Numerical calculation of the diffusion process in multicomponent hydrocarbon gas mixtures

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Abstract—Knowledge of the laws of diffusion is necessary in the description, design and calculation of the mass transfer process in the production, operation and transportation of gaseous fuels. In this article, the calculation of diffusion processes for five natural hydrocarbon gas mixtures into the air was carried out. The effective diffusion coefficients and matrix coefficients of multicomponent diffusion were determined. Also the advantages of using effective coefficients in the description of mass transfer were shown.

Keywords—Diffusion, effective diffusion coefficients, hydrocarbon, mass transfer.

I. INTRODUCTION

TODAY, natural gas is one of the cheapest and most environmentally friendly fuels. It also serves as a raw material for the production of various substances and materials. As a rule, pipeline networks are used for gas transportation from the gas production site to the destination, and their safe operation must be provided with complete information about the thermophysical properties of natural gases in a wide range of temperatures and pressures [1]-[4]. This statement also applies to the use of natural gases in technological processes related to heat and mass transfer. However, most reference books and manuals provide very brief (or no information at all [5]) information about the mass transfer properties of hydrocarbon gases, their mixtures, and natural gases, although such data is extremely necessary.

Studies of multicomponent mass transfer have shown the complex nature of the mixing process [6]-[9]. The occurrence of a pressure drop (diffusive baroeffect) in closed diffusion devices, due to the unequal mobility of the gas mixture molecules, leads to the appearance of a hydrodynamic flow of the gas mixture as a whole. The division of the total transfer of components into the diffusion flow, characterized by true diffusion coefficients, and hydrodynamic allows us to give a correct physical picture of the diffusion process not only in the case of two components, but also in a more complex mixture [10]-[11].

When conducting experiments on the measurement of diffusion coefficients in multicomponent systems, a number of features that are not characteristic of binary gas mixtures were revealed: "reverse diffusion", "osmotic diffusion", "diffusive barrier". These "Tour effects" have become a classic example of the manifestation of the features of multicomponent diffusion. The physics of the occurrence of these seemingly strange phenomena is that the transfer of components observed during diffusion is the result of the addition (vector) of the actual diffusion and hydrodynamic components of the transfer.

The common device for diffusion measurements is a two-flask device. It is used to determine the coefficients of mutual diffusion in a wide range of temperatures, pressures, concentrations, and thermal diffusion.

The paper presents the results of a computational study of diffusion in eight-component systems for five natural hydrocarbon gas mixtures into the air by the computational method applied to a two-column diffusion apparatus. The described EDC (effective diffusion coefficient) method turned out to be convenient for describing multicomponent mass transfer and its features.

II. PHYSICAL-MATHEMATICAL FORMULATION

The phenomenon of multicomponent mass transfer is described in a number of theories. These theories are divided into: strict kinetic Curtiss–Hirschfelder, hydrodynamic Stefan–Maxwell, and thermodynamic [12].

Solving problems of multicomponent diffusion by methods of strict kinetic, thermodynamic and hydrodynamic theories leads to cumbersome and complex expressions, with a large number of different characteristics, which are sometimes not uniquely defined [13]. It is extremely inconvenient to use such expressions in engineering calculations. To significantly simplify the calculation of multicomponent mass transfer, an EDC was introduced, which in the limiting case of a two-component system is identically equal to the usual mutual diffusion coefficient [14]-[15]. It is assumed that the flow of each of the components in the n-component mixture of gases is described by Fick's law:

$$ j_i = -D_i^{eff} \frac{dc_i}{dx}, \qquad (1) $$

where j_i, c_i – the density of the diffusion flow and the concentration of the i-th component, respectively.

Thus, the flow of i-th component in the n-component gas mixture is determined only by the gradient of this component and it's EDC.

Since the flow of each of the components of the mixture depends on the distribution of the concentration of all the components, the EDC is a complex quantity that changes during the transition from one point of the system to another. If this dependence is weakly expressed, we can already use the well-known solution of the corresponding diffusion problem for binary systems by simply replacing the mutual diffusion coefficient (MDC) - D_{12} for D_i^{eff}. Such special cases of diffusion include:

- for systems in which the components of $2,3...S$ move at the same speed or are stationary, the following relation is valid:

$$D_i^{eff} = \frac{1 - C_i}{\sum_{j=2}^{s} \frac{C_j}{D_{1j}}};$$ (2)

- for the trace diffusion of component i into a homogeneous mixture of the others, we have:

$$D_i^{eff} = \frac{C_i}{\sum_{j=2}^{s} \frac{C_j}{D_{1j}}};$$ (3)

- the diffusion of small impurities (trace diffusion) of substances $2,3,...n$ in almost pure substance 1, while $D_i^{eff} = D_{i1}$.

- diffusion in systems in which the mutual diffusion coefficients of the gas pairs entering the mixture are the same, $D_i^{eff} = D_{ij}$;

In other cases, it is necessary to find some EDC values averaged over the length of the diffusion layer. Despite the fact that there are currently no reasonable ways to find the average values of EDC, and therefore this method is widely used.

