



## HIGH-PRESSURE TERNARY DIFFUSION IN AN N<sub>2</sub>-C<sub>1</sub>-C<sub>5</sub> MIXTURE

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***Abstract.** This work describes a general procedure for the determination of Fick's diffusion coefficients associated to the evaporation process in ternary mixtures involving light hydrocarbons at high pressure. The non-idealities in the vapor and liquid phases that naturally arise in this situation are taken into account by the application of thermodynamical corrections to perfect gas results. Application of the theory is then exemplified for the case of an C<sub>5</sub>-C<sub>1</sub>-N<sub>2</sub> mixture at 101 bar abs and 38.5°C. Numerical results are presented and the applicability of the perfect gas model to this case is discussed.*

***Keywords:** Ternary diffusion, Porous media, Oil recovering.*

### 1. INTRODUCTION

The petroleum extracted in the primary exploitation phase of fractured reservoirs comes essentially from the fractures. In fact, at the end of this phase, a great deal of oil is still trapped in the pores of the solid matrix. One solution to recover this matrix oil is to inject a dry gas in the reservoir. The oil in the matrix vaporizes and feeds by diffusion the gas circulating in the fractures. This technique allows, theoretically, a total recovering of light and intermediate fractions of the oil.

The analysis and modeling of this recovering process is complex, since an oil is made up by dozens of different components (Gravier, 1986). For this reason, it is common practice to perform laboratory experiments to try to isolate the important parameters. One such experiment was carried out by Morel et al. (1990), who took a sample bar of chalk, initially saturated with a mixture of pentane and methane (C<sub>5</sub>-C<sub>1</sub>) under typical conditions of an oilfield (101 bar abs and 38.5°C), and used methane as injection gas (experiment C<sub>5</sub>-C<sub>1</sub>) and nitrogen (experiment C<sub>5</sub>-C<sub>1</sub>-N<sub>2</sub>). They noticed that the methane was more efficient as a recovering agent than the nitrogen and also that these two cases were phenomenologically different. However, the explanation for this difference and even an optimization of the process would demand results for a wide range of the operating parameters, and also more information about what happens at the pore level of the matrix block, what can be more easily

accomplished with the aid of numerical simulations in porous media. Examples of such simulations can be found in Laurindo and Prat (1998) and Prat (1995).

This is a problem of transport in porous media, where an appropriate theoretical macroscopic representation must be preceded by the complete comprehension of the diffusion phenomena that occur at the microscopic level. In each pore, an elementary diffusional process takes place, where a binary mixture of pentane and methane C5-C1, initially at liquid-vapor equilibrium, is exposed to the passage of a gas current. This gas diffuses in the mixture at the same time that methane and pentane are recovered at the outlet. If the injection gas is also methane, it is a binary diffusion problem, and the diffusion molar fluxes  $J$  (moles/m<sup>2</sup>.s) can be represented by Fick's law:

$$\vec{J}_1 = -c_t \mathcal{D}_{12} \nabla x_1 \quad (1)$$

where  $c_t$  is the total molar fraction (moles/m<sup>3</sup>),  $x_1$  is the molar fraction of species 1, and where the coefficient  $\mathcal{D}_{12}$  (m<sup>2</sup>/s), called "molecular diffusion coefficient" ( $\mathcal{D}_{12} = \mathcal{D}_{21}$ ), is constant. On the other hand, in the recovering with nitrogen, the basic event is the ternary diffusion in C5-C1-N2. In this case, the modeling can be done by an extension of Fick's law:

$$\vec{J}_1 = -c_t D_{11} \nabla x_1 - c_t D_{12} \nabla x_2 \quad \text{and} \quad \vec{J}_2 = -c_t D_{21} \nabla x_1 - c_t D_{22} \nabla x_2 \quad (2)$$

Equation (2) keeps the structurally simple form of the binary case, but in this case there are "crossed coefficients" establishing connections between flux of one component and gradient of the other and the relationship between the ternary coefficients ( $D_{ij}$ ) and the molecular ones ( $\mathcal{D}_{ij}$ ) involves a strong dependence on the concentrations, even for perfect gases. Besides, under high-pressure conditions and for liquids, the perfect gas model cannot be applied and the real coefficients may present significant deviations from the ideal ones.

