied. Open type devices take into account the interaction of a mixture of flammable liquids with the atmosphere. The flash point of a mixture is a critical property, but experimental data for many mixtures are lacking and obtaining such data is expensive and time-consuming. Therefore, the development of mathematical models for analyzing the state of the environment under conditions of increasing risk of emergency situations is an important scientific and practical task. This paper examines the possibility of predicting the flash point in closed-type devices, i.e. the influence of atmospheric conditions is not taken into account in this approximation. Several models for predicting the flash point for mixtures of various types have been proposed previously.

Keywords: binary mixtures, flash point, boiling point, activity coefficients, solvation coefficient, association coefficient.

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1. Introduction

Safety in the production and storage of flammable liquids is one of the most important tasks, since dangerous situations often occur, such as explosions at gas stations, accidents during the transportation of flammable substances, etc. When assessing the fire hazard of liquids with a melting point of less than 50°C, a number of indicators of fire and explosion hazard are used: flammability group, flash point, ignition temperature, auto-ignition temperature, lower and upper concentration limits of flame propagation, ability to explode and burn when interacting with water, atmospheric oxygen and others substances, etc. These parameters are interconnected. For example, the flash point is linearly related to the boiling point: fp b t a bt = +, which depends on the ambient pressure. In this regard, the task of assessing the influence of pressure and temperature on the thermodynamic characteristics of mixtures of liquids is relevant. It is especially important to study the behavior at various temperatures, pressure, concentration, thermophysical parameters of the environment, etc. of such flammable liquid mixtures as fuel mixtures, brake fluids, antifreezes used for cooling internal combustion engines, coolants in heating and air conditioning systems. Along with this, it is necessary to solve the problem of heat transfer, which is divided into external or internal depending on the characteristics of the heat source and the shape of the heat accumulator. Thus, predicting the flash point is only part of the problem of studying fire and explosion safety conditions

Models developed for ideal solutions [4–6] are unsuitable for nonideal mixtures, which are most common. The imperfection of solutions is due to the interaction of molecules. The influence of liquid phase non-ideality on activity coefficients is taken into account by constructing appropriate thermodynamic models, which can be divided into two categories: models using experimental data and so-called a priori predictive models. To model the properties of a solution, it is necessary to be able to calculate the activity coefficients of the components of the mixture in the liquid phase.

2. Methodological part

Thermodynamic models NRTL, Wilson and UNIQUAC are often used, but in these models the binary interaction parameters are calculated from experimental data. Therefore, these models are not predictive. The UNIFAC model [12] is predictive, since it does not require experimental data to calculate the parameters of binary interaction. To assess the interaction, group contribution parameters are used [10, 13]. The general model for predicting the flash point of a mixture is known [1] and improved by Liu et al. [2] based on the modified Le Chatelier equation (1), Antoine equation (2) and a model for estimating activity coefficients [3]:

$$\sum_i x_i y_i P_i / P_{i,fr}^0 = 1; \qquad (1)$$

$$log P_i = A - B_i/(T + C_i), \qquad (2),$$

where x_i is the mole fraction of the i-th component in the liquid; γ_i is the activity coefficient of the component; P_i is the saturated vapor pressure of the i-th component of the mixture at a given temperature; P_i, 0 fp is the vapor pressure of pure combustible component i at its flash point, respectively.

The Le Chatelier equation $L = 1/\sum_i p_i/L_i$ is used to determine the explosion limit of a vapor mixture and is a formula for calculating the harmonic mean value, where L is the ignition limit of the mixture; L_i – flammability limits of individual components; pi is the contribution of individual components to the studied property of the liquid mixture. In particular, the equation allows us to determine the average pressure P_{fp} at which a flare can occur, i.e. at $L_i = P_{i,fr}^0$. Unde

