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Modeling Viscosity of Alkanolamine Aqueous Solution Using the Friction Theory and the Cubic Plus Association Equation of State

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Abstract

Friction theory is one of the suitable theories for calculation of the viscosity. Using the theory and an appropriate equation of state (EoS), the viscosity could be estimated. Combination of the cubic-plus-association (CPA) EoS and the friction (f) theory called CPA ƒ-theory model can be applied for viscosity estimation of the aqueous solutions of alkanolamines. In this work, we investigated the capability of the CPA ƒ-theory model for evaluating viscosity of the aqueous solutions of alkanolamines: mono-ethanolamine (MEA), diethanolamine (DEA), and methyl diethanolamine (MDEA). The overall absolute average deviation (overall AAD) for the CPA ƒ-theory model in the temperature range of (293 – 353 K), mass fraction range of (0.10 – 0.90), and at atmospheric pressure for the aqueous solutions of MEA, DEA, and MDEA shows excellent accordant with experimental data and overall AAD for the aqueous solutions of MEA, DEA, and MDEA in the same temperature range and pressure range of (1 – 1200 bar), mass fraction range of (0.10 – 0.40) ,also shows very good accordant with experimental data.

Keywords*: Viscosity; Cubic-plus-association EOS; Alkanolamine; MEA; DEA; MDEA.*

1. Introduction

Alkanolamines are widely used in natural gas sweetening unit, oil refineries, petrochemical plants, and synthetic ammonia industry where aqueous solutions of alkanolamines are used to remove acid gases (hydrogen sulfide (H_2S) and carbon dioxide (CO_2)) from gas streams [1-3]. For the separation of acid gases from natural gases, several absorptions based methods such as amine, Sulfinol, and carbonate based processes have been introduced [4-6]. In CO_2 capture process, carbon dioxide can be captured from different gaseous sources according to its partial pressure, operating conditions and composition of the gas mixture. CO_2 capture process is very effective in reducing the amount of carbon dioxide from flue gases inpulverized coal (PC) and natural gas combined cycle (NGCC) power plants using the alkanolamines aqueous solutions such as MEA, DEA, and MDEA aqueous solutions [7]. In the mixed amines processes, a mixture of tertiary amines and secondary (or primary) amines are used for natural gas sweetening. In the mixed amines processes, synergy between methods of chemical absorption are exploited [8-14]. Single amine for gas treating is economically unfavorable due to high energy demand. Therefore, searching for alternative solvents is important. Blended amines can play an important role in gas absorption and desorption processes and properties of amines and blended amines including viscosity are important to understand the processes [15-17]. Viscosity measurement is generally costly and cannot be carried out for all fluids. Therefore, developing methods and models is of great interest [18].

Many researchers have developed different models for viscosity estimation. Most of these models are semi-theoretical methods that could be based on the corresponding states principles, the hard-sphere theory, the modified Chapman-Enskog theory, the empirical residual concept by Monnery *et al.*, equations of state (EOS), etc. Using the friction theory (f-theory), the basic principles of mechanics and thermodynamics are combined. In this theory, the viscosity of dense fluid is approached as a mechanical property rather than a transport property [19-21]. Thus, by combining the Amontons-Coulomb friction law and the van der Waals attractive and repulsive terms of a cubic EOS, viscosity is modeled for fluids in wide temperatures and pressures ranges. For aqueous solutions of alkanolamines due to the presence of OH group, a proper equation of state for combining with *f*-theory is required [22]. Oil and gas industries normally apply cubic equations of state and it is more convenient to use the CPA EOS. The CPA-EOS [23] generally is a good choice for modeling of systems consisting of OH groups in particular alkanolamine containing systems. Kontogeorgis *et al.* [24-26], Sobrino *et al.* [27], etc. investigated alkanolamine containing systems. In this work, we combined the *f*-theory and the CPA-EOS to model viscosities of aqueous solutions of three typical alkanolamines, i.e. monoethanolamine (MEA), diethanolamine (DEA), and methyl diethanolamine (MDEA). Satisfactory agreement is observed between literature data and modeling results.