Therefore, a precise determination of Fick's coefficients is essential for the theoretical modeling of recovering by diffusion in a fractured reservoir. Such an evaluation is particularly difficult in the ternary case, where the coefficients have to be determined to a wide range of concentration values and in conditions that must resemble those of a real reservoir. This work presents a complete description of a numerical determination of the Fick's diffusion coefficients associated to the evaporation process in an C5-C1-N2 mixture at 101 bar abs and 38.5°C, taking perfect gas results and applying thermodynamical corrections for the non-idealities in the vapor and liquid phases.

The final coefficients are the combination of the results of three separated analysis: i) the dependence of the ternary coefficients on the concentrations for perfect gases, ii) the quantification of the non-idealities for the two phases and iii) the determination of the molecular coefficients for the two phases.

## 2. CALCULATION OF FICK'S COEFFICIENTS

### 2.1 Fick's coefficients for the ternary diffusion in perfect gases

For perfect gases, the relationship between binary and ternary diffusion coefficients comes from the Kinetic Theory of Gases, as given by Maxwell-Stefan's formulation:

$$\vec{d}_i = \nabla x_i = \sum_{j=1}^3 \frac{(x_i \vec{J}_j - x_j \vec{J}_i)}{c_t \mathcal{D}_{ij}} \quad (3)$$

where  $\bar{d}_i$  is the diffusion potential for component “i” and  $\mathcal{D}_{ij}$  is the molecular coefficient associated to the pair “i-j”. This formula can be inverted, to produce:

$$(\vec{J}) = -c_i [B]^{-1} (\vec{d}) \quad (4)$$

where B is a coefficient matrix defined by (i=1,2 and j=1,2):

$$B_{ij} = -x_i \left( \frac{1}{\mathcal{D}_{ij}} - \frac{1}{\mathcal{D}_{i3}} \right) \quad \text{and} \quad B_{ii} = \frac{x_i}{\mathcal{D}_{i3}} + \sum_{\substack{k=1 \\ i \neq k}}^3 \frac{x_k}{\mathcal{D}_{ik}} \quad (5)$$

Therefore, for ideal ternary mixtures, Fick’s coefficients [ $D^{id}$ ] can be expressed as functions of the molecular coefficients ( $\mathcal{D}$ ) related to each pair i-j and of the molar concentrations  $x_i$  as [ $D^{id}$ ] = [ $B$ ]<sup>-1</sup>, that is,

$$D_{11}^{id}(x_1, x_2) = \mathcal{D}_{13} \frac{[x_1 \mathcal{D}_{23} + (1 - x_1) \mathcal{D}_{12}]}{S} \quad (6)$$

$$D_{12}^{id}(x_1, x_2) = x_1 \mathcal{D}_{23} \frac{(\mathcal{D}_{13} - \mathcal{D}_{12})}{S} \quad (7)$$

$$D_{21}^{id}(x_1, x_2) = x_2 \mathcal{D}_{13} \frac{(\mathcal{D}_{23} - \mathcal{D}_{12})}{S} \quad (8)$$

$$D_{22}^{id}(x_1, x_2) = \mathcal{D}_{23} \frac{[x_2 \mathcal{D}_{13} + (1 - x_2) \mathcal{D}_{12}]}{S} \quad (9)$$

where the function S is given by:

$$S(x_1, x_2) = x_1 \mathcal{D}_{23} + x_2 \mathcal{D}_{13} + (1 - x_1 - x_2) \mathcal{D}_{12} \quad (10)$$

## 2.2 Thermodynamical correlations for the non-idealities

Taylor and Krishna (1993) suggest that a good way to adapt Eqs. (6) to (10) to high-pressure gases and to liquids is to redefine the potential “d” on the basis of the chemical potential  $\mu$ :

$$\bar{d}_i = \frac{x_i}{RT} \nabla_{T,P} \mu_i = \frac{x_i}{RT} \sum_{j=1}^{n-1} \left. \frac{\partial \mu_i}{\partial x_j} \right|_{T,P} \nabla x_j \quad (11)$$