At the moment, there is enough information on the study of multicomponent mass transfer (for the most common cases) using EDC. Along with experimental studies, methods for calculating multicomponent diffusion were developed, which were based on the Stefan-Maxwell equations. Testing of this method on numerous experiments has shown that it is correct, has sufficient accuracy and is easy to use [16]-[19].

In the literature, a number of records of expressions for EDC are given. We will use the expression which is easily verified in diffusion experiments:

$$D_i^{eff} = D_{ii}^* + \sum_{\substack{j=1 \\ j \neq i}}^{n-1} D_{ij}^* \frac{dc_j}{dc_i},$$ (4)

where D_{ii}^*, $D_{ij}^* = f(D_{ij}, y_i, y_j)$ – the main and cross "practical" diffusion coefficients (PDC) or they are sometimes called matrix coefficients of multicomponent diffusion (MCMD); dc_j / dc_i – the ratio that relates the change in the concentration of the j-th component to the change in the concentration of the i-th component; D_{ij} – MDC of gas pairs i and j; y_i, y_j – molar fractions of components i and j.

The expression (4) in local quantities is quite difficult to apply, so it is simplified by moving to an approximate calculation of the integral (averaged over the entire diffusion layer) EDC of the i-th component in the n-component mixture. The values D_{ii}^*, D_{ij}^* are calculated for the averaged (arithmetic mean) molar fractions, and the ratio of the gradients is replaced by the ratio of the differences in the concentrations of the components between points 0 and L at the boundaries of the diffusion layer:

$$\bar{D}_i^{eff} = \bar{D}_{ii}^* + \sum_{\substack{j=1 \\ j \neq i}}^{n-1} \bar{D}_{ij}^* \left(\frac{c_j^L - c_j^0}{c_i^L - c_i^0} \right).$$ (5)

It follows from (5) that, depending on the distribution of components within the system, the EDC sign depends, which can be either positive or negative. For the simplest multicomponent system – a triple mixture, the expression for calculating the EDC has the form:

$$\bar{D}_i^{eff} = \frac{(1 - \bar{y}_i) D_{ij} D_{ik} + \bar{y}_i D_{jk} D_{ik} + \bar{y}_i \left(D_{ik} Djk - D_{ij} D_{jk} \right) \left(\frac{c_j^L - c_j^0}{c_i^L - c_i^0} \right)}{\bar{y}_i D_{jk} + \bar{y}_j D_{ik} + \bar{y}_k D_{ij}},$$ (6)

i, j, k = 1, 2, 3.

$\bar{y}_i, \bar{y}_j, \bar{y}_k$ - are an averaged molar fractions of components.

Determination of effective coefficients (EDC) requires the presence of data on the coefficient of interdiffusion (MDC). For a binary system (consisting of A and B components), it is a function of temperature, pressure and composition. To obtain the necessary coefficients, we apply the Chapman – Enskog kinetic theory. The Chapman – Enskog formulas in the case of rarefied gases have the form:

$$cD_{AB} = 2.2646 \times 10^{-5} \frac{\sqrt{T(\frac{1}{M_A} + \frac{1}{M_B})}}{\sigma_{AB}^2 \Omega_{D,AB}}.$$ (7)

If we accept that, according to the law of ideal gases $c = p / RT$, we find the following expression for D_{AB}:

$$D_{AB} = 0.0018583 \frac{\sqrt{T^2(\frac{1}{M_A} + \frac{1}{M_B})}}{p \sigma_{AB}^2 \Omega_{D,AB}}.$$ (8)

Here D_{AB} is expressed in sm^2/s; c - in mol/sm^3; T - in K; p -

in atm; σ - in A; $\Omega_{D,AB}$ - is a dimensionless function of temperature and intermolecular potential field for one molecule A and one molecule B. It is convenient to express this potential field by the Lennard-Jones function:

$$\varphi_{AB}(r) = 4\varepsilon_{AB}[(\frac{\sigma_{AB}}{r})^{12} - (\frac{\sigma_{AB}}{r})^{6}]. \qquad (9)$$

Parameters of the forces of intermolecular interaction are shown in Table 1 and functions required for calculating the transport coefficients in rarefied gases are shown in Table 2.

