Modeling the Viscosity for (nC$_5$+nC$_8$), (nC$_5$+nC$_{10}$), (nC$_8$+nC$_{10}$) and (nC$_5$+nC$_8$+nC$_{10}$) Systems with Peng-Robinson Viscosity Equation of State

Shahin Khosharay[1*], Reza Karimi[2], Khashayar Khosharay[3]

Abstract

The aim of this modeling study is to improve the performance of the Peng-Robinson viscosity equation of state. To achieve this aim, the couple of Peng-Robinson viscosity equation of state and the proposed mixing rules has been applied for modeling the viscosities of the binary and ternary systems containing (nC$_5$+nC$_8$), (nC$_5$+nC$_{10}$), (nC$_8$+nC$_{10}$) and (nC$_5$+nC$_8$+nC$_{10}$) for temperatures and pressures ranged (297.75-373.35) K and (49.95-246.26) bar, respectively. First, the pressure temperature-dependent and constant expressions for the binary interaction coefficients of binary systems have been determined. Subsequently, these empirical correlations of binary interaction coefficients have been applied to predict the viscosities for ternary mixture of (nC$_5$+nC$_8$+nC$_{10}$). For this ternary mixture, the results of model show acceptable accuracy (overall AAD~7.77 % and 8.02 for mixing rule 1 and 2, respectively).

[1] School of Chemical, Gas and Petroleum Engineering, Semnan University, Semnan, Iran

[2] Department of Chemical Engineering, Faculty of Engineering, University of Isfahan, Isfahan, Iran

[3] Department of Computer Science, College of Engineering, Buinzahra Branch, Islamic Azad University, Buinzahra, Iran

[*] Corresponding author, e-mail: sh.khosharay@semnan.ac.ir

1 Introduction

Viscosity is a transport property which can be related to the strength of the forces acting between the molecules. Viscosity modelling is of high importance in many engineering purposes such as designing petrochemical equipments, the underground movement of the oil, heat and mass transfer rate and the numerical modeling of fluid flow. Since viscosity is a significant parameter in diffusion analysis and the hydrodynamic processes modeling require the accurate dynamic viscosities, the precise values of viscosity over wide ranges of pressure and temperature are of fundamental importance. Although the reliable viscosities can be determined with the experimental methods, it is not possible for researchers to determine the viscosity values for all mixtures by using experimental measurements (especially at high pressures); therefore, according to this limitation, the viscosity must be described by applying numerical simulation methods [1,2].

Several methods exist for modeling the viscosities of the mixtures such as the empirical correlations [3] and thermodynamical methods [4,5,6]. The empirical correlation of Lohrenz et al. [3] and the corresponding states model of [7] are conventional approaches to describe the viscosity of hydrocarbon mixtures. The narrow ranges of the thermodynamic conditions in their application and the requirement of fluid densities to calculate viscosities are two main limitations of these methods hindering their usage. On the other hand, the statistical physics approaches are very complicated and they are insufficiently developed to describe the viscosity of mixtures. According to the similarities between the *PvT* and *TμP* relationships and diagrams, several researchers have developed the viscosity equations of state. The viscosity equations of state can be applied for pure components and their mixtures at high and low pressures. The main advantage of using viscosity equations of state is that no fluid density is needed for modeling the viscosity [4]. In recent years, several researches have been conducted on modeling the viscosity of hydrocarbons and their mixtures. Khosharay [6] proposed a mixing rule for the viscosity based Peng-Robinson equation of state to compute the viscosity of light liquid hydrocarbon mixtures. The results of this model

proved that it worked well for binary, ternary, quaternary and five-component hydrocarbon mixtures (AAD=4.83 %). Fan and Wang [4] suggested the Peng-Robinson model to describe the viscosity for pure light hydrocarbons and their mixtures. The proposed model was successfully applied for computing viscosities of light hydrocarbons and their mixtures (AAD=9.47 %). Wang et al. [8] utilized the PR viscosity EOS for hydrocarbons containing heptane, octane, nonane, hexyl benzene and their ternary mixtures. The results of the proposed model showed the good performance of model for specified systems (AAD=0.13 %). Guo et al. [9] suggested two viscosity EOSs containing Petal–Teja and PR EOSs. Guo et al. [10] used Peng-Robinson viscosity EOS for pure hydrocarbons and their mixtures. This model could be applied for both high and low pressures. Derevich [5] applied a thermodynamic model for describing the viscosities of pure hydrocarbons and their mixtures. This model successfully reproduced the experimental viscosities not only pure components in vapor and liquid phases but also solutions in the wide ranges of thermodynamic conditions with AAD of 6 %. Tan et al. [11] applied Friction theory in combination with statistical associating fluid theory (SAFT1 and PC-SAFT) to model the viscosities of *n*-Alkane mixtures. Based on the results of the model, the accuracy was adequate for engineering applications. Lawal [12] proposed a four-parameter viscosity EOS based on Lawal–Lake–Silberberg model for pure hydrocarbons and their mixtures.