Moreover, using the more practical concept of fugacity,  $d\mu_i = RT d(\ln f_i)$ , Eq. (11) can be rewritten as:

$$\vec{d}_i = x_i \sum_{j=1}^{n-1} \left. \frac{\partial \ln f_i}{\partial x_j} \right|_{T,P} \nabla x_j \quad (12)$$

allowing Maxwell-Stefan's diffusion potential for non-ideal fluids to be written in a compact form as:

$$d_i = \sum_{j=1}^{n-1} \Gamma_{ij} \nabla x_j \quad (13)$$

where the matrix  $\Gamma$  is defined by:

$$\Gamma_{ij} = \delta_{ij} + x_i \left. \frac{\partial \ln \varphi_i}{\partial x_j} \right|_{T,P} \quad (14)$$

where  $\delta_{ij}$  is the identity matrix and  $\varphi$  is the fugacity coefficient, defined by  $\varphi_i = f_i/(x_i P)$ .

**Calculation of the fugacity coefficient,  $\varphi$ .** Prausnitz et al. (1986) show that an expression for the calculation of  $\varphi$  can be obtained using volumetric relations, such as  $V = F(T, P, n_1, n_2, \dots)$ , and applying the definition of fugacity, which leads to:

$$RT \ln \varphi_i = RT \ln \frac{f_i}{x_i P} = \int_0^P \left[ \bar{v}_i - \frac{RT}{P} \right] dP \quad (15)$$

where  $\bar{v}_i$  is the partial molar volume. A good precision for Eq. (15) depends only on the precision achieved by the PVT relation chosen to represent component i's properties inside a mixture.

According to Conrard and Gravier (1980), one of the most precise PVT relations to the representation of natural hydrocarbons is the Peng-Robinson (1976) equation of state:

$$P = \frac{RT}{v-b} - \frac{a}{v(v+b)+b(v-b)} \quad (16)$$

where  $P$  is pressure (Pa),  $v$  is molar volume ( $\text{m}^3/\text{mol}$ ),  $R$  is the universal constant of gases ( $R=8.3143 \text{ (J/gmol.K)}$ ), and "a" and "b" are constants that depend on the substance being represented. Equation (16) is originally associated to the description of a pure substance, but it can also represent a mixture of hydrocarbons, provided that appropriate values to the parameters "a" and "b" be supplied. The values of "a" and "b" for mixtures can be obtained from the corresponding values to pure substances with the aid of "mixing rules", as described by Peng and Robinson (1976). The values of the critical properties and the acentricity factor for the components of the ternary mixture C5-C1-N2, needed for the application of Peng-Robinson equation, can be found in Walas (1985) and Perry (1950).

The substitution of Eq. (16) into Eq. (15) leads, after the integration, to the expression shown by Peng and Robinson (1976) for the theoretical determination of the fugacity coefficient of a component "i" in a mixture, as a function of the molar concentration:

$$\ln(\varphi_i) = \ln\left(\frac{f_i}{x_i P}\right) = \frac{b_i^{pur}}{b^{mixt}}(Z-1) - \ln(Z-B) - \frac{A}{2\sqrt{2}B} \Phi_i \Theta \quad (17)$$

where  $A = a^{mixt} P / (R^2 T^2)$ ,  $B = b^{mixt} P / (RT)$ ,  $Z = P_V / (RT)$  and:

$$\Phi_i = \left( \frac{2 \sum_r x_r a_{ri}}{a^{mixt}} - \frac{b_i^{pur}}{b^{mixt}} \right) \quad \text{and} \quad \Theta = \ln\left(\frac{Z + 2.414B}{Z - 0.414B}\right) \quad (18)$$

In order to be used in the thermodynamical corrections, the derivative of Eq. (17) must be obtained, which leads to:

$$\begin{aligned} \frac{\partial \ln \varphi_i}{\partial x_k} = & \frac{b_i^{pur}}{(b^{mixt})^2} \left( b^{mixt} \frac{\partial Z}{\partial x_k} - \frac{\partial b^{mixt}}{\partial x_k} (Z-1) \right) - \frac{1}{(Z-B)} \left( \frac{\partial Z}{\partial x_k} - \frac{\partial B}{\partial x_k} \right) + \\ & - \frac{A}{2\sqrt{2}B} \left\{ \left( \frac{1}{A} \frac{\partial A}{\partial x_k} - \frac{1}{B} \frac{\partial B}{\partial x_k} \right) \Phi_i \Theta + \frac{\partial \Phi_i}{\partial x_k} \Theta + \Phi_i \frac{\partial \Theta}{\partial x_k} \right\} \end{aligned} \quad (19)$$