Table 1.
Parameters of the forces of intermolecular interaction

Compon ent	Molecular weight	Lennard-Jones potential parameters	
		σ, Å	ε/k, K
Light elements			
H₂	2.016	2.915	38.0
He	4.003	2.576	10.2
Noble gases			
Ne	20.183	2.789	35.7
Kr	83.80	3.498	225.0
Simple polyatomic substances			
Air	28.97	3.617	97.0
CO₂	44.01	3.996	190.0
Hydrocarbons			
CH₄	16.04	3.822	137.0
C₂H₆	30.07	4.418	230.0
C₃H₈	44.09	5.061	254.0
n-C₄H₁₀	58.12	5.341	313.0
n- C₅H₁₂	72.15	5.769	345.0

Table 2.
Functions required for calculating the transport coefficients

kT/ε_{AB}	$\Omega_{D,AB}$	kT/ε_{AB}	$\Omega_{D,AB}$
0.30	2.662	2.50	0.9996
0.70	1.729	3.20	0.9328
1.00	1.439	3.70	0.8998
1.35	1.253	4.50	0.8610
1.80	1.116	4.80	0.8492
2.10	1.057	5.0	0.8422
6.0	0.8124	40.0	0.5960
7.0	0.7896	60.0	0.5596
8.0	0.7712	70.0	0.5464
9.0	0.7556	80.0	0.5352
10.0	0.7424	90.0	0.5256
20.0	0.6640	100.0	0.5170

In the above Tables 1 and 2 for the model with potential energy the values of $\Omega_{D,AB}$ are given as a function of kT/ε_{AB}. Using these data, it can be calculated that D_{AB} increases approximately in proportion to the power of 2.0 at low temperatures and to the power of 1.65 at very high temperatures. Combining the Lennard-Jones parameters for molecules A and B, it can be calculates the values in the range of the average deviation of about 6%:

$$\sigma_{AB} = \frac{1}{2}(\sigma_A \sigma_B)$$
$$\varepsilon_{AB} = \sqrt{\varepsilon_A \varepsilon_B} \qquad (10)$$

In this way the necessary coefficients can be determined to calculate the effective diffusion coefficients.

III.   CALCULATION METHODOLOGY

When measuring the EDC in most tasks, we used the method of a two-flask diffusion device [20]. The design of the diffusion apparatus, devices and components included in the experimental setup, as well as the operating procedure are described in detail in [21]. One of the devices used in the work had the following parameters: the volumes of the upper and lower flasks: $V_u = V_l = 76.9$ sm³; the length and diameter of the diffusion channel $L = 7.055$ sm and $d = 0.4$ sm, respectively.

For this device, the complex of geometric dimensions (the constant of the device, where S is the cross-sectional area of the channel) was equal to 2261 sm². For the devices available to us, the geometric characteristics could differ slightly, and their constants were in the range from 2100 to 2600 sm². In the presented calculations, we used the geometric parameters of the diffusion apparatus, the constant of which was equal to 2500 sm².

The method of operation on the device is as follows: the flasks of the device are separated from each other and the fore vacuum pump is pumped out of one of the flasks, which is then washed (filled and pumped out again 2-3 times) with gas from the corresponding cylinder and finally filled with gas to a pressure slightly higher than the pressure of the experiment. Similar procedures are performed with the second flask and fill it with a pressure slightly lower than the pressure of the experiment.

After the installation reaches the desired temperature (usually at a temperature of T=298.0 K), the pressure in the flasks is equalized through a special container. According to the model pressure gauges, the pressure in the flasks of the device is noted. The absolute value of the pressure P_{exp} is found by summing the atmospheric pressure and the pressure

taken from the pressure gauge (overpressure).

The diffusion channel opens after equalizing the pressure and simultaneously turns on the stopwatch. At the end of the experiment, the flasks are separated, and the diffusion time is noted. The analysis of gases after diffusion is carried out either by the interferometry method (binary mixtures) or by the chromatographic method (three-component mixtures and higher) from both the upper and lower flasks.

The calculation of the MDC in binary mixtures and the EDC in multicomponent mixtures for a given pressure and temperature is performed as follows:

$$D = \frac{L}{St} \times \frac{1}{(\dfrac{1}{V_{upper}} + \dfrac{1}{V_{lower}})} \ln \frac{\Delta C_0}{\Delta C_t}. \quad (11)$$

Where L, S – the length and cross-sectional area of the diffusion capillary; V_{upper}, V_{lower} - the volumes of the upper and lower flasks of the device; ΔC_0 and ΔC_t - are the concentration difference between the flasks at the initial and final moments of time; t – the diffusion time.

In this paper, the diffusion of five natural hydrocarbon gas mixtures into the air at $T = 298$ K and $P = 0.101$ MPa was investigated by numerical experiment. The composition and concentrations of the components in the mixtures were taken from [5]. At the same time, the choice of a particular gas field was not related to the classification in [5], but was determined only by the concentration of the main gas – methane. This range ranged from its minimum values in the mixture to its maximum values. In our calculations, we took into account all the components, although many of the gases were present in the form of "traces". The concept of "trace" concentration has the following meaning, when the molecules of a given gas do not experience collisions with each other, but only collide with other molecules. In quantitative terms (depending on what gases are in the mixture) this can be up to 5-7%.

Nowadays an important stage in the development of the gas and oil deposits is its exploration. Detailed exploration of a gas deposit, in particular, the availability of information about its chemical composition, physical characteristics contributes to the safety of the working area.

The transition to the integrated design of gas field development, the intensification of gas flow to wellbores, and the automation of installations in gas fields have significantly increased the working flow rates of wellbores, improved gas preparation for transportation, and reduced the cost of natural gas. The results of a computational study of the diffusion process of a multicomponent gas mixture are presented below.