2. Friction theory

In *f*-theory, viscosity of fluids mixture can be calculated as below [18,21,28]:

$$\eta = \eta_{\mathrm{o,mix}} + \eta_{f,\mathrm{mix}} \tag{1}$$

$$\eta_{\mathrm{o,mix}} = \exp[\textstyle\sum_{i=1}^{n} x_i \ln(\eta_{0,i})] \tag{2}$$

$$\eta_{f,\mathrm{mix}} = k_{\mathrm{r,mix}} P_{\mathrm{r}} + k_{\mathrm{a,mix}} P_{\mathrm{a}} + k_{\mathrm{rr,mix}} P_{\mathrm{r}}^{2} \tag{3}$$

where $k_{\mathrm{r,mix}}$, $k_{\mathrm{a,mix}}$, and $k_{\mathrm{rr,mix}}$ are the coefficients that are calculated as below [18,21,28]:

$$k_{\mathrm{r,mix}} = \textstyle\sum_{i=1}^{n} z_i \frac{\eta_{c,i}\hat{k}_{r,i}}{P_{c_i}} \tag{4}$$

$$k_{\mathrm{a,mix}} = \textstyle\sum_{i=1}^{n} z_i \frac{\eta_{c,i}\hat{k}_{a,i}}{P_{c_i}} \tag{5}$$

$$k_{\mathrm{rr,mix}} = \textstyle\sum_{i=1}^{n} z_i \frac{\eta_{c,i}\hat{k}_{rr,i}}{P_{c_i}^{2}} \tag{6}$$

2.1. Viscosity modeling

The viscosity modeling requires experimental viscosity data for tuning Kc parameter, which is a parameter that is tuned with the model of Uyehara and Watson for critical viscosity, given as [29]:

$$\eta_{\mathrm{c,i}} = K_c \frac{\sqrt{Mw} P_{c_i}^{2/3}}{T_{c_i}^{1/6}} \tag{7}$$

with replacement of Eq. (7) into the previous equations, the new equations can be obtained as follows [29]:

$$\eta = \eta_{\mathrm{I}} + K_c \eta_{\mathrm{II}} \tag{8}$$

$$\eta_{\mathrm{I}} = \eta_{\mathrm{o,mix}} + k_{\mathrm{r,I}} P_{\mathrm{r}} + k_{\mathrm{a,I}} P_{\mathrm{a}} + k_{\mathrm{rr,I}} P_{\mathrm{r}}^{2} \tag{9}$$

$$\eta_{\mathrm{II}} = k_{\mathrm{r,II}} P_{\mathrm{r}} + k_{\mathrm{a,II}} P_{\mathrm{a}} + k_{\mathrm{rr,II}} P_{\mathrm{r}}^{2} \tag{10}$$

$k_{\mathrm{r,I}}, k_{\mathrm{a,I}}, k_{\mathrm{rr,I}}, k_{\mathrm{r,II}}, k_{\mathrm{a,II}}$, and $k_{\mathrm{rr,I}}$ are specified as follows [29]:

$$k_{\mathrm{r,I}} = \textstyle\sum_{i=1}^{m} z_i \frac{\eta_{c,i} k_{r,i}}{P_{c_i}} \tag{11}$$

$$k_{\mathrm{a,I}} = \textstyle\sum_{i=1}^{m} z_i \frac{\eta_{c,i} k_{a,i}}{P_{c_i}} \tag{12}$$

$$k_{\mathrm{rr,I}} = \textstyle\sum_{i=1}^{m} z_i \frac{\eta_{c,i} k_{rr,i}}{P_{c_i}^{2}} \tag{13}$$

$$k_{\mathrm{r,II}} = \textstyle\sum_{i=m+1}^{n} z_i \left(\frac{\sqrt{Mw} P_{c_i}^{2/3}}{T_{c_i}^{1/6}}\right) \frac{k_{r,i}}{P_{c_i}} \tag{14}$$

$$k_{\mathrm{a,II}} = \textstyle\sum_{i=m+1}^{n} z_i \left(\frac{\sqrt{Mw} P_{c_i}^{2/3}}{T_{c_i}^{1/6}}\right) \frac{k_{a,i}}{P_{c_i}} \tag{15}$$

$$k_{\mathrm{rr},\mathrm{II}} = \sum_{i=m+1}^{n} z_i \left(\frac{\sqrt{Mw} P_{c_i}{}^{2/3}}{T_{c_i}{}^{1/6}}\right) \frac{k_{rr,i}}{P_{c_i}{}^{2}} \qquad (16)$$

for all components with i>m; and

$$z_{\mathrm{i}} = \frac{x_i}{M_{w_i}^{\varepsilon} MM} \qquad (17)$$

$$MM = \sum_{i=1}^{n} \frac{x_i}{M_{w_i}^{\varepsilon}} \qquad (18)$$

Using a least-squares method and experimental viscosity data, Kc can be determined by applying Eq. (8) [29]:

$$K_c = \frac{\eta_{exp} - \eta_I}{\eta_{II}} \qquad (19)$$

where experimental viscosity data are represented as η_{exp}[29].