where:

$$\frac{\partial \Phi_i}{\partial x_k} = \frac{2}{(a^{mixt})^2} \left( a^{mixt} (a_{ki} - a_{3i}) - \sum_r x_r a_{ri} \frac{\partial a}{\partial x_k} \right) + \frac{b_i^{pur}}{(b^{mixt})^2} \frac{\partial b^{mixt}}{\partial x_k} \quad (20)$$

$$\frac{\partial \Theta}{\partial x_k} = \frac{1}{(Z + 2.414B)} \left( \frac{\partial Z}{\partial x_k} + 2.414 \frac{\partial B}{\partial x_k} \right) - \frac{1}{(Z - 0.414B)} \left( \frac{\partial Z}{\partial x_k} - 0.414 \frac{\partial B}{\partial x_k} \right) \quad (21)$$

and where the derivatives of the parameters “a” and “b” are obtained directly from the definitions shown in Peng and Robinson’s work.

Finally, Peng and Robinson showed in their work that the state equation that they propose can be inverted to the form:

$$Z^3 - \lambda_1 Z^2 - \lambda_2 Z - \lambda_3 = 0 \quad (22)$$

where:

$$\lambda_1 = (1-B) \quad , \quad \lambda_2 = -(A - 3B^2 - 2B) \quad , \quad \lambda_3 = (AB - B^2 - B^3) \quad (23)$$

from where it is obtained that:

$$(3Z^2 - 2\lambda_1 Z - \lambda_2) \frac{\partial Z}{\partial x_k} = -\frac{\partial B}{\partial x_k} (Z^2 + \lambda_2) - Z \left[ \frac{\partial A}{\partial x_k} - 2(3B+1) \frac{\partial B}{\partial x_k} \right] + B \frac{\partial A}{\partial x_k} \quad (24)$$

which completes the formulation of the fugacity coefficients.

### 2.3 Molecular coefficients for the vapor phase

According to Bird et al. (1960), the diffusion coefficients for the gaseous mixtures can be determined by the Kinetic Theory of Gases with an error of less than 5%. The expression presented by Bird et al. for a perfect gas at low density is:

$$\mathcal{D}_{ij}^V = CT^{3/2} \frac{\sqrt{(M_i + M_j)/(M_i M_j)}}{P \sigma_{ij}^2 \Omega_D} \quad (25)$$

$$C = 0.02199 - 0.00507 \sqrt{\frac{M_i + M_j}{M_i M_j}} \quad \text{and} \quad \Omega_D = \frac{1.06}{\tau^{0.156}} + \frac{0.193}{e^{0.476\tau}} + \frac{1.53}{e^{1.036\tau}} + \frac{1.765}{e^{3.894\tau}} \quad (26)$$

In the expressions above,  $\mathcal{D}$  is the molecular diffusion coefficient ( $\text{m}^2/\text{s}$ );  $M_i$  is the molar mass of component  $i$  ( $\text{g}/\text{mol}$ );  $P$  is the pressure ( $\text{Pa}$ );  $T$  is the temperature ( $\text{K}$ );  $\sigma_{ij} = (\sigma_i + \sigma_j)/2$  is the average collision diameter ( $\text{\AA}$ );  $\tau = kT/\varepsilon_{ij}$ , where  $\varepsilon_{ij} = \sqrt{\varepsilon_i \varepsilon_j}$  is an average energy ( $\text{J}/\text{molecule}$ ) and  $k$  is the Boltzmann constant;  $\Omega_D$  is a dimensionless function of the temperature and of the intermolecular field for one molecule of “ $i$ ” and one of “ $j$ ”. The values of the physical constants are shown in Table 1.

Table 2. Constants for the calculation of molecular diffusion coefficients (Latil, 1991).