Since the applied model in the work of Khosharay [6] can be applied for limited light liquid hydrocarbon mixtures, in this modeling investigation, two suggested mixing rules have been coupled with the PRμ model. In these two mixing rules, the binary interaction parameters have been considered, taking advantage of the well described viscosity. These binary interaction parameters have been determined based on the experimental viscosities for binary mixtures of the (nC_5+nC_8), (nC_5+nC_{10}), (nC_8+nC_{10}) binary systems. These binary interaction parameters have been utilized for modeling the viscosities of ($nC_5+nC_8+nC_{10}$) ternary liquid hydrocarbon system. The performance of this model has been compared with the previous model [6].

2 Brief description of applied model
2.1 PRμ₀ model

The PRμ$_0$ model has been described extensively in several modeling investigations [4,6,8] so that the description of this model is limited to the important parts of this model. It is known that the PvT and $T\mu P$ relationships and diagrams are similar, so the positions of T and P in the PR EOS can be changed and v can be substituted with μ. Subsequently, gas constant R can be substituted with (defined subsequently) [4,8]. The Peng-Robinson viscosity based equation of state is expressed as follows:

$$T' = \frac{R'P}{\mu - b} - \frac{a}{\mu^2 + 2\mu b - b} \tag{1}$$

In Eq. (1), a presumptive temperature, T', is computed by using following equations:

$$T' = T - T_d \tag{2}$$

$$T_d = 0.45T_c \tag{3}$$

where P shows the pressure, T is the temperature of system, μ represents the viscosity and subscript c is the critical state. According to the minimum deviation of the modeled viscosities from the experimental viscosities, the coefficient of 0.45 has been chosen for T_d [4].

The coefficients of a, b and r_c are computed as follows:

$$a = 0.45724 \frac{(r_c P_c)^2}{T'_c} \tag{4}$$

$$b = 0.0778 \frac{r_c P_c}{T'_c} \tag{5}$$

$$r_c = \frac{\mu_c T'_c}{0.3074 P_c} \tag{6}$$

Based on the [13], the critical viscosity, μ_c, is calculated in terms of critical temperature (T_c), critical pressure (P_c) and molecular weight (M_W)

$$\mu_c = 7.7 T_c^{-1/6} M_W^{1/2} P_c^{2/3} \tag{7}$$

In Eq. (7), μ_c has been expressed in terms of micropoise. In Eq. (1), R' is computed as follows:

$$R' = \beta(P) r_c \tag{8}$$

In Eq. (8), $\beta(P)$ is a function of pressure. The value of this parameter is 1 at critical pressure and it is determined as follows:

$$\beta(P) = e_0 \left(1 - P_r^{-1}\right) - 0.02715 P_r^{-1} \left((P_r + 0.25)^{-1} - 0.8\right) + P_r^{-1} \tag{9}$$

In Eq. (9)

$$e_0 = 0.03192 - 3.3125 \times 10^{-4} M_W \omega \tag{10}$$

The above viscosity model developed from PR EOS is named PRμ$_0$ model.

2.2 PRμ model

The PRμ$_0$ model poorly represented liquid viscosity so that the correction of viscosity must be applied. This correction is written as follows:

$$\mu = \mu^{PR} + c_0 + c \tag{11}$$

In Eq. (11), μ^{PR} shows the viscosity that is calculated by using Eq. (1) and c_0 shows the parameter which is a function of reduced pressure.

$$c_0 = 6.714(P_r - 1) - 127.8((P_r + 1)^{-1} 0.5) \qquad (12)$$

In Eq. (11), the parameter of c is a function μ^{PR} and it can be expressed by Eq. (21).

$$\mu_r = \frac{\mu^{PR}}{\mu_c} \qquad (13)$$

$$c = e_3 \ln \mu_r + e_4 \left((\mu_r + 1.25)^{-1} - 0.4444 \right) \qquad (14)$$