The gas fields and their composition are listed as [5, P.178, 182]:

1. Markovian plast, investigated system: Air(1) – 0,7240 CH4(2) + 0,0930 C2H6(3) + 0,0780 C3H8(4) + 0,0490 n-C4H10(5) + 0,0305 n-C5H12(6) + 0,0100 CO2(7) + 0,0155 N2(8);

2. Urengoy field, investigated system: Air(1) – 0,8531 CH4(2) + 0,0581 C2H6(3) + 0,0536 C3H8(4) + 0,0200 n-C4H10(5) + 0,0018 n-C5H12(6) + 0,0044 CO2(7) + 0,0090 N2(8);

3. Berezanskoye field (average composition), investigated system: Air(1) – 0,8878 CH4(2) + 0,0475 C2H6(3) + 0,0056 C3H8(4) + 0,0015 n-C4H10(5) + 0,0129 n-C5H12(6) + 0,0387 CO2(7) + 0,0060 N2(8);

4. Srednevilyuyskoye field (average composition), investigated system: Air(1) – 0,9220 CH4(2) + 0,0510 C2H6(3) + 0,0090 C3H8(4) + 0,0025 n-C4H10(5) + 0,0009 n-C5H12(6) + 0,0075 CO2(7) + 0,0071 N2(8);

5. Gazli-XII field (average composition), investigated system: Air(1) – 0,9444 CH4(2) + 0,0258 C2H6(3) + 0,0031 C3H8(4) + 0,0015 n-C4H10(5) + 0,0039 n-C5H12(6) + 0,0020 CO2(7) + 0,0193 N2(8).

In the future, for convenience, we will use not the chemical symbols of gases, but their numerical numbering, given after them. To perform the calculations, the MDC of the gas pairs entering the systems was necessary. Calculations of the MDC were carried out according to the Chapman-Enskog theory using the Lennard-Jones potential [19] for $T = 298$ K, $P = 0.101$ MPa.

IV. RESULTS OF NUMERICAL CALCULATION

The MDC values are as follows:

D12 = 0,217; D13 = 0,144; D23 = 0,151; D14 = 0,111;
D24 = 0,121; D34 = 0,077; D15 = 0,79; D25 = 0,105;
D35 = 0,066; D45 = 0,050; D16 = 0,082; D26 = 0,092;
D36 = 0,057; D46 = 0,043; D56 = 0,036; D17 = 0,151;
D27 = 0,165; D37 = 0,104; D47 = 0,079; D57 = 0,067;
D67 = 0,058; D18 = 0,203; D28 = 0,217; D38 = 0,144;
D48 = 0,112; D58 = 0,096; D68 = 0,083; D78 = 0,151 sm^2/s.

Also, air was relied on as one component. This is quite justified if we assume that the concentrations of oxygen and nitrogen do not undergo strong changes [22-23].

The calculated values of EDC and MCMD for the initial distribution of the component concentrations of the five natural gas systems are given below. ($T = 298$ K and $P = 0.101$ MPa).

EDC of components for the **1st system**:
1 – 0,181; **2** – 0,206; **3** – 0,138; **4** – 0,107; **5** – 0,068; **6** – 0,079; **7** - 0,144; **8** – 0,193 sm^2/s.

MCMD for the **1st system** for independent flows and gradients (first seven components):

D(1,1) = 0,1868	D(1,2) = -0,0066	D(1,3) = 0,0273
D(1,4) = 0,0430	D(1,5) = 0,0623	D(1,6) = 0,0570
D(1,7) = 0,0246	D(2,1) = 0,0004	D(2,2) = 0,1968
D(2,3) = 0,0209	D(2,4) = 0,0299	D(2,5) = 0,0316
D(2,6) = 0,0382	D(2,7) = 0,0164	D(3,1) = 0,0001
D(3,2) = -0,0005	D(3,3) = 0,1346	D(3,4) = 0,0042
D(3,5) = 0,0045	D(3,6) = 0,0055	D(3,7) = 0,0025
D(4,1) = 0,0001	D(4,2) = -0,0006	D(4,3) = 0,0023
D(4,4) = 0,1063	D(4,5) = 0,0039	D(4,6) = 0,0048

D(4,7) = 0,0023   D(5,1) = 0,0009   D(5,2) = -0,0004
D(5,3) = 0,0015   D(5,4) = 0,0023   D(5,5) = 0,0831
D(5,6) = 0,0031   D(5,7) = 0,0015   D(6,1) = 0,0001
D(6,2) = -0,0003   D(6,3) = 0,0009   D(6,4) = 0,0015
D(6,5) = 0,0016   D(6,6) = 0,0783   D(6,7) = 0,0009
D(7,1)= 0,00001   D(7,2) = -0,0001   D(7,3) = 0,0003
D(7,4) = 0,0005   D(7,5) = 0,0005   D(7,6) = 0,0006
D(7,7) = 0,1397 sm²/s.