Component	$\sigma$ ( $\text{\AA}$ )	$\varepsilon/k$ (K)
n-Pentane (C5)	5.784	341.1
Methane (C1)	3.758	148.6
Nitrogen (N2)	3.798	71.4

Application of Eq. (25) for the ternary gaseous mixture of pentane ( $i=1$ ), methane ( $i=2$ ) and nitrogen ( $i=3$ ) at  $38.5^\circ\text{C}$  and  $101$  bar abs yielded the values:  $\mathcal{D}_{12}^V = 9.067 \times 10^{-8}$  ( $\text{m}^2/\text{s}$ ),  $\mathcal{D}_{13}^V = 8.912 \times 10^{-8}$  ( $\text{m}^2/\text{s}$ ) and  $\mathcal{D}_{23}^V = 23.955 \times 10^{-8}$  ( $\text{m}^2/\text{s}$ ).

### 2.4 Molecular coefficients for a concentrated liquid mixture

Differently from the case of the ideal gaseous mixtures, the molecular coefficients for the liquid mixtures can be strongly dependent of the concentration. According to Taylor and Krishna (1993), in a concentrated liquid mixture, this dependence can be well expressed by:

$$\mathcal{D}_{ij}^L = \left(\mathcal{D}_{ij}^{L,\infty}\right)^{\frac{1+x_j-x_i}{2}} \left(\mathcal{D}_{ji}^{L,\infty}\right)^{\frac{1+x_i-x_j}{2}} \quad (27)$$

where  $x_i$  is the molar fraction of component “ $i$ ” in the mixture and  $\mathcal{D}_{ij}^\infty$  is the coefficient of molecular diffusion of component “ $i$ ” in “ $j$ ” when  $i$  is infinitely diluted in  $j$ .

For the evaluation of the binary molecular coefficients in a liquid mixture at high dilution state, Taylor and Krishna suggest the use of the semi-empirical correlation proposed by Wilke and Chang (1955):

$$\mathcal{D}_{ij}^{L,\infty} = 7.4 \times 10^{-8} \frac{(\phi_j M_j)^{1/2} T}{\mu_j (v_i^{boil})^{0.6}} \quad (28)$$

In Eq. (28),  $\mathcal{D}_{ij}^{\infty}$  is the coefficient (cm<sup>2</sup>/s);  $M_j$  is the molar mass of the solvent (g/mol);  $T$  is the temperature (K);  $\mu$  is the viscosity of the solvent (mPa.s);  $v_i^{boil}$  is the molar volume of the solute  $i$  at his normal boiling point (at 1 atm) (cm<sup>3</sup>/mol);  $\phi_j$  is an association parameter for the solvent, whose values are 2.26 for water and 1.0 for non-polar solvents like hydrocarbons.

## 2.5 Fick's coefficients for diffusion in non-ideal fluids

Combining Eqs. (4) and (13), Fick's coefficients for ternary diffusion, corrected to take into account the thermodynamical non-idealities of the real mixture,  $D^{ni}$ , can be written as a function of the binary (molecular) coefficients and of the composition of the mixture as:

$$[D^{ni}] = [D^{id}] [\Gamma] \quad (29)$$

which means, explicitly, for  $i=1,2$  and  $j=1,2$ :

$$D_{ij}^{ni}(x_1, x_2) = D_{i1}^{id} \Gamma_{1j} + D_{i2}^{id} \Gamma_{2j} \quad (30)$$

So, the characterization of Fick's coefficients for the ternary case reduces to the calculation of 4 molecular coefficients for the composition of the ideal matrix  $D^{id}$  of the vapor phase and 4 for the liquid phase. Then, when the 2 ideal matrices are assembled, the 2 fugacity coefficients ( $\phi_i$ ) for the vapor phase and 2 for the liquid phase must be calculated, for the composition of the correction matrix  $\Gamma$ . Equation (29) completes the operation.

## 3. RESULTS

With the formulation described above, ternary diffusion coefficients were calculated for a liquid-vapor mixture of C5-C1-N2 ( $i=1,2,3$ , respectively) at 38.5°C and 101 bar abs, assuming thermodynamical equilibrium at the interface. The hypothesis of equilibrium at the interface restricts the range of values that can be assumed by the molar concentrations of both phases. As shown in Fig. 1, depending on the combination of the three molar concentrations, one point in the ternary diagram of equilibrium may fall in one of three regions: the vapor region, the liquid region and liquid-vapor equilibrium region. Values of the diffusion coefficients need only to be calculated for the respective region.