$$k_{a,i} = k_a^c + \Delta k_{a,i} \qquad (20)$$

$$\Delta k_{a,i} = k_{a,0,0}(\Gamma_i - 1) + \left(k_{a,1,0} + k_{a,1,1}\psi_i\right)\left(e^{(\Gamma_i - 1)} - 1\right) + \left(k_{a,2,0} + k_{a,2,1}\psi_i + k_{a,2,2}\psi_i{}^2\right)\left(e^{(2\Gamma_i - 2)} - 1\right) \qquad (21)$$

$$k_{r,i} = k_r^c + \Delta k_{r,i} \qquad (22)$$

$$\Delta k_{r,i} = k_{r,0,0}(\Gamma_i - 1) + \left(k_{r,1,0} + k_{r,1,1}\psi_i\right)\left(e^{(\Gamma_i - 1)} - 1\right) + \left(k_{r,2,0} + k_{r,2,1}\psi_i + k_{r,2,2}\psi_i{}^2\right)\left(e^{(2\Gamma_i - 2)} - 1\right) \qquad (23)$$

$$k_{rr,i} = k_{rr}^c + \Delta k_{rr,i} \qquad (24)$$

$$\Delta k_{rr,i} = k_{rr,2,1}\psi_i\left(e^{(2\Gamma_i)} - 1\right)(\Gamma_i - 1)^2; \text{ and}; \qquad (25)$$

$$\psi_i = \frac{RT_{c,i}}{P_{c,i}} \quad (26); \qquad \Gamma_i = \frac{T_{c,i}}{T} \quad (27)$$

3. CPA equation of state

The cubic-plus-association equation of state [30-32] contains a fraction of cumulative sites of A on the *i*th molecule that have not formed any bonds (X_{A_i}) [33-37], cumulative power ($\Delta^{A_i B_j}$) and radial distribution function [31] as follows:

$$\mathrm{X}_{A_i} = \left[1 + \rho \sum_j X_j \sum_{Bj} X_B \Delta^{A_i B_j}\right]^{-1} \qquad (28)$$

$$\Delta^{A_i B_j} = g(\rho)\left[exp\left(\frac{\varepsilon^{A_i B_j}}{RT}\right) - 1\right] b_{ij} \beta^{A_i B_j} \qquad (29)$$

$$\mathrm{g}(\rho) = \frac{1}{1 - 1.9\mathrm{y}} \qquad (30)$$

$$\mathrm{y} = \frac{1}{4} b\rho \qquad (31)$$

Using the Elliott combining rule (ECR) [33]:

$$\Delta^{A_i B_j} = \sqrt{\Delta^{A_i B_i} . \Delta^{A_j B_j}} \qquad (32)$$

CR - 1 :

$$\varepsilon^{A_i B_j} = \frac{\varepsilon^{A_i B_i} + \varepsilon^{A_j B_j}}{2} \qquad (33)$$

$$\beta^{A_i B_j} = \sqrt{\beta^{A_i B_i} . \beta^{A_j B_j}} \qquad (34)$$

and

$$a = \sum \sum x_i x_j a_{ij} \qquad (35)$$

$$a_{ij} = \sqrt{a_i a_j}\left(1 - k_{ij}\right) \qquad (36)$$

$$b = \sum x_i b_i \qquad (37)$$

The thermodynamic properties of alkanolamines and water are reported in Table 1.

Table 1. Thermophysical properties of alkanolamines and water.

Parameter		Critical temperature	Critical pressure	Critical volume	Acentric factor	Molecular weight	Number of OH	Dipole moment
		K	bar	cm^3/mol		g/mol		
Ref.		[38]	[38]	[38]	[38]	[38]		[39]
MEA	C_2H_7NO	678.2	71.24	225	0.447	61.083	1	0.776
DEA	$C_4H_{11}NO_2$	736.6	42.7	349	0.953	105.136	2	0.851
MDEA	$C_5H_{13}NO_2$	675	38.8	368	1.165	119.163	2	2.86
Water	H_2O	647.13	220.55	55.95	0.3449	18.015	2	1.85