EDC of components for the **2ⁿᵈ system**:

1 – 0,199; **2** – 0,212; **3** – 0,141; **4** – 0,107; **5** – 0,064; **6** – 0,077; **7** - 0,146; **8** – 0,198 sm²/s.

MCMD for the **2ⁿᵈ system** for independent flows and gradients (first seven components):

D(1,1) = 0,1985   D(1,2) = -0,0069   D(1,3) = 0,0287
D(1,4) = 0,0455   D(1,5) = 0,0669   D(1,6) = 0,0605
D(1,7) = 0,0259   D(2,1) = 0,0002   D(2,2) = 0,2071
D(2,3) = 0,0261   D(2,4) = 0,0372   D(2,5) = 0,0392
D(2,6) = 0,0476   D(2,7) = 0,0203   D(3,1) = 0,00001
D(3,2) = -0,0003   D(3,3) = 0,1415   D(3,4) = 0,0028
D(3,5) = 0,0030   D(3,6) = 0,0036   D(3,7) = 0,0017
D(4,1) = 0,0001   D(4,2) = -0,0004   D(4,3) = 0,0017
D(4,4) = 0,1119   D(4,5) = 0,0029   D(4,6) = 0,0035
D(4,7) = 0,0017   D(5,1) = 0,0004   D(5,2) = -0,0002
D(5,3) = 0,0006   D(5,4) = 0,0010   D(5,5) = 0,0865
D(5,6) = 0,0014   D(5,7) = 0,0006   D(6,1) = 0,0000
D(6,2) = -0,00002   D(6,3) = 0,0001   D(6,4) = 0,0001
D(6,5) = 0,0001   D(6,6) = 0,0817   D(6,7) = 0,0007
D(7,1) = 0,0000   D(7,2) = -0,00004   D(7,3) = 0,0001
D(7,4) = 0,0002   D(7,5) = 0,0002   D(7,6) = 0,0003
D(7,7) = 0,1487 sm²/s.

EDC of components for the **3ʳᵈ system**:

1 – 0,206; **2** – 0,214; **3** – 0,142; **4** – 0,107; **5** – 0,063; **6** – 0,077; **7** - 0,146; **8** – 0,200 sm²/s.

MCMD for the **3ʳᵈ system** for independent flows and gradients (first seven components):

D(1,1) = 0,2029   D(1,2) = -0,0070   D(1,3) = 0,0292
D(1,4) = 0,0464   D(1,5) = 0,0685   D(1,6) = 0,0618
D(1,7) = 0,0264   D(2,1) = 0,00003   D(2,2) = 0,2108
D(2,3) = 0,0277   D(2,4) = 0,0394   D(2,5) = 0,0414
D(2,6) = 0,0505   D(2,7) = 0,0214   D(3,1) = 0,0000
D(3,2) = -0,0002   D(3,3) = 0,1440   D(3,4) = 0,0023
D(3,5) = 0,0025   D(3,6) = 0,0030   D(3,7) = 0,0014
D(4,1) = 0,00001   D(4,2) = -0,0001   D(4,3) = 0,0002
D(4,4) = 0,1118   D(4,5) = 0,0003   D(4,6) = 0,0004
D(4,7) = 0,0002   D(5,1) = 0,00003   D(5,2)=-0,00001
D(5,3) = 0,0001   D(5,4) = 0,0001   D(5,5) = 0,0871
D(5,6) = 0,0001   D(5,7) = 0,0001   D(6,1)= 0,00002
D(6,2) = -0,0001   D(6,3) = 0,0004   D(6,4) = 0,0007
D(6,5) = 0,0007   D(6,6) = 0,0842   D(6,7) = 0,0004
D(7,1) = 0,0000   D(7,2) = -0,0004   D(7,3) = 0,0013
D(7,4) = 0,0020   D(7,5) = 0,0021   D(7,6) = 0,0026
D(7,7) = 0,1530 sm²/s.

EDC of components for the **4ᵗʰ system**:

1 – 0,210; **2** – 0,215; **3** – 0,142; **4** – 0,107; **5** – 0,062; **6** – 0,076; **7** - 0,148; **8** – 0,201 sm²/s.

MCMD for the **4ᵗʰ system** for independent flows and gradients (first seven components):