the condition of phase equilibrium of liquid and vapor, the partial pressure of the vapor of a component of a nonideal liquid has the form $p_i = x_i\gamma_i P_i$. The activity coefficient is introduced as a characteristic of the imperfection of the mixture. If the activity coefficient is equal to one, this means that the interactions between different or the same molecules are identical and the mixture is in an ideal state; if the activity coefficient is different from unity, the mixture is in a non-ideal state. In the case of an ideal solution, $\gamma_i = 1$, the partial pressure of a component is proportional to its fraction in the solution $p_i = x_i y_i P_i$, which is the formulation of Raoult's law. The Antoine equation is an approximate representation of the Clausius - Clapeyron equation for the equilibrium of the liquid and vapor phases and makes it possible to move from pressure to the temperature characterizing the flash point, and to find the dependence of the flash point on the composition. To calculate the flash point on the basis of equilibrium models, the activity coefficients of the components γ_i are determined for a given solution composition, then the vapor pressure of the mixture components $P_{i.fr}^{0}$ is found using the Antoine equation at the flash point of the pure components. The flash point T_{fp} for the mixture is found by solving Antoine's equation (2) and equation (1) together. Substituting the pressure P_i from the Antoine equation into the Le Chatelier equation makes it possible to calculate the flash point for the entire range of solution compositions, i.e. dependence T(x). The main problem is to determine the activity coefficients of the components of a non-ideal mixture. This problem can be solved if mathematical modeling methods are used.

Thermodynamic models of phase equilibrium of solutions From models of activity coefficients of the liquid phase, two types can be distinguished: 1) models used for non-polar systems, for example, mixtures of hydrocarbons, isomers, and homologues, which include regular solution theory (RST) and Flory-Huggins models; 2) models used for non-polar and polar systems. These models are commonly used to predict liquid phase activity coefficients and include the Van Laar equation, Wilson equation, NRTL, UNIQUAC equation, UNIFAC equation, etc.

Vidal et al. [13] combined the flash point prediction of Liu et al. [2] with the model UNIFAC to predict the minimum for highly nonideal solutions. Gmeling and Rasmussen calculated the flash point of binary systems using the UNIFAC model to estimate activity coefficients [10, 14]. At the minimum flash point of the mixture, the flash point may drop by several degrees, increasing the risk of explosion, which is often accompanied by a positive deviation of the liquid–vapor equilibrium from Raoult’s law [9, 13, 15]. The maximum on the flash point behavior curve is associated with a negative deviation from Raoult’s law, which leads to a decrease in the risk of explosion [16].

In the original UNIFAC model [12], the logarithm of the activity coefficient consists of the combinatorial and residual parts. To improve the performance of the original UNIFAC model predicting liquid–vapor equilibrium (VLE), liquid–liquid equilibrium (LLE) and excess enthalpies, several versions of the model were proposed [17−20].

The main disadvantage of the UNIFAC model and its versions is the need to create a database of group parameters of pure components and interaction parameters. The group contributions of these parameters are systematically improved by the UNIFAC consortium [21]. In [22], the flash point of mixtures is predicted (Liaw and Chiu flash point prediction model [3]); to calculate the behavior of the activity coefficients of the liquid phase, UNIFAC type models are used, which do not require experimental data to find the parameters of the binary system. In Fig. Figure 1 presents experimental data [22], as well as the results of calculating the activity coefficients and flash point temperature for the systems ethanol - butanol-1, ethanol - acetone. Various versions of the UNIFAC model and the NRTL model were used for the ideal ethanol

butanol-1 system (Fig. 1, a) and the ethanol - acetone system with a positive deviation from ideality (Fig. 1, b). The phase behavior of mixtures becomes more complex if there is a large difference in their physical properties, types of polarity, or critical properties. As can be seen in Fig. 1, there is a significant scatter in the results of calculations of activity coefficients and flash point for different models.

One form of the equation relating various parameters of a system to internal energy can be written as

$$dU = TdS - PdV + \sum_{i=1}^{n} \mu_i dN_i, \qquad (3)$$

where U = f(S,V, N_1, N_2,…,N_n) is the internal energy of the phase as a function of entropy, volume and number of moles of components forming the phase; $\mu = {}_i (\partial U/\partial N_i)_{S\,V\,N,j\neq I}$ chemical potential of the i-th component; $T = (\partial U/\partial S)_{V\,N,j\neq i}$ − temperature; P= (∂U/∂V) S,N J≠i − pressure.