The parameters of e_3, e_4 and e_7 are functions of molecular weight and acentric factor and they are computed as follows:

$$e_5 = 3337.201 - 717.955 M_W \omega \qquad (15)$$

$$e_5 = 17000 \qquad \omega \geq 0.3 \qquad (16)$$

$$e_3 = 216.643 + 0.231 e_5 \qquad \omega < 0.3 \qquad (17)$$

$$e_3 = 4130.636 \qquad (18)$$

$$e_4 = \left(\frac{e_5}{(e_7 + 1.25)^2} - \frac{e_3}{e_7} \right) \qquad (19)$$

In Eq. (19), e_7 is a function of acentric factor and can be calculated as follows:

$$e_7 = 1.767 + 18.384\omega - 32.728\omega^2 + 80.299\omega^3 \qquad (20)$$

This model of viscosity correction is PRµ viscosity model.

2.3 Mixing Rules

According to the modeling investigation of Fan and Wang [4], the viscosity equation of state can be applied for the mixtures by this mixing rule:

$$z_m = \sum_i z_i x_i \qquad z = a, b, c, r_c, T_d \text{ and } \beta \qquad (21)$$

Khosharay [6] suggested this mixing rule for light liquid hydrocarbon mixtures.

$$z_m = \sum_i \sum_j z_{ij} x_i x_j \qquad z = a, b, r_c, T_d \text{ and } \beta \qquad (22)$$

$$c_m = \sum_i c_i x_i \qquad (23)$$

In Eq. (22)

$$z_{ij} = \sqrt{z_i z_j} \qquad (24)$$

In this study, two mixing rules have been applied for viscosity equation of state. The first mixing rule (mixing rule 1) is expressed as follows:

$$z_m = \sum_i \sum_j z_{ij} x_i x_j \qquad z = a, b, r_c \qquad (25)$$

Also, this expression has been used to calculate z_m:

$$z_{ij} = \sqrt{z_i z_j} \qquad (26)$$

$$c_m = \sum_i c_i x_i \qquad (27)$$

Since the viscosity depends on the strength of the forces acting between the molecules, in this work, the binary interaction parameters is considered as an applicable factor for improving the performance of the present model. To achieve this aim, the binary interaction parameters of the model are used as follows:

$$T_{dm} = \sum_i \sum_j x_i x_j \sqrt{T_{di} T_{dj}} \left(1 - k_{ij} \right). \qquad (28)$$

$$\beta_m = \sum_i \sum_j x_i x_j \sqrt{\beta_i \beta_j} \left(1 - l_{ij} \right). \qquad (29)$$

In Eqs. (28) and (29), k_{ij} and l_{ij} are the binary interaction parameters. These parameters are determined according to the minimization of the average absolute deviation (AAD) of viscosity data for binary systems.

The second mixing rule (mixing rule 2) is suggested as follows:

$$z_m = \sum_i \sum_j z_{ij} x_i x_j \qquad z = a, b, c, r_c \qquad (30)$$

$$z_{ij} = \frac{\left((z_i)^{\frac{1}{3}} + (z_j)^{\frac{1}{3}} \right)^3}{8} \qquad (31)$$

$$T_{dm} = \sum_i \sum_j x_i x_j \frac{\left((T_{di})^{\frac{1}{3}} + (T_{dj})^{\frac{1}{3}} \right)^3}{8} \left(1 - k_{ij} \right) \qquad (32)$$

$$\beta_m = \sum_i \sum_j x_i x_j \frac{\left((\beta_i)^{\frac{1}{3}} + (\beta_j)^{\frac{1}{3}} \right)^3}{8} \left(1 - l_{ij} \right) \qquad (33)$$

Also, k_{ij} is considered as a constant parameter and l_{ij} have been considered as a pressure dependant parameter.

$$l_{ij} = a_0 + a_1 P + a_2 P^2 \qquad (34)$$

| Mixture | Mixing Rule 1 | | | | | Mixing Rule 2 | | | | |
	k_{ij}	$l_{ij} = a_0 + a_1P + a_2P^2$			AAD	k_{ij}	$l_{ij} = a_0 + a_1P + a_2P^2$			AAD
		$a_0{}^*10^6$	a_1	a_2			$a_0{}^*10^6$	a_1	a_2	
nC_5+nC_8	-0.07145	-12.51	0.003125	-0.2821	2.61 %	-0.0494	-15.58	0.00393	-0.3390	2.90 %
nC_5+nC_{10}	-0.1779	-9.761	0.001458	-0.03259	3.01 %	-0.0721	-6.884	0.00073	-0.3657	3.42 %
nC_8+nC_{10}	-0.07811	-11.31	0.001130	-0.00148	2.90 %	-0.0643	-10.25	0.00078	-0.0460	3.00 %