D(1,1) = 0,2054   D(1,2) = -0,0071   D(1,3) = 0,0296
D(1,4) = 0,0470   D(1,5) = 0,0697   D(1,6) = 0,0627
D(1,7) = 0,0267   D(2,1) = 0,00003   D(2,2) = 0,2131
D(2,3) = 0,0291   D(2,4) = 0,0415   D(2,5) = 0,0435
D(2,6) = 0,0532   D(2,7) = 0,0225   D(3,1) = 0,0000
D(3,2) = -0,0003   D(3,3) = 0,1460   D(3,4) = 0,0025
D(3,5) = 0,0027   D(3,6) = 0,0033   D(3,7) = 0,0015
D(4,1) = 0,00001   D(4,2) = -0,0001   D(4,3) = 0,0003
D(4,4) = 0,1136   D(4,5) = 0,0005   D(4,6) = 0,0006
D(4,7) = 0,0003   D(5,1) = 0,0001   D(5,2) = -0,00002
D(5,3) = 0,0001   D(5,4) = 0,0001   D(5,5) = 0,0884
D(5,6) = 0,0002   D(5,7) = 0,0001   D(6,1) = 0,00000
D(6,2) = -0,00001   D(6,3) = 0,00003   D(6,4) = 0,0001
D(6,5) = 0,0001   D(6,6) = 0,0847   D(6,7) = 0,00003
D(7,1) = 0,0000   D(7,2) = -0,0001   D(7,3) = 0,0002
D(7,4) = 0,0004   D(7,5) = 0,0004   D(7,6) = 0,0005
D(7,7) = 0,1543 sm²/s.

EDC of components for the **5ᵗʰ system**:

1 – 0,212; **2** – 0,216; **3** – 0,142; **4** – 0,107; **5** – 0,061; **6** – 0,075; **7** - 0,148; **8** – 0,202 sm²/s.

MCMD for the **5ᵗʰ system** for independent flows and gradients (first seven components):

D(1,1) = 0,2071   D(1,2) = -0,0072   D(1,3) = 0,0298
D(1,4) = 0,0474   D(1,5) = 0,0704   D(1,6) = 0,0632
D(1,7) = 0,0269   D(2,1) = 0,00002   D(2,2) = 0,2146
D(2,3) = 0,0301   D(2,4) = 0,0428   D(2,5) = 0,0449
D(2,6) = 0,0549   D(2,7) = 0,0232   D(3,1) = 0,0000
D(3,2) = -0,0001   D(3,3) = 0,1464   D(3,4) = 0,0013
D(3,5) = 0,0014   D(3,6) = 0,0017   D(3,7) = 0,0008
D(4,1) = 0,0000   D(4,2) = -0,00003   D(4,3) = 0,0001
D(4,4) = 0,1143   D(4,5) = 0,0002   D(4,6) = 0,0002
D(4,7) = 0,0001   D(5,1) = 0,00003   D(5,2) = -0,00001
D(5,3) = 0,0001   D(5,4) = 0,0001   D(5,5) = 0,0891
D(5,6) = 0,0001   D(5,7) = 0,0001   D(6,1) = 0,00001
D(6,2) = -0,00004   D(6,3) = 0,0001   D(6,4) = 0,0002
D(6,5) = 0,0002   D(6,6) = 0,0857   D(6,7) = 0,0001
D(7,1) = 0,0000   D(7,2) = -0,00002   D(7,3) = 0,0001
D(7,4) = 0,0001   D(7,5) = 0,0001   D(7,6) = 0,0001
D(7,7) = 0,1555 sm²/s.

At the Figures 1-2 presented the appearance of the screen with input data and the text of the report file with the results of calculations for the 1ˢᵗ system (Markovian plast): Air – 0, 7240CH4 + 0, 0930C2H6 + 0, 0780C3N8 + 0.0490 n - C4H10 + 0.0305 n-C5H12 + 0, 0100CO2 + 0, 0155N2 (at T=773 K and P=30 MPa).

```
Расчет диффузии в двухколбовом аппарате
**********************************
Введите число компонентов - 8
Обзовите газы по номерам :
1 - Air
2 - CH4
3 - C2H6
4 - C3H8
5 - C4H10
6 - C5H12
7 - CO2
8 - N2
Введите число газов в верхней колбе - 1
Введите число газов в нижней колбе - 7
Введите начальное распределение концентраций газов
(N газа , концентрация в мольных долях <Enter>)
Концентрация компонентов в верхней колбе :
 -1.1.0
Концентрация компонентов в нижней колбе :
 -2.0.7240
 -3.0.0930
 -4.0.0780
 -5.0.0490
 -6.0.0305
 -7.0.0100
 -8.0.0155
Введите коэффициенты взаимной диффузии, см2/с (Р норм, атм.):
D(1,2) = 0.045
D(1,3) = 0.030
D(2,3) = 0.034
D(1,4) = 0.024
D(2,4) = 0.027
D(3,4) = 0.018
D(1,5) = 0.021
D(2,5) = 0.024
D(3,5) = 0.016
D(4,5) = 0.012
D(1,6) = 0.018
D(2,6) = 0.021
D(3,6) = 0.014
D(4,6) = 0.011
D(5,6) = 0.009
D(1,7) = 0.033
D(2,7) = 0.036
D(3,7) = 0.024
D(4,7) = 0.018
D(5,7) = 0.016
D(6,7) = 0.014
D(1,8) = 0.041
D(2,8) = 0.045
D(3,8) = 0.030
D(4,8) = 0.024
D(5,8) = 0.021
D(6,8) = 0,018
D(7,8) = 0.032
Введите постоянную прибора в см2 - 2500
Атмосферное давление в мм. рт. ст. - 760
Давление в кгс/см2 :
Р_нач - 0
Р_кон - 30
Шаг по давлению - 30
Введите начальное время для вывода в минутах - 0
Введите конечное время для вывода в минутах - 150
Интервал вывода в минутах - 15
< Ввод данных завершен. Для начала расчета используйте Ctrl+E >
-------------------------------------------------------
```

