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ON THE CONDENSATION OF MULTICOMPONENT VAPOURS IN THE PRESENCE OF INERT GASES†

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Abstract—The paper deals with the analysis of simultaneous heat and mass transfer at the condensation of multicomponent vapour in the presence of inert components. The results of numerical calculations of condensers, based on the solution of the individual versions of process model, resulting from the different description of multicomponent mass transfer, have been compared with the experimental data available. It has been found that best agreement was obtained in the case of variant based on Krishna–Standart method, whereas the shortest computing time was needed for the variant based on Burghardt–Krupiczka method yielding the results differing not significantly from the most accurate ones.

NOMENCLATURE

A_{ij},	coefficients of the inverted matrix of diffusion coefficients in a multicomponent mixture;
A	inverted matrix of diffusion coefficients in a multicomponent mixture;
c_p,	specific heat at constant pressure;
D_{ij},	binary coefficient of diffusion of component i through j;
D, D',	matrix of diffusion coefficients in multicomponent mixture;
F	geometric surface area of wall on the vapour–gas side;
G,	mass flow rate;
J_i^*,	diffusional mass flux of component i with respect to the molal average velocity of mixture;
J^*,	column vector of diffusional mass fluxes;
N_i,	total mass flux of component i;
N,	column vector of total mass fluxes;
p_i,	saturation vapour pressure of component i;
P,	total pressure;
P,	bootstrap solution matrix;
P_{ij},	elements of bootstrap solution matrix;
q,	heat flux per unit area;
q_t,	gas-interface heat flux per unit area of surface F;
q_w,	interface-coolant heat flux per unit area of surface F;
r,	latent heat of vaporization;
t,	temperature;
x_i,	mole fraction of component i in liquid phase;
y_i,	mole fraction of component i in gaseous phase;
y,	column vector of mole fractions;
$\tilde{y}_i$,	arithmetic-mean of concentrations of component i in diffusional layer;
$\tilde{y}_I$,	arithmetic-mean of total concentrations of inert components in diffusional layer;
y_{Im},	integral-mean of total concentrations of inert components in diffusional layer.

Greek symbols

α,	local heat-transfer coefficient;
α^*,	corrected heat-transfer coefficient;
α_0,	combined heat-transfer coefficient;
β_{ij}^*,	corrected mass-transfer coefficient in a binary system i, j;
$\beta^*(D), \beta(D')$,	matrix of mass-transfer coefficients;
γ,	liquid-phase activity coefficient;
δ_{ij},	Kronecker's delta;
ϕ,	fugacity coefficient;
φ,	coefficient defined by equation (4b);
θ_h,	correction factor accounting for the effect of finite fluxes N_i on α value;
θ,	correction factor accounting for the effect of finite fluxes N_i.

Subscripts

c,	refers to liquid phase;
g,	refers to gaseous phase;
i,j,	refer to mixture components;
m,	mean value;
w,	refers to coolant;
z,	refers to conditions at gas–liquid interface.

1. INTRODUCTION

CONDENSATION of mixed vapours in the presence of inert gases occurs very often in technological processes. Because of simultaneous momentum, heat and mass transfer, the exact quantitative approach is

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rather difficult, particularly for systems most often occurring in practice, i.e. multicomponent ones.

So, in spite of the fact that the fundamental laws of condensation process have been already formulated in the 1930s by Othmer, Colburn, Hougen and Drew [1–3], the majority of studies carried out in the past concerned the simplest case of condensation in the presence of inerts, i.e. the condensation of a single vapour. An exact and general mathematical model of this case, enabling the design of condensers, has been worked out only two years ago [4, 5].

The problem of condensation of multicomponent vapours in the presence of inert gases has been subject of a few works only. Among them, two principal groups may be distinguished, namely:

1. The group of works [6–8], which apart from the suggestion of methods of the design of condensers, basing on larger or smaller number of simplifying assumptions (constancy of physico-chemical properties, absence of interaction between diffusing components, use of mean driving forces and of mean flowrates, restriction to the systems: binary vapour plus inert gas), present the results of experimental studies carried out on purpose to verify the method.
2. Group of works [9–13], which propose more general methods, basing on a smaller number of assumptions as above; if some calculational results, if any, are given, they do not refer to any experimental results.