3 Results and discussions

3.1 Binary mixtures

In this section, three binary mixtures have been considered. First of all, the binary mixture of (nC_5+nC_8) has been considered. The acentric factor and critical properties are taken from [14]. The experimental viscosities of this binary mixture are given from the experimental investigation of Barrufet et al. [15]. The experimental viscosities used for modeling purpose are in the temperatures ranged (297.95-373.35) K and pressures ranged (1.01-246.26) bar. The composition of this mixture is in the range of (10.48-89.46) mole% of nC_5. A part of experimental viscosities and modeling results (by using mixing rule 1) are reported in Fig. 1. The AAD of the viscosities are calculated based on the following equation:

$$AAD = \frac{1}{N_P} \frac{\mu_i^{exp} - \mu_i^{calc}}{\mu_i^{exp}} \times 100 \qquad (35)$$

The overall absolute deviations of the applied model for (nC_5+nC_8) mixture are 2.61 % and 2.90 % for mixing rule 1 and mixing rule 2, respectively. The binary interaction parameters of the model are presented in Table 1. To our knowledge, the AAD of this system was 9.8 % by using PRVIS model [16] in previous work. The results prove that the application of binary interaction parameter is necessary for PR viscosity equation of state.
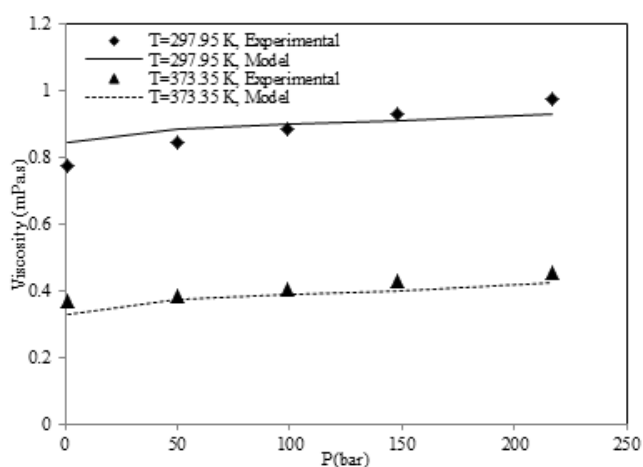
Fig. 1 Viscosity versus pressure for the binary mixture of
(0.8946 nC_5+0.1054 nC_8)

The second binary mixture considered in this study is (nC_5+nC_{10}) mixture. The experimental viscosity data have been given from the experimental study of Estrada-Baltazar et al. [17]. The modeling study has been conducted for viscosity data in the temperatures range of (297.95-373.35) K and pressure range of (1.01-246.26) bar. The composition of this binary system changes from 10.31 to 90.08 mole% of nC_5. As it is shown in Table 1, the average absolute deviations of the present model for (nC_5+nC_{10}) mixture are 3.01 % and 3.42 % by using mixing rule 1 and mixing rule 2, respectively. Also, Fig. 2 compares the experimental and modeled viscosities for (nC_5+nC_{10}) mixture by using mixing rule 1.
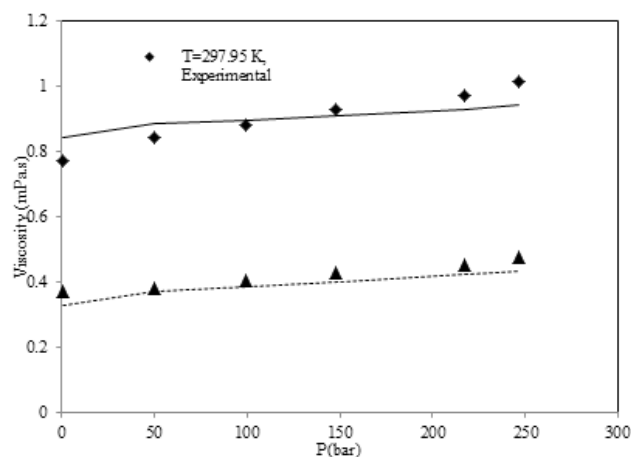
Fig. 2 Viscosity versus pressure for the binary mixture of
(0.1031 nC_5+0.8969 nC_{10})