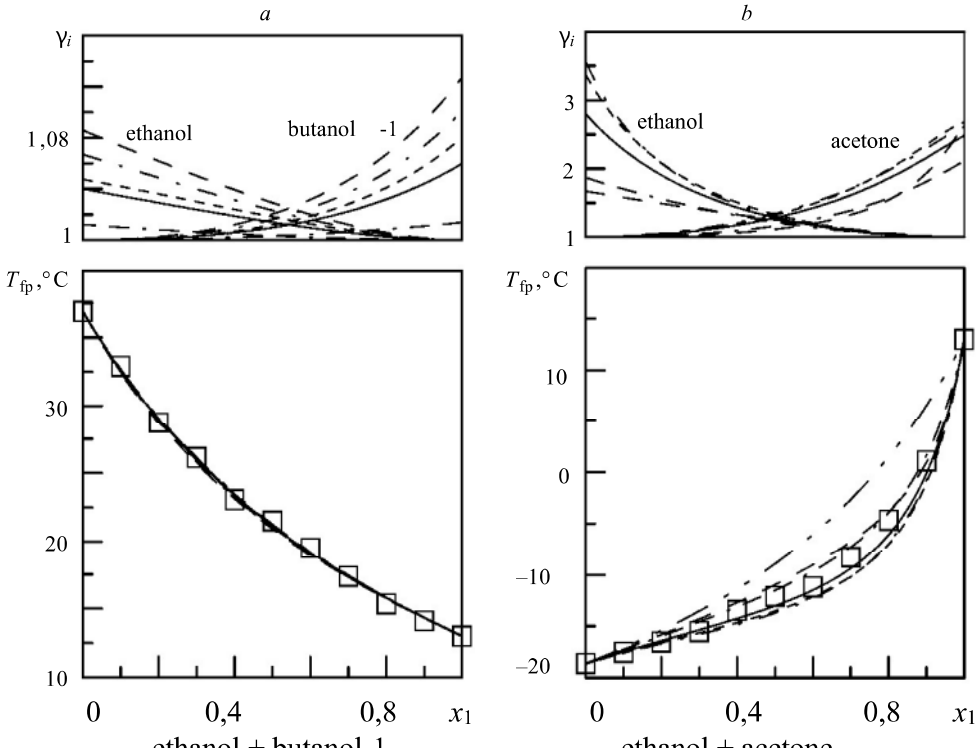

Fig. 1. Comparison of activity coefficients and the predicted flash point with experimental data [22] for an ideal solution ethanol - butanol-1 (a) and a solution with a positive deviation from ideality ethanol - acetone (b): " " - experimental data, " - " - original UNIFAC; "----" − UNIFAC DORTMUND; « ─ · ─ :» − UNIFAC Linghby; "─ ─ ─" – version of UNIFAC Bastos and others; "─ ─ · ─" – NRTL; "─ --" – ideal solution

This paper discusses a method for calculating the flash point based on the results of simulating liquid–solid equilibrium at constant pressure. The relationship between the activity coefficients γ_i and the excess Gibbs free energy GE:

$$RT\sum x_i \ln \gamma = {}_i \; G^E.$$

Equation (3) can be written in another form using the Gibbs energy GE (P,T,x_1,x_2,.., x_n) as the characteristic function:

$$dG^E = -SdT + VdP + \sum^n_{i=1} \mu_i \, dx_i.$$

The equation can also be written as

$$-SdT + VdT - \sum^n_{i=1} x_i \, d\mu_i = 0,$$

where the transition is made from the number of moles of components to their molar fractions x_i. The difference between the equations of state of a binary system for the real and ideal equilibrium phases can be represented as [23]

$$-(H^E/RT^2)\, dT + (V^E/RT)\, dP = \sum^2_{i=1} x_i \, d\ln \gamma_i,$$

where γ_i is the activity coefficient of the component, i = 1, 2; H^E – enthalpy of mixing; V^E – excess volume; P – solution pressure; T – absolute temperature; R is the universal gas constant.

The Gibbs equation is used to represent phase equilibrium over a wide range of temperatures and pressures, and to calculate thermal and volumetric properties.