Fig. 1. The input data for the Markovian plast system.

```
-------------------------------------------------------
Результаты расчета
-------------------------------------------------------
Избыточное давление по манометру P = 0 кгс/см2 ( 0 МПа )
Коэффициент перевода давления Кр = 1
М к м д , см2/с (для условий опыта) :
D(1,1) = 0,03881 ; D(1,2) = -0,00197 ; D(1,3) = 0,00525
D(1,4) = 0,00812 ; D(1,5) = 0,00961 ; D(1,6) = 0,01115 ;
D(1,7) = 0,00382 ; D(2,1) = 0,00000 ; D(2,2) = 0,04148 ;
D(2,3) = 0,00338 ; D(2,4) = 0,00555 ; D(2,5) = 0,00642 ;
D(2,6) = 0,00726 ; D(2,7) = 0,00284 ; D(3,1) = 0,00000 ;
D(3,2) = -0,00024 ; D(3,3) = 0,02932 ; D(3,4) = 0,00071
D(3,5) = 0,00083 ; D(3,6) = 0,00093 ; D(3,7) = 0,00036 ;
D(4,1) = 0,00000 ; D(4,2) = -0,00019 ; D(4,3) = 0,00037
D(4,4) = 0,02355 ; D(4,5) = 0,00079 ; D(4,6) = 0,00081 ;
D(4,7) = 0,00040 ; D(5,1) = 0,00000 ; D(5,2) = -0,00013
D(5,3) = 0,00022 ; D(5,4) = 0,00042 ; D(5,5) = 0,02059 ;
D(5,6) = 0,00057 ; D(5,7) = 0,00024 ; D(6,1) = 0,00000 ;
D(6,2) = -0,00009 ; D(6,3) = 0,00013 ; D(6,4) = 0,00022
D(6,5) = 0,00031 ; D(6,6) = 0,01779 ; D(6,7) = 0,00014 ;
D(7,1) = -0,00001 ; D(7,2) = -0,00002 ; D(7,3) = 0,00005
D(7,4) = 0,00009 ; D(7,5) = 0,00010 ; D(7,6) = 0,00011 ;
D(7,7) = 0,03093 ;
```

имя газа	концентрации компонентов, мольные доли		разность концен., мольные доли	мгновен. значения эффект. коэфф. диффузии, см2/с	интегр. значения эффект. коэфф. диффузии см2/с
	верхняя колба	нижняя колба			
t = 0 мин					
Air	1,00000	0,00000	1,00000	0,03826	0,03826
CH4	0,00000	0,72400	-0,72400	0,04329	0,04329
C2H6	0,00000	0,09300	-0,09300	0,02885	0,02885
C3H8	0,00000	0,07800	-0,07800	0,02311	0,02311
C4H10	0,00000	0,04900	-0,04900	0,02012	0,02012
C5H12	0,00000	0,03050	-0,03050	0,01705	0,01705
CO2	0,00000	0,01000	-0,01000	0,03172	0,03172
N2	0,00000	0,01550	-0,01550	0,03935	0,03935
t = 15 мин					
Air	0,99316	0,00684	0,98632	0,03825	0,03825
CH4	0,00560	0,71840	-0,71280	0,04330	0,04330
C2H6	0,00048	0,09252	-0,09204	0,02886	0,02885
C3H8	0,00032	0,07768	-0,07735	0,02312	0,02312
C4H10	0,00018	0,04882	-0,04865	0,02014	0,02013
C5H12	0,00009	0,03041	-0,03031	0,01706	0,01706
CO2	0,00006	0,00994	-0,00989	0,03173	0,03172
N2	0,00011	0,01539	-0,01528	0,03937	0,03936
t = 30 мин					
Air	0,98642	0,01358	0,97284	0,03823	0,03825
CH4	0,01111	0,71289	-0,70178	0,04332	0,04330
C2H6	0,00096	0,09204	-0,09109	0,02887	0,02886
C3H8	0,00064	0,07736	-0,07671	0,02313	0,02312
C4H10	0,00035	0,04865	-0,04829	0,02015	0,02014
C5H12	0,00019	0,03031	-0,03013	0,01708	0,01706
CO2	0,00011	0,00989	-0,00977	0,03174	0,03173
N2	0,00022	0,01528	-0,01507	0,03938	0,03937
t = 45 мин					
Air	0,97977	0,02023	0,95954	0,03822	0,03824
CH4	0,01654	0,70746	-0,69092	0,04333	0,04331
C2H6	0,00143	0,09157	-0,09015	0,02889	0,02887
C3H8	0,00096	0,07704	-0,07608	0,02315	0,02313
C4H10	0,00053	0,04847	-0,04795	0,02016	0,02014
C5H12	0,00028	0,03022	-0,02994	0,01710	0,01707
CO2	0,00017	0,00983	-0,00966	0,03175	0,03174

```
-------------------------------------------------------
Конец расчета
-------------------------------------------------------
```

Fig. 2. The results of calculations for the Markovian plast
system.