Some results for the vapor phase are shown in Fig. 2, for  $y_{C5}=0.033$  ( $0 \leq y_{C1} \leq 0.945$ ). In a real diffusion problem in a reservoir, these variables are limited by the injected gas condition (pure nitrogen) on one side and by the equilibrium at the interface with the liquid on the other side. So, variable  $y_{C5}$  is fixed to fall into the vapor region and variable  $y_{C1}$  is determined in order to respect the equilibrium curve corresponding to the vapor in the ternary diagram. From this figure, it is apparent that coefficient  $D_{12}$  is always very small compared to the others and can be neglected, while the other crossed coefficient,  $D_{21}$ , is significant and must be considered. The graphic shows also the importance of taking into account the non-idealities: the ideal model would be acceptable for the calculation of  $D_{12}$  and  $D_{22}$ , but  $D_{11}$  and  $D_{21}$  must be corrected. Besides, it is clear that coefficients  $D_{11}$ ,  $D_{12}$  and  $D_{22}$  present very little variation in all the relevant range, while coefficient  $D_{21}$  undergo a strong variation with  $y_{C1}$ .

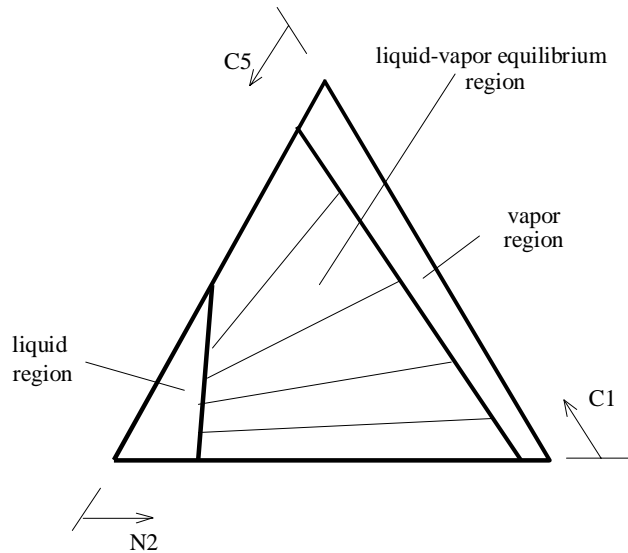


Figure 1 - Ternary diagram for C5-C1-N2 (P=101 bar abs., T=38.5°C).

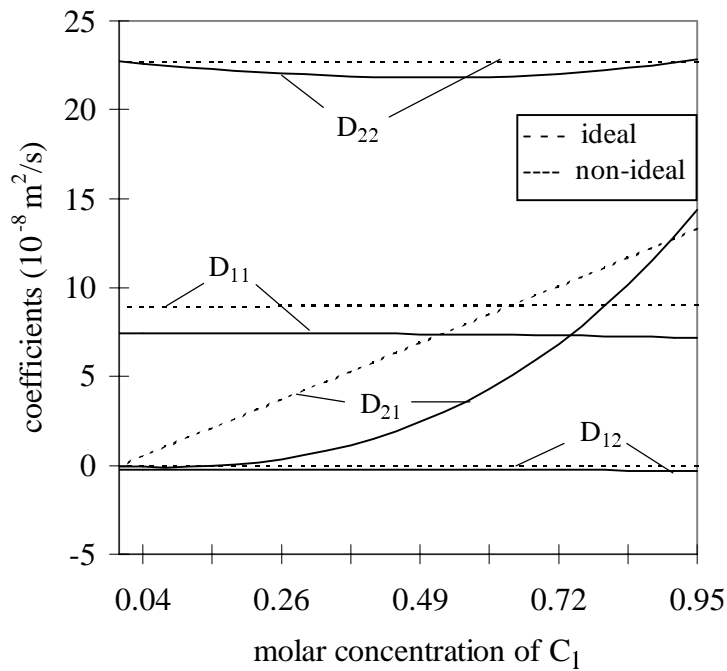


Figure 2 - Fick's coefficients for diffusion in the vapor phase of a liquid-vapor mixture of C5-C1-N2 ( $y_{C5} = 0.033$ ).