The third binary system studied in this work contains (nC_8+nC_{10}). The experimental viscosity data for this binary mixture are from Estrada-Baltazar et al. [18]. The present model is used in order to reproduce viscosities in the temperatures ranged (297.95-373.35) K and pressures ranged (1.01-246.26) bar. The composition of this binary system is in the range of (13.84-88.58) mole% of nC_8. Similar to two previous binary mixtures, the adjustable parameters (l_{ij} and k_{ij}) are determined for this binary system and they have been shown in Table 1. The experimental data and modeling results (by using mixing rule 1) are compared in Fig. 3. The average absolute deviations

of the viscosity equation of state for (nC$_5$+nC$_{10}$) mixture are 2.90 % and 3.00 % in combination with the mixing rule 1 and mixing rule 2, respectively.
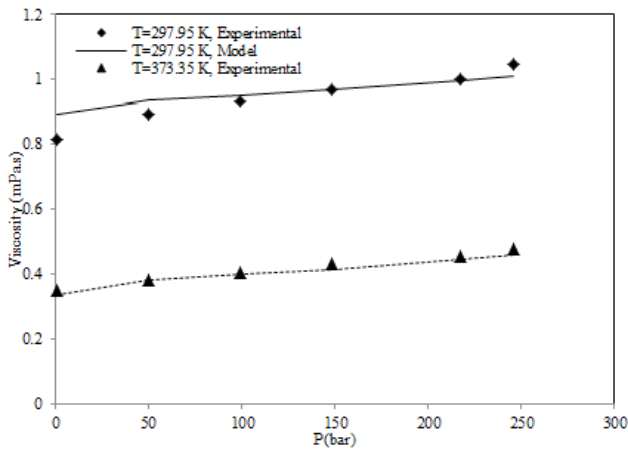
Fig. 3 Viscosity versus pressure for the binary mixture of (0.1384 nC$_8$+0.8616 nC$_{10}$)

In order to demonstrate the central hypothesis of this proposed mixing rule, the binary interaction coefficients of the present model are set to zero (k_{ij}=0 and l_{ij}=0). The results reported in Table 2 indicate that the present model is not reliable for (nC$_5$+nC$_8$), (nC$_5$+nC$_{10}$) and (nC$_8$+nC$_{10}$) binary systems without binary interaction coefficients. Also, the AAD of PRVIS model [16] was 5.69 % and 12.7 % for second and third binary systems, respectively.

Table 2 Comparison of the applied mixing rules with and without binary interaction coefficients (AAD)

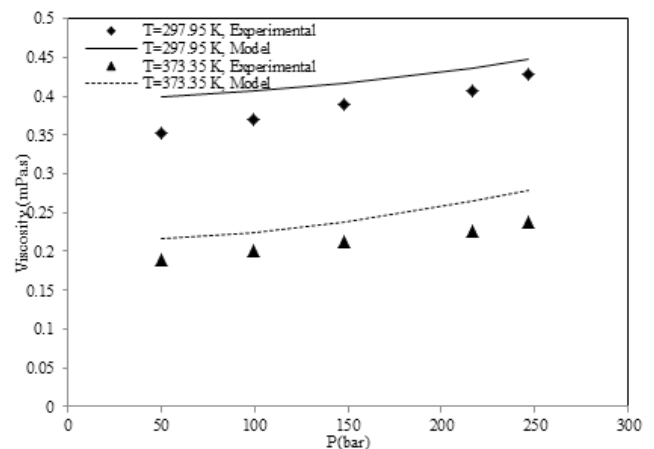| Mixture	N_P	Mixing Rule 1		Mixing Rule 2	
		k_{ij} & l_{ij}=0	k_{ij} & $l_{ij}\neq$0	k_{ij} & l_{ij}=0	k_{ij} & $l_{ij}\neq$0
nC$_5$+nC$_8$	295	8.25 %	2.61 %	8.54 %	2.90 %
nC$_5$+nC$_{10}$	312	14.74 %	3.01 %	15.35 %	3.42 %
nC$_8$+nC$_{10}$	324	10.36 %	2.90 %	10.37 %	3.00 %