Among the works of the latter group, the most general and exact model of the condensation process of multicomponent vapours in the presence of inert components is given by Krishna *et al.* [11]. Here, as well as in our own work [14], one has rejected for the first time one simplifying assumption, introduced formerly at the description of mass transport. One has used, namely, general matrix equations which in a physically correct manner predict mass fluxes in multicomponent systems. Krishna *et al.* employed the equations derived [16] by the exact solution (at the assumption of the film theory) of Stefan–Maxwell equations and, alternatively, the method worked out by Toor [17] and independently by Stewart and Prober [18].

In [14], while formulating the general mathematical model of the process, one has proposed the direct calculation of mass fluxes from the relationship worked out by Burghardt and Krupiczka [19], which enables to dispense with the iterative procedure. The proposed model has been next solved [15] for the experimental condensers of Potter and Jeffreys [6], Mizushina [7] and Schrodt–Gerhard [8] and the results of calculations have been compared with the experimental ones. It came out — as expected — that the suggested model gave the results most close to the experimental data, compared to the simplified methods ([7, 10]).

The aim of the present work was the application to the general mathematical model of multicomponent condensation in the presence of inerts, formulated in [14], of the remaining two methods of the description of mass transport in multicomponent systems (Toor and Stewart and Prober; Krishna and Standart), its solution for the case of availability of experimental data, followed by the comparison of all results obtained at present and previously (computational as well as experimental ones).

The model formulated in [14], discussed in short below, is — like the models of Schrodt and of Krishna *et al.* — a one-dimensional model. Such an approach has been determined by the fact that at the solution of a model for the turbulent flow it is necessary to make use either of turbulence models, which render the problem very complex and are not reliable for all geometries, or of turbulence parameters determined experimentally. Thus, both in the case of two-dimensional model and one-dimensional one, the use of empirical parameters is inevitable. The assumption *a priori* which of these models will yield more accurate results is difficult, since the final result will depend not only on model's exactness but also on errors due to empirical parameters. It is certain, however, that the two-dimensional model will be more sophisticated and its solution will be connected with considerable computational difficulties and will require larger time of computation as well as digital computers of higher speed and larger memory. The proposed one-dimensional model is not, however, quite general, since it does not allow for fog-containing vapour mixtures. Such mixtures had to be disregarded of necessity, since in the case of multicomponent vapours no criterion, even of estimative character, has been suggested which would enable the determination both of the extent of its precipitation and of composition. This model neither takes into account the diffusional resistance in the liquid phase (it is assumed to be negligible [11]).

2. ANALYSIS OF SIMULTANEOUS HEAT AND MASS TRANSFER IN MULTICOMPONENT SYSTEMS

The simultaneous interphase heat and mass transport in the case of finite mass fluxes, causing a disturbance of temperature and concentration profiles in the boundary layer, may be most generally described by equations (1) and (2)[21]

$$q = \alpha^* \Delta t \tag{1}$$

$$N_i - y_i \sum_{j=1}^{n} N_j = J_i^* = \sum_{j=1} \beta_{ij}^* \Delta y_j \tag{2a}$$

$$i, j = 1, 2, \ldots, n$$

or — in matrix notation

$$N - y \sum_{j=1}^{n} N_j = J^* = \beta^*(D) \Delta y. \tag{2b}$$

Relationship (1) resembles Newton's equation with heat transfer-coefficient α replaced by the coefficient α^*, allowing for the effect of finite mass fluxes on the temperature profile

$$\alpha^* = \alpha \theta_h. \tag{3}$$

The form of the correction factor θ_h in equation (3) depends on the assumed transport theory. For example, at the assumption of film theory it may be determined for a multicomponent system from the relationship

$$\theta_h = \frac{\varphi}{\exp\varphi - 1} \tag{4a}$$

where

$$\varphi = \frac{\sum_{i=1}^{n} N_i c_{pi}}{\alpha}. \tag{4b}$$

Expression $\beta^*(D)$ in equation (2b) represents the matrix of mass-transfer coefficients, whereas D is the matrix of diffusivities. There exist, as already mentioned, two methods of determining the matrix of mass-transfer coefficients: method of Toor and Stewart and Prober [17, 18] and method of Krishna and Standart [16]. The main difference between these methods consists in the assumption, made by the former authors at the linearization of the system of equations, of the constancy of diffusivities' matrix along the path of diffusion, whereas Krishna and Standart take into account the concentration profile in the film. So, from the theoretical view-point, the solution of the latter authors is more exact. On the other hand, there is no possibility in this case to use correction factors θ according to various transport theories (as it may be done in the solution of Toor and Stewart and Prober), since it has been derived only for the film theory assumptions.