Since the diffusion process in a two-column apparatus is non-stationary, the behavior of the diffusion coefficients of the components in the systems over time is naturally of interest. As an example, this relationship is presented in Figure 3 below for the Urengoy field (2nd system).

```
-----------------------------------------------------------------
|имя   |концентрации       |разность |мгновен. |интегр.  |
|газа  |компонентов,       |концен., |значения |значения |
|      |мольные доли       |мольные  |эффект.  |эффект.  |
|      |-------------------|доли     |коэфф.   |коэфф.   |
|      |верхняя  |нижняя   |         |диффузии,|диффузии,|
|      |колба    |колба    |         |см2/с    |см2/с    |
-----------------------------------------------------------------
t = 0 мин
| Air| 1,00000 | 0,00000 | 1,00000 | 0,19871 | 0,19871 |
| CH4| 0,00000 | 0,85310 |-0,85310 | 0,21208 | 0,21208 |
|C2H6| 0,00000 | 0,05810 |-0,05810 | 0,14082 | 0,14082 |
|C3H8| 0,00000 | 0,05360 |-0,05360 | 0,10720 | 0,10720 |
|C4H10|0,00000 | 0,02000 |-0,02000 | 0,06445 | 0,06445 |
|C5H12|0,00000 | 0,00180 |-0,00180 | 0,07718 | 0,07718 |
| CO2| 0,00000 | 0,00440 |-0,00440 | 0,14646 | 0,14646 |
| N2 | 0,00000 | 0,00900 |-0,00900 | 0,19826 | 0,19826 |
t = 300 мин
| Air| 0,62184 | 0,37816 | 0,24367 | 0,19298 | 0,19610 |
| CH4| 0,33554 | 0,51756 |-0,18202 | 0,21775 | 0,21455 |
|C2H6| 0,01864 | 0,03946 |-0,02083 | 0,14404 | 0,14247 |
|C3H8| 0,01461 | 0,03899 |-0,02437 | 0,11124 | 0,10946 |
|C4H10|0,00409 | 0,01591 |-0,01181 | 0,07920 | 0,07316 |
|C5H12|0,00039 | 0,00141 |-0,00101 | 0,08231 | 0,08021 |
| CO2| 0,00145 | 0,00295 |-0,00151 | 0,15126 | 0,14892 |
| N2 | 0,00344 | 0,00556 |-0,00212 | 0,20355 | 0,20064 |
t = 600 мин
| Air| 0,53133 | 0,46867 | 0,06266 | 0,18355 | 0,19236 |
| CH4| 0,40833 | 0,44477 |-0,03644 | 0,23107 | 0,21897 |
|C2H6| 0,02540 | 0,03270 |-0,00730 | 0,14711 | 0,14401 |
|C3H8| 0,02137 | 0,03223 |-0,01085 | 0,11327 | 0,11091 |
|C4H10|0,00673 | 0,01327 |-0,00653 | 0,08436 | 0,07768 |
|C5H12|0,00062 | 0,00118 |-0,00055 | 0,08392 | 0,08175 |
| CO2| 0,00195 | 0,00245 |-0,00050 | 0,15592 | 0,15124 |
| N2 | 0,00426 | 0,00474 |-0,00047 | 0,21408 | 0,20439 |
-----------------------------------------------------------------
```

Fig. 3. The text of the report file with the results of calculations for the Air system (1) – 0.8531 CH4 (2) + 0.0581 C2H6(3) + 0.0536 C3H8(4) + 0.0200 n-C4H10(5) + 0.0018 n-C5H12(6) + 0.0044 CO2(7) + 0.0090 N2(8) at T = 298 K and P = 0.101 MPa.

The results showed a change in the EDC of the system components not exceeding 5% over a time interval of 600 minutes. This behavior of gases is explained by the fact that their MDC in the air differ slightly. Similar results were obtained for the other systems studied.

Note a very important point in the use of EDC and MCMD in the calculation of the diffusion fluxes of components in systems. In this case, when using the EDC, only 8 of them are needed, and the MCMD for independent streams - 48. It follows that the number of necessary coefficients to describe multicomponent diffusion is easier and simpler using EDC than MCMD.

Using these methods it is planning to hold experimental studies that aimed at establishing the boundaries of the transition from diffusion to convection, which also involves the study of developed convective mixing and the determination of the boundaries of stability of various modes of motion [24].

V. Conclusion

In conclusion, it can be said that: 1) to describe multicomponent diffusion is easier and simpler using EDC; 2) the calculated diffusion coefficients (EDC and MCMD) for the diffusion processes of natural hydrocarbon gas systems into

the air can serve as reference information in practical applications.

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