Thus, equation (4) allows us to find the activity coefficients of the components if the enthalpy of mixing and excess volume are known. If the formation of associates of molecules of pure components occurs, then the effective molar mass of the component in the solution can be calculated using the formula M' $\lambda_i M_i$, where M_i is the molar mass of the component before mixing, λ_i are correction factors. The average ratio of the number of molecules in associates of pure components $\lambda = \lambda_1/\lambda_2$ characterizes the stable structure of the solution. The difference between the coefficient λ and unity indicates the presence of a deviation from ideality in the binary system and the need to move to effective mole fractions to obtain thermodynamically consistent models.

In [24 − 27], a method for modeling phase equilibrium diagrams of liquid–solid and liquid–vapor in real solutions and the possibility of predicting the flash point of a binary liquid mixture is considered.

In most real solutions, the components interact, leading to the formation of molecular compounds of the AB type. The solvation coefficient $\lambda = \lambda_1/\lambda_2$ shows the ratio of the number of molecules A to the number of molecules B in the resulting molecular compound. Molecules of components A and B can also form clusters consisting of molecules of the same type. The ratio of the number of molecules of component A to the number of molecules of component B united into associates will be characterized by the association coefficient $k = k_1/k_2$. The association coefficient k1 shows how many molecules of component A in the liquid phase have combined into a cluster of type AA, similarly, the association coefficient k_2 shows how many molecules of component B in the liquid phase have combined into a cluster of type BB. Minimizing the excess Gibbs energy with respect to the solvation parameter λ leads to an equation that models the liquid–solid phase equilibrium diagram at P = const:

$$T(z_1) = [H_1^E z_1 + H_2^E(1 - z_1)]/[(H_1^E/T_1^0)\, z_1 + (H_2^E/T_2^0)(1 - z_1) -$$

$$- R(z_1 \ln z_1 + (1 - z_1) \ln(1 - z_1))], \qquad (5)$$

where T is the liquidus temperature; H_i^E – enthalpy and T_i^0 – melting temperature of the component forming the one-component phase, i = 1, 2; $z_1 = x_1/(x_1 + \lambda x_2)$, $z_2 = x_1/(x_1/\lambda + x_2)$ are the effective mole fractions of the components of the binary mixture.

The PCEAS (Phase Chart Eutectic and Azeotropic Systems) model proposed by the authors of this work [28] is based on minimizing the excess Gibbs energy using the solvation parameter λ, which characterizes the ratio of the number of molecules A to the number of molecules B in a molecular compound. Equation (5) allows us to find the dependence of the temperature of the

mixture on the composition at constant pressure. The PCEAS program makes it possible to calculate the liquid-solid equilibrium, as well as the liquid-vapor equilibrium at constant pressure or at constant temperature. The input data is the temperature T_i and the enthalpy of phase transitions H_i^0 of pure components. The program allows you to determine the average values of the solvation coefficient λ, as well as the association coefficient k, and calculate activity coefficients.

Phase equilibria liquid - solid and liquid - vapor in binary systems, as well as the dependence of the flash point on the composition of the solution were calculated at atmospheric pressure.

3. Results and discussion

Figure 2 presents the results of calculating the flash point temperature using the PCEAS model for the ideal ethanol - butanol-1 system (Fig. 2, a) and the ethanol - acetone system with a positive deviation from ideality (Fig. 2, b).