The determination of the fluxes N_i from equation (2b), if the J_i^* values are known, does not present any serious difficulties. For the case under consideration, these fluxes may be calculated from the relationship (5)

$$N = P \cdot J^* \tag{5}$$

where P is the matrix consisting of the following elements

$$P_{ij} = \delta_{ij} + y_i/y_I \qquad i,j = 1,2,\ldots,m \tag{6}$$

δ_{ij} is Kronecker's delta, and y_I—sum of concentrations of inert components.

At the determination of the fluxes N_i by either of both methods mentioned above, while calculating the matrix $\beta^*(D)$ one has to assume initially the values of mass fluxes and then to correct it iteratively. This procedure, however, is rapidly convergent.

On the other hand, the method of Burghardt and Krupiczka [19, 20] permits to calculate directly the mass fluxes, which reduces the computational time considerably. The latter authors suggest the calculation of fluxes from equation (7)

$$N = \beta(D')\frac{\Delta y}{y_{Im}}. \tag{7}$$

The matrix of mass-transfer coefficients $\beta(D')$ is determined analogically as the matrix $\beta^*(D)$ in the method of Toor and Stewart and Prober, matrix D' being, however, defined differently than matrix D:

$$D' = A^{-1} \tag{8a}$$

where

$$A_{ij} = \frac{1}{\bar{y}_I} \sum_{\substack{k=1 \\ k \neq i}}^{n} \frac{\bar{y}_k}{D_{ik}} \qquad i = 1,2,\ldots,m \tag{8b}$$

$$A_{ij} = -\frac{\bar{y}_i}{\bar{y}_I}\frac{1}{D_{ij}} \qquad \begin{matrix} i,\ j = 1,2,\ldots,m. \\ i \neq j \end{matrix} \tag{8c}$$

This solution has been derived from the system of Stefan–Maxwell equations under the assumption of film theory, similarly as Krishna and Standart did. It does not take into account, however, the concentration profiles in the film. Moreover, the method of approach to the solution of this problem, used by the before mentioned authors, differs considerably from the method of Krishna and Standart. The value of the integral mean y_{Im} may be calculated—according to authors' suggestion—as the logarithmic mean, without any appreciable error.

The above methods deal with mass fluxes determined solely by concentration gradients. Since the magnitudes of mass fluxes caused by other factors, e.g. by temperature or pressure, are in the case of condensation negligibly small compared to mass fluxes referring to common diffusion [21], one has concluded to disregard all remaining kinds of diffusion in the description of mass phenomena of the process under consideration.

3. DIFFERENTIAL EQUATIONS OF ENERGY AND MASS BALANCES OF THE PROCESS AND EQUILIBRIUM RELATIONSHIPS

Let us consider the differential element of a condenser in Fig. 1. A mixture of vapours of m components together with n–m inert components is flowing along condenser's wall. The condensate formed, consisting in the most general case of m components, flows down the wall in the same direction as vapour–gas mixture. On the other side of the wall the cooling agent is flowing in co- or counter-current to the vapour–gas mixture. The mass balance of individual components in the gas- and liquid phase and the energy balance of the gas phase and of cooling agent, assuming that vapour–gas mixture is monophasic (absence of fog) and the process takes place without crossing the saturation line, yield the following system of differential equations.

$$\frac{\mathrm{d}y_i}{\mathrm{d}F} = \frac{1}{G_g}\left[y_i \sum_{j=1}^{m} N_j - N_i\right] \tag{9}$$

$$i = 1,2,\ldots,m$$

$$\frac{\mathrm{d}x_i}{\mathrm{d}F} = \frac{1}{G_c}\left[N_i - x_i \sum_{j=1}^{m} N_j\right] \tag{10}$$

$$i = 1,2,\ldots,m$$

$$\frac{\mathrm{d}G_g}{\mathrm{d}F} = -\frac{\mathrm{d}G_c}{\mathrm{d}F} = -\sum_{i=1}^{m} N_i \tag{11}$$