3.2 The (nC$_5$+nC$_8$+nC$_{10}$) ternary system

In the previous part of this study, the results obtained for the (nC$_5$+nC$_8$), (nC$_5$+nC$_{10}$) and (nC$_8$+nC$_{10}$) binary systems prove that the viscosity equation of state in combination with the proposed mixing rules works well for these binary systems. Hence, the present model can be applicable for (nC$_5$+nC$_8$+nC$_{10}$) ternary mixture. The experimental viscosities of this ternary system are given from the experimental investigation of Iglesias-Silva et al. [19] who have measured the liquid viscosity for (nC$_5$+nC$_8$+nC$_{10}$) ternary mixture in the temperatures ranged (297.95-373.35) K and pressures ranged (1.01-246.26) bar for 530 points. The compositions of this mixture are represented in

Table 3. In this section, the adjustable parameters of the model (l_{ij} and k_{ij}) obtained in previous section have been applied in order to predict the viscosities of (nC$_5$+nC$_8$+nC$_{10}$) ternary mixture. The pressure and temperature range of experimental data are given in Table 3. Also, a part of experimental viscosities and modeling results (by using mixing rule 1) are reported in Fig. 4. The average absolute deviations of these systems are 7.77 % and 8.02 % for the mixing rule 1 and mixing rule 2, respectively; therefore, the present model performed well for these two ternary mixtures. The AAD of PRVIS model [16] was 7.41 % for this system. The AAD of previous model is 10.1 % for this system.

Table 3 Components of (nC$_5$(1)+nC$_8$(2)+nC$_{10}$(3)) ternary mixture

x_1	x_2	T range (K)	P range (bar)
0.6014	0.2505	297.79-373.35	1.01-246.26
0.1489	0.7033	297.79-373.35	1.01-246.26
0.3008	0.5425	297.79-373.35	1.01-246.26
0.4487	0.3989	297.79-373.35	1.01-246.26
0.7507	0.1001	297.79-373.35	1.01-246.26
0.1499	0.5407	297.79-373.35	1.01-246.26
0.3008	0.4001	297.79-373.35	1.01-246.26
0.4483	0.2488	297.79-373.35	1.01-246.26
0.5994	0.0998	297.79-373.35	1.01-246.26
0.1498	0.4013	297.79-373.35	1.01-246.26
0.3002	0.2494	297.79-373.35	1.01-246.26
0.4485	0.0999	297.79-373.35	1.01-246.26
0.1502	0.2500	297.79-373.35	1.01-246.26
0.3007	0.0996	297.79-373.35	1.01-246.26
0.1501	0.0994	297.79-373.35	1.01-246.26

Fig. 4 Viscosity versus pressure for the ternary mixture of (0.6014 nC$_5$+0.2505 nC$_8$+0.1481 nC$_{10}$)

4 Conclusions

The coupling of the Peng-Robinson viscosity equation of state and two proposed mixing rules have been applied for describing the viscosities of (nC_5+nC_8), (nC_5+nC_{10}), (nC_8+nC_{10}) and ($nC_5+nC_8+nC_{10}$) mixtures. Also, the pressure dependent and constant expressions for the binary interaction coefficients of binary systems containing (nC_5+nC_8), (nC_5+nC_{10}), (nC_8+nC_{10}) are determined, taking the advantage of well described viscosity for these binary systems. The obtained binary interaction coefficients are applied for describing the viscosity of ($nC_5+nC_8+nC_{10}$) mixture. The results of the model prove that the present model successfully describes the viscosities of these mixtures.

Nomenclature

a	energy parameter for PR equation of state ($K.s^2.Pa^2$)
b	volumetric parameter for PR equation of state (Pa.s)
c, c_0	parameters of viscosity
e	constant parameters of the model
M_w	molecular mass (gr/mol)
N_P	number of experimental data
P	pressure (bar)
r_c	parameter of the PRμ model (K.s)
R	universal gas constant (Jmol/K)
R'	parameter of the PRμ model (Jmol/K)
T	Temperature (K)
T_d	a specific temperature for calculated viscosities (K)
T'	a presumptive temperature (K)
v	molar volume (mol/m^3)
x	liquid mole fraction
PR	Peng Robinson
EOS	Equation Of State
$SAFT$	Statistical Associating Fluid Teory
PC-$SAFT$	perturbed-chain statistical associating fluid theory

Greek letters

$\beta(P)$	pressure dependent function in PRμ model
μ	dynamic viscosity (mPa s)
ω	acentric factor

Subscripts

c	critical property
m	mixture property
r	reduced property

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