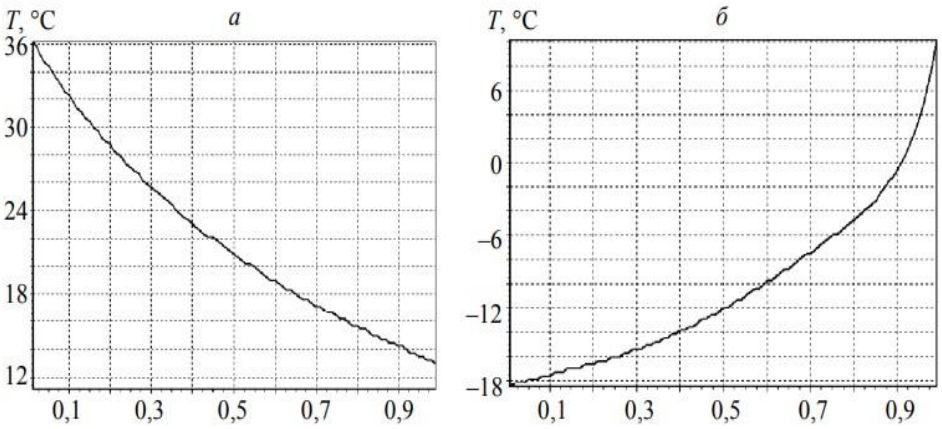

Fig. 2. Dependence of the flash point temperature on the composition of the solution according to the PCEAS model: a – for the ethanol – butanol-1 system; b – for the ethanol – acetone system

In Fig. Table 3 shows the results of calculating activity coefficients using the PCEAS model for the systems ethanol - butanol-1 and ethanol - acetone at normal atmospheric pressure. To predict the flash point of mixtures close to ideal, it is sufficient to use the activity coefficients of the liquid phase obtained from the results of modeling the liquid–solid equilibrium. The work [22] presents modeling results and experimental curves of the dependence of the flash point on the composition for systems with the formation of an extremum of the flash point. It is noted that existing thermodynamic models do not accurately describe such systems. The reason for this is insufficient attention to the processes of solvation and association in the liquid and vapor phases, as well as the consistency of thermodynamic data.

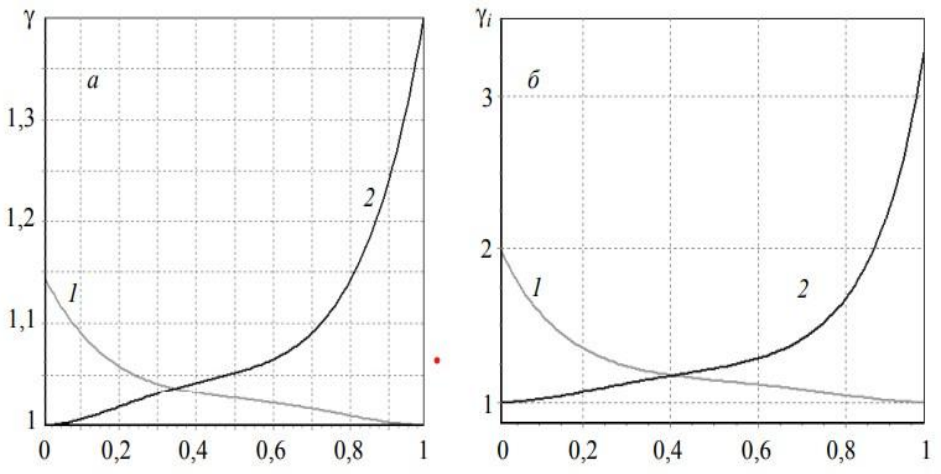

Fig. 3. Calculation of activity coefficients at P = 101.4 kPa using the PCEAS model for solutions: a – ethanol – butanol-1: cr. 1 – activity coefficient (ethanol), cr. 2 – activity coefficient (butanol-1); b – ethanol – acetone with a positive deviation from ideality, line 1 – activity coefficient (ethanol), line 2 – activity coefficient (acetone)

In Fig. 4, 5 show the calculation results and experimental data for systems with the formation of a minimum flash point [22].