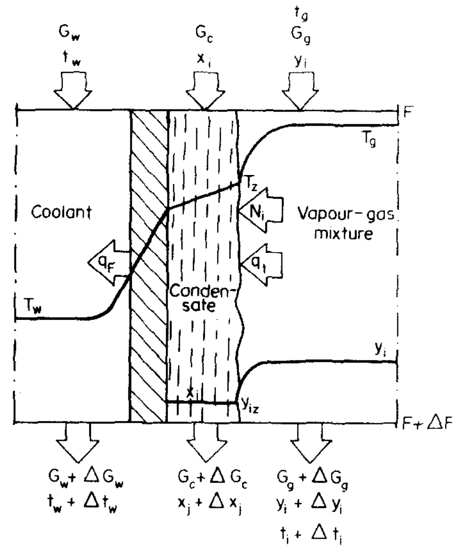


Fig. 1. Process scheme for parallel flow condenser.

$$\frac{\mathrm{d}t_g}{\mathrm{d}F} = -\frac{1}{G_g c_{pmg}} \alpha_g^*(t_g - t_z) \tag{12}$$

$$\frac{\mathrm{d}t_w}{\mathrm{d}F} = \pm\frac{1}{G_w c_{pw}} \alpha_0(t_z - t_w). \tag{13}$$

In energy balances only the enthalpies of fluids and heat fluxes transferred by convection or conduction have been taken into account. All other energy kinds have been disregarded.

If one assumes the thermodynamic equilibrium at the interface, then the interphase equilibrium may be most generally described by the relationship

$$\phi_i y_i P = p_i \gamma_i x_i \qquad i = 1, 2, \ldots, m. \tag{14}$$

This equation enables to determine $m-1$ concentrations y_i at the interface in equilibrium with the liquid phase of the composition x_i [22]. On the other hand, the condition of thermal equilibrium at the interface may be expressed as

Table 1. Results of statistical computations for the mole fractions of ethanol in the condensate, calculated according to various versions of the proposed model and simplified Schrodt's model, compared with the experimental data of Porter and Jeffreys (11 experimental data)

	Toor and Stewart–Prober (1)	Krishna–Standart (2)	Burghardt–Krupiczka (3)	Schrodt
Mean	0.9260	0.9322	0.9301	0.9232
Variance	0.0029	0.0032	0.0025	0.0010
Standard error of variance	0.0162	0.0171	0.0150	0.0098
Standard deviation	0.0536	0.0567	0.0497	0.0324
t	4.5796	3.9685	4.6696	7.8628
Result of t-test	positive	positive	positive	negative

Table 2. Results of statistical computations for the value of surface area of Mizushina's experimental condenser, compared with the values obtained on the basis of calculations according to various versions of the proposed model, simplified Schrodt's model and three-point Mizushina's method (23 experimental data)

	Toor and Stewart–Prober (1)	Krishna Standart (2)	Burghardt–Krupiczka (3)	Schrodt	Mizushina
Mean	0.1253	0.1253	0.1223	0.1172	0.1440
Variance	0.0001	0.0001	0.0001	0.0001	0.0001
Standard error of variance	0.0017	0.0017	0.0020	0.0023	0.0016
Standard deviation	0.0082	0.0080	0.0096	0.0109	0.0075
t	1.5992	1.5992	2.8141	4.7674	10.1645
Result of t-test	positive	positive	positive	negative	negative

Table 3. Results of statistical computations for the amount of condensate formed, calculated according to various versions of the proposed model and simplified Schrodt's model, compared with the experimental data of Schrodt–Gerhard (7 experimental data)

	Toor and Stewart–Prober (1)	Krishna Standart (2)	Burghardt–Krupiczka (3)	Schrodt
Mean	1.2306	1.2339	1.2364	1.2589
Variance	0.0214	0.0224	0.0215	0.0218
Standard error of variance	0.0553	0.0566	0.0554	0.0559
Standard deviation	0.1462	0.1497	0.1466	0.1478
t	4.1726	4.1359	4.2666	4.6361
Result of t-test	positive	positive	positive	negative

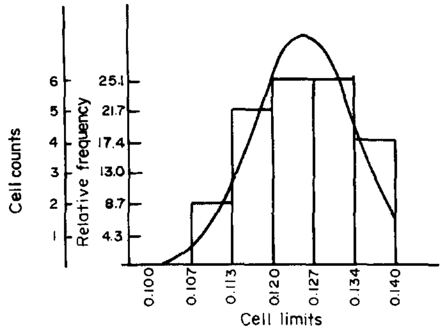
$$q_w = q_t + \sum_{i=1}^{m} N_i r_{iz}. \qquad (15)$$

This nonlinear equation permits to calculate the unknown interfacial temperature by one of iterative procedures [23].