In the liquid phase, the relevant values of molar concentration are limited by ternary equilibrium condition at the interface with the vapor and by the condition that corresponds to the binary equilibrium C5-C1, found before N<sub>2</sub> injection. Figure 3 shows the values of the coefficients for  $x_1=0.717$  and  $0.195 \leq x_2 \leq 0.283$  (for situating in the relevant region). Like the vapor case, D<sub>12</sub> is the less important coefficient, and, taking non-ideal results, it represents no more than 20% of the leading coefficient D<sub>11</sub>. Also evident from the figure is the strong variation with concentration of all the coefficients. Besides, figure 3 shows, as expected, that the thermodynamical corrections in the liquid phase lead to much more significant differences between ideal and non-ideal models than in the vapor phase.



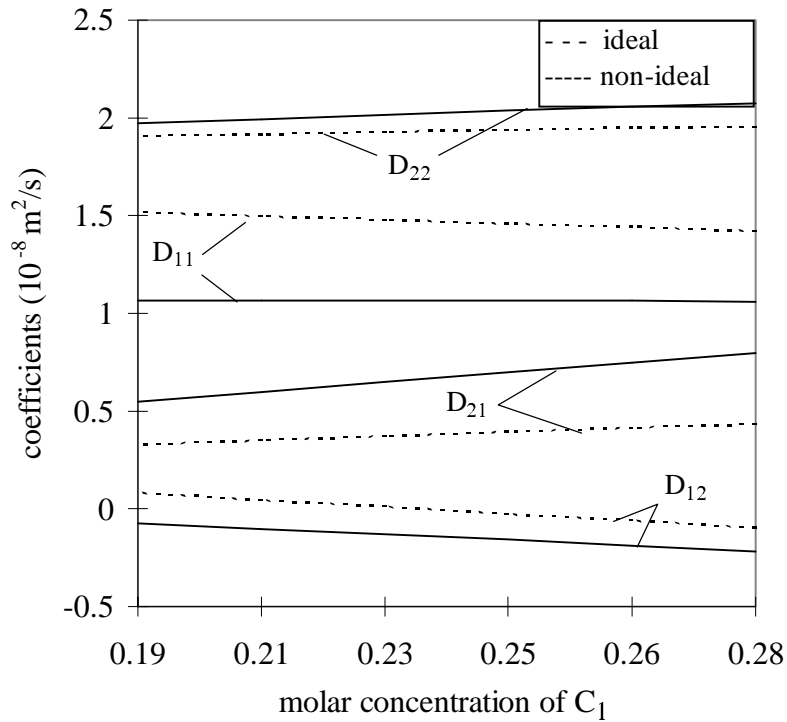


Figure 3 - Fick's coefficients for diffusion in the liquid phase of a liquid-vapor mixture of C5-C1-N2 ( $x_{CS}=0.717$ ).

As a final observation, Fig. 2 indicates that the diffusion coefficients in the vapor phase have an order of about  $10^{-7}$ , while Fig. 3 denotes that in the liquid phase these coefficients are of order  $10^{-8}$ . Hence, the diffusion coefficients in the vapor phase are roughly 10 times bigger than those in the liquid phase. This is not enough to take the process in the vapor as quasi-static in comparison to a transient in the liquid, as is currently done, for example, at normal atmospheric conditions (Jaffrenou, 1995), where vapor coefficients can be of the order of a thousand times the liquid coefficients.

#### 4. CONCLUSION

This work discussed the modeling of Fick's coefficients for ternary diffusion. Such an analysis is essential for the numerical simulation of light oil secondary exploitation in fractured reservoirs by means of evaporation. These coefficients are usually determined with the aid of experiments, very complicated in the conditions of a real reservoir, or with the application of excessively simplified models, which treat liquids as perfect gases.

Applying thermodynamical corrections to the expressions derived from the Kinetic Theory of Gases, it was possible to obtain theoretically the values of Fick's coefficients for the mixture C5-C1-N2 under the conditions of a typical reservoir (101 bar abs and 38.5°C). Analysis of the results revealed that from the four coefficients associated with the vapor phase,  $D_{12}$  could be neglected in the use of Fick's law for both phases. The other coefficients are all important and must be considered.

Numerical results allowed also to quantify the error committed in considering the fluids in a reservoir at high-pressure as perfect gases. In a general way, the perfect gas model can only be used as the starting point for the determination of more realistic coefficients. As a final conclusion, the results obtained indicate that, at high-pressure conditions, Fick's coefficients for the vapor are roughly ten times bigger than those for the liquid.

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