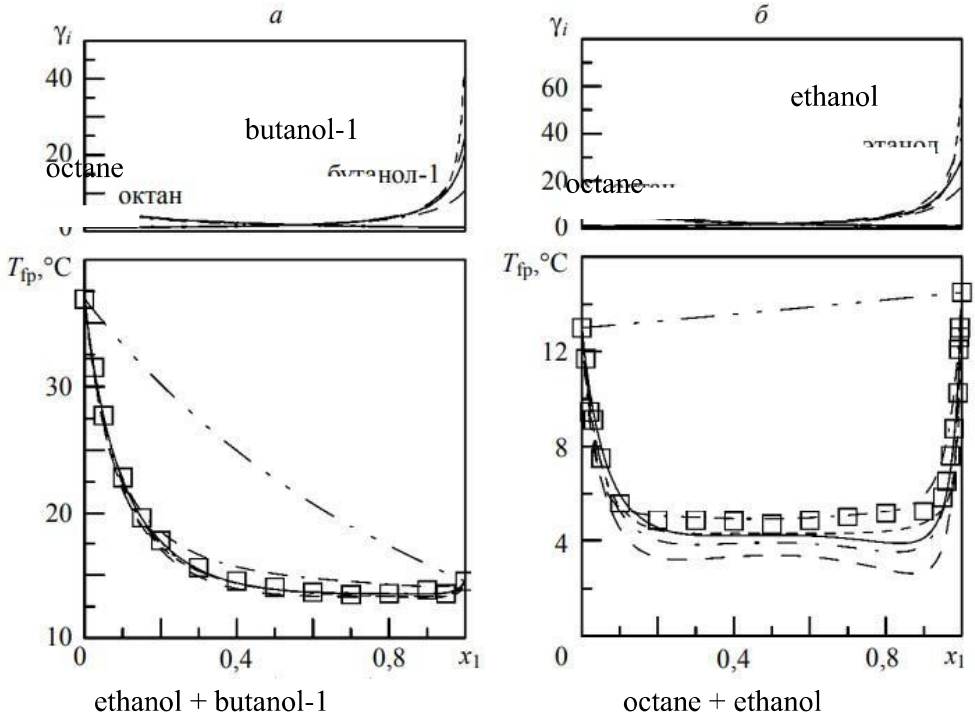

Fig. 4. Comparison of activity coefficients and predicted flash point with experimental data [3] for solutions with the formation of a minimum flash point octane - butanol-1 (a) and octane - ethanol (b): " " - experimental data, " - " - original UNIFAC; "----"– UNIFAC DORTMUND; « ─ · ─ :» – UNIFAC Linghby; “─ ─ ─” – version of UNIFAC Bastos and others; “─ ─ · ─” – NRTL; “─ --” – ideal solution