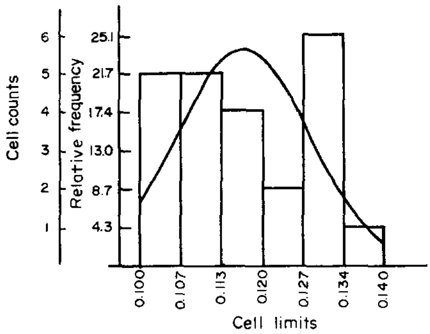
4. RESULTS OF COMPARATIVE CALCULATIONS

The differential equations of energy and mass balances (5)–(13), together with equations (14), (15) determining the interfacial equilibrium, relationships (1)–(8) describing the transport of heat and mass as well as with equations of the kinetics of heat and mass transfer, constitute a system of equations which describe the process of simultaneous heat and mass transfer at the condensation of multicomponent vapours in the presence of inerts. This system has been solved numerically for the experimental condensers of: Potter and Jeffreys, Mizushina and Schrodt and Gerhard, applying for the assumptions of the film theory the above-discussed versions of the description of mass transport and assuming that the diffusional resistance in ideally mixed liquid phase may be disregarded [11].

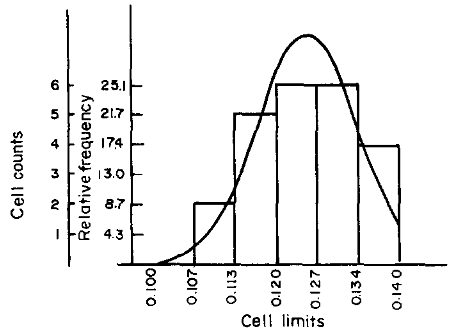
The adequacy of the individual versions of the proposed model and of Schrodt's model has been estimated by means of statistical methods. In the case of Porter and Jeffreys' data the estimation basis was the random variable equal to the ratio of the value of mole fraction of ethanol, determined for the individual sections of the condenser from the solution of individual models, to the experimental value. The null hypothesis $H_0{:}\mu = 1$ has been verified by means of Student's t test. The results of statistical calculations are given in Table 1. For the statistical estimation of the considered models on the basis of experimental



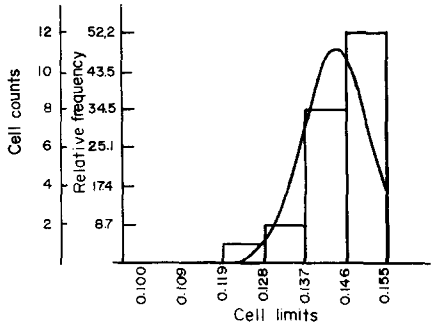
Toor and Stewart–Prober version



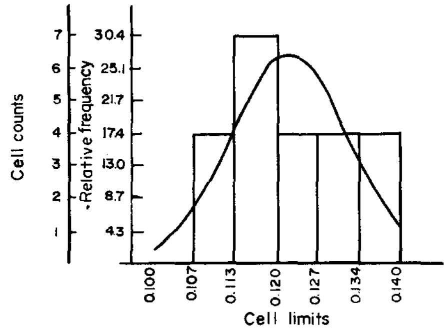
Schrodt 's model



Krishna-Standart version



Mizushina's method



Burghardt–Krupiczka version

FIG. 2. Histograms of distribution of random variable obtained from the individual versions of the proposed model, Schrodt's model and Mizushina's three-point method, on the basis of Mizushina's experimental data.

data of Mizushina *et al.* one has assumed directly the magnitude of the computed surface area, necessary to attain the degree of condensation equal to the experimental value. Also the three-point method, proposed by Mizushina, has been subject to estimation. The value of the area of the experimental condenser was the basis for the formulation of null hypothesis $H_0:\mu=0.128$. Table 2 contains the calculational results. In order to carry out the estimation of the models under consideration on the basis of Schrodt–Gerhard's data, one has calculated the amount of condensed vapours. The random variable and the null hypothesis have been defined analogically as in the case of Porter and Jeffreys' data. The results of statistical calculations are shown in Table 3.