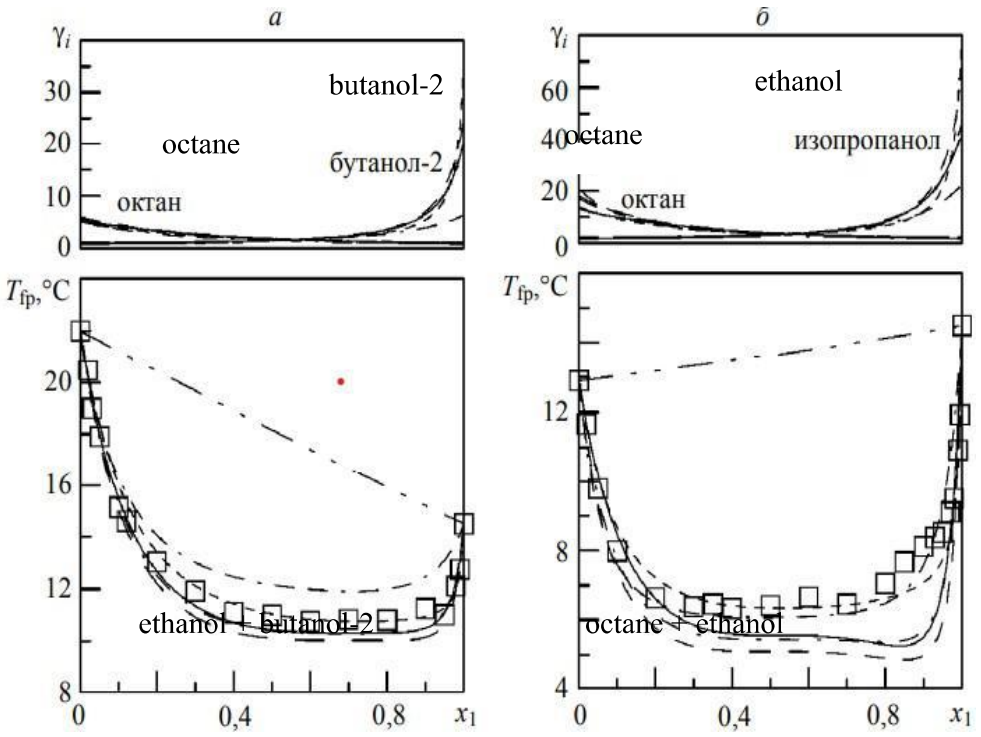

Fig. 5. Comparison of activity coefficients and predicted flash point with experimental data [22] for solutions with the formation of a minimum flash point octane - butanol-2 (a) and octane - isopropanol (b): " " - experimental data, " - " - original UNIFAC; “----” – UNIFAC DORTMUND; « — · — :» — UNIFAC Linghby; “— — —” – version of UNIFAC Bastos and others; “— — —” – NRTL; “— --” – ideal solution

In Fig. 6. The results of calculating the boiling point and flash point temperature using the PCEAS model for an octane - butanol-1 solution with the formation of flash point extrema are presented.
In Fig. 7 – results of calculating the boiling point and flash point using the PCEAS model for the octane-ethanol system at normal atmospheric pressure.
In Fig. 8 – results of calculating the boiling point and flash point using the PCEAS model for the octane - butanol -2 system at normal atmospheric pressure.
In Fig. 9 – results of calculating the boiling point and flash point using the PCEAS model for the octane - isopropanol system at normal atmospheric pressure.

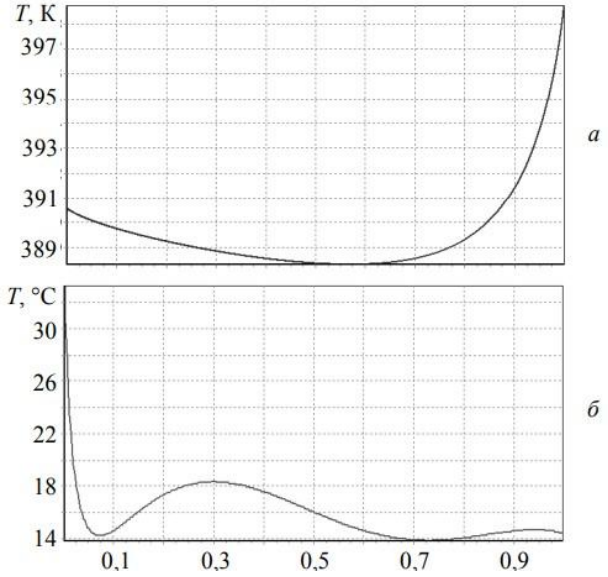

Fig. 6. Calculation of the boiling point and flash point temperature using the PCEAS model for a solution of octane-butanol-1 with the formation of flash point extrema: a – boiling point - calculation; b – flash point temperature – calculation

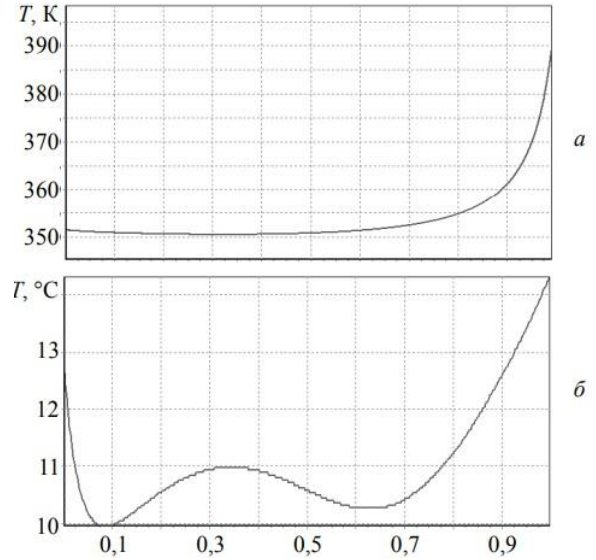

Fig. 7. Calculation of the boiling point and flash point temperature using the PCEAS model for an octane-ethanol solution with the formation of flash point extrema: a – boiling point - calculation; b – flash point temperature – calculation