Analysing the results in Tables 1 and 2 one can find that at the significance level 0.001 the null hypothesis is true only for the versions of the proposed model. Analogical conclusion may be drawn in the case of comparison with Schrodt–Gerhard's experimental data, but assuming a slightly higher significance level.

The lowest value of test t, i.e. the smallest probability that the value obtained from the model will differ significantly from the experimental value was yielded by the version of calculations according to the method of Krishna and Standart. In order to ascertain, if the differences in computational results between individual versions of the proposed model are significant, the analysis of variance and estimation by means of F test has been carried out. Under the assumption of the significance level 0.05 it has been found that the results obtained from individual versions do not differ significantly.

The histograms presented in Fig. 2 show the distribution of the random variable obtained on the basis of Mizushina's experimental data. These histograms illustrate the correctness of the last statement.

Recapitulating, it may be concluded that adequacy of all three versions of the proposed model (1), (2) and (3) is nearly the same. The most accurate version proved to be one based on Krishna and Standart's method but the difference with respect to the remaining two versions is rather insignificant. Hence, the choice of some version or other should be additionally determined by the corresponding time consumption of computer. In this respect the most rapid version of the proposed model is, of course, the non-iterative method of Burghardt and Krupiczka, whereas the slowest one (*ca* 3 times slower) is the version based on the method of Krishna and Standart.

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SUR LA CONDENSATION DE MELANGES DE VAPEURS EN PRESENCE DE GAZ INERTES

Résumé—On analyse les transferts simultanés de chaleur et de masse pour la condensation de mélanges de vapeurs en présence de gaz inertes. Les résultats des calculs numériques des condenseurs, basés sur des versions individuelles de modèle de mécanisme et résultant de la description différente du transfert massique ont été comparés avec les données expérimentales connues. On trouve que le meilleur accord est obtenu dans le cas d'une variante de la méthode de Krishna–Standart, tandis que le plus court temps de dalcul est obtenu par une variante de la méthode de Burghardt–Krupiezka qui donne des résultats ne différant pas très sensiblement des plus précis.

ÜBER DIE KONDENSATION VON MEHRSTOFFDAMPFGEMISCHEN BEI ANWESENHEIT VON INERTGASEN

Zusammenfassung — Der Bericht befaßt sich mit der Analyse von gleichzeitigem Wärme- und Stoffübergang bei der Kondensation von Mehrstoffdampfgemischen in Anwesenheit von inerten Komponenten. Die Ergebnisse der numerischen Berechnung von Kondensatoren, die auf der Lösung der einzelnen Modellversionen basieren, welche sich aus den verschiedenen Beschreibungen des Stoffübergangs von Mehrstoffsystemen ergaben, wurden mit den vorhandenen experimentell ermittelten Werten verglichen. Es wurde festgestellt, daß die beste Übereinstimmung mit der Variante erhalten wurde, die auf der Krishna–Standart-Methode basierte, während die kürzeste Rechenzeit für die Variante nach der Burghardt–Krupiczka-Methode benötigt wurde, wobei sich die hierbei erhaltenen Ergebnisse nicht signifikant von denen der genauesten Berechnungen unterschieden.

О КОНДЕНСАЦИИ МНОГОКОМПОНЕНТНЫХ ПАРОВ В ПРИСУТСТВИИ ИНЕРТНЫХ ГАЗОВ

Аннотация — Проведен анализ одновременного тепло- и массопереноса при конденсации многокомпонентного пара в присутствии инертных компонентов. Результаты численных расчетов конденсаторов, основанных на решении различных вариантов модели процесса, обязанных различной интерпретации многокомпонентного переноса массы, сравнивались с имеющимися экспериментальными данными. Наилучшее совпадение результатов получено для варианта, основанного на методе Кришны–Штандарта, в то время как вариант, основанный на методе Бургардта–Крупички, потребовал самого минимального количества машинного времени и дал результаты, довольно близко совпадающие с точными данными.