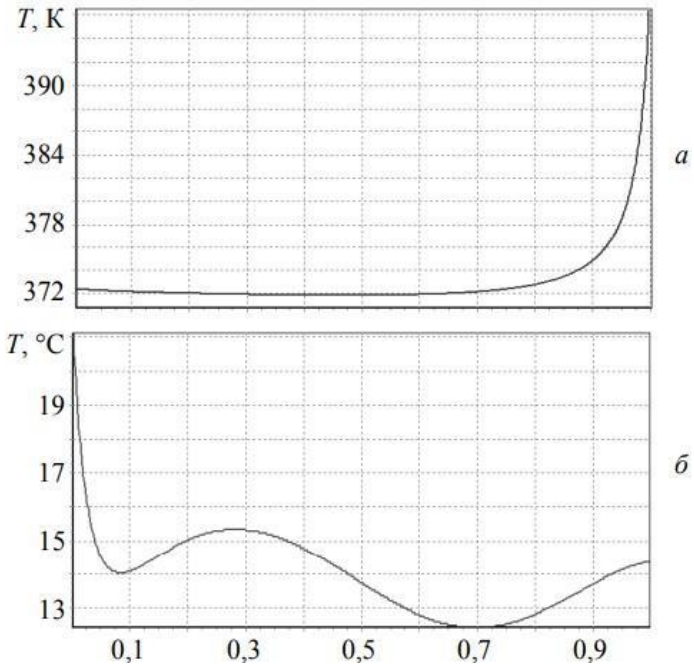

Fig. 8. Calculation of the boiling point and flash point temperature using the PCEAS model for a solution of octane - butanol-2 with the formation of flash point extremes: a - boiling point - calculation; b – flash point temperature – calculation

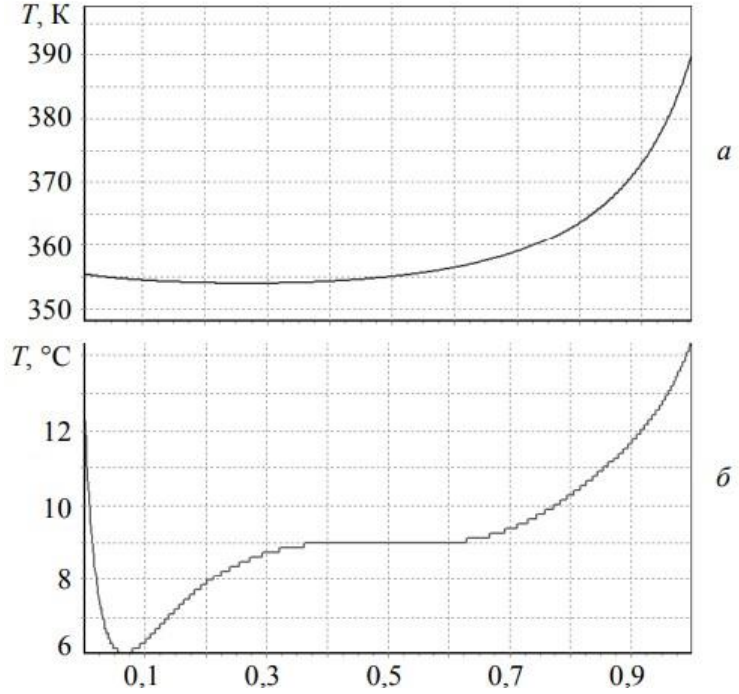

Fig. 9. Calculation of the boiling point and flash point temperature using the PCEAS model for an octane - isopropanol solution with the formation of a minimum flash point: a - boiling point - calculation; b – flash point temperature – calculation

4. Conclusion

A method for predicting the dependence of the flash point of binary flammable liquid mixtures on the composition of the solution is presented. The method is based on the use of mixture pressure data obtained using mathematical modeling of phase equilibrium under isobaric conditions. Predicting the flash point of a binary mixture requires data on the enthalpy and melting point, enthalpy and boiling point, and flash point of the pure components

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research

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Study of the designs of devices for centrifugal extraction

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Abstract

This article provides an overview of centrifugal extractors of various configurations, their advantages are noted advantages and disadvantages. It is noted that, despite the variety of types of