4. Results and discussion

Using the friction theory and the CPA equation of state at the same time, a model called CPA ƒ-theory was used for estimating the viscosity of aqueous solutions of MEA, DEA and MDEA, as mentioned earlier. The CPA EoS parameters for the studied systems are reported in Table 2. In the friction theory, 16 friction constants must be adjusted / optimized, which can be done using the least-squares method (Table 3), and the ε value was also adjusted. By examining the ε values, it was found that this value is in the range of 0-1. In this study, the best performance of the CPA ƒ-theory model was obtained at ε = 0.7 for MEA, DEA and MDEA aqueous solutions. The CPA equation of state is used to calculate the repulsive and attractive terms in the friction theory. In the CPA equation of state, it is necessary to select an appropriate association scheme. By selecting the association scheme and replacing the five CPA parameters and the k_{ij} value (Table 2) in the CPA equation of state, the repulsive and attractive terms required in the CPA ƒ-theory model are calculated. Also, to determine the properties of the mixtures, the CR-1 mixing rule was used in this work, which has shown better performance than ECR [30].

Table 2. The CPA EoS Parameters for alkanolamines and water with the 4C association scheme [30,33]

Parameter		b	a_0	c_1	ε	$\beta \times 10^{-3}$	k_{ij}
		L/mol	bar.L^2/mol^2		bar.L/mol		
MEA	C2H7NO	0.05656	14.112	0.7012	181.77	5.35	-0.165
DEA	C4H11NO2	0.09435	20.942	1.5743	161.59	33.2	-0.12
MDEA	C5H13NO2	0.11145	21.659	1.3371	161.59	33.2	-0.087
Water	H2O	0.0145	1.2277	0.6736	166.55	69.2	

k_{ij} values are for the aqueous solutions of alkanolamines (MEA + Water), (DEA + Water) and (MDEA + Water)

Table 3. The friction constants of the CPA ƒ-theory model for aqueous solutions of alkanolamines at atmospheric pressure

System	MEA + Water	DEA + Water	MDEA + Water
k_a^c	40913.1044	1718394.938	-793749.6483
$k_{a,0,0}$	292205.1602	-7556613.833	5428316.599
$k_{a,1,0}$	-223584.7457	3440996.32	-4586235.288
$k_{a,1,1}$	-166839.9537	987318.7961	245446.8973
$k_{a,2,0}$	20573.71997	22393.10005	434378.5686
$k_{a,2,1}$	-30753.84883	-184827.0286	315172.9498
$k_{a,2,2}$	70148.52858	-70905.49085	-227009.9631
k_r^c	39329.378	1668057.011	-795545.7691
$k_{r,0,0}$	299273.3222	-7357425.84	5425090.05
$k_{r,1,0}$	-229130.4989	3350251.854	-4572108.491
$k_{r,1,1}$	-166281.5481	963767.2827	239308.0129
$k_{r,2,0}$	-3647.314109	76361.85949	396377.5666
$k_{r,2,1}$	102433.6038	-435475.6373	484041.1417
$k_{r,2,2}$	-58580.71786	82482.13001	-325897.5348
k_{rr}^c	0.37274133	5.446554095	-0.604509994
$k_{rr,2,1}$	-0.01813896	-0.029779522	0.013347147

An important step in calculating the pressure through the CPA equation of state is to select an appropriate association scheme. MEA has a hydroxyl group and an amine group. It has two sites for hydroxyl and two sites for amine group. It is known that, in amine groups, the association force is weaker than alcohol, and by not considering the amine group and leaving only the hydroxyl group (for both sites) in MEA, it can be shown that the most suitable scheme for this arrangement is 4C scheme.

Study of DEA and MDEA reveals that they have two hydroxyl groups and one amine group and ignoring the amine group, it can be indicated that the most suitable scheme is 4C [33,18]. Considering the aforementioned explanations and using the CPA ƒ-theory model, the viscosity of aqueous solutions of the aforementioned alkanolamines can be calculated. The calculations were performed at atmospheric pressure and different mass fractions of alkanolamines in aqueous solutions. This model shows excellent accuracy and also the overall modeling results for most of the studied aqueous solutions overlaps with the experimental data. Figures 1 to 6 show that the viscosity of the studied aqueous solutions decreases with increasing temperature at atmospheric pressure and constant mass fraction. This trend can also be observed with increasing temperature for different mass fractions. Experimental data and the results of the CPA ƒ-theory model for MEA, DEA and MDEA aqueous solutions show a good agreement in a wide range of temperature and atmospheric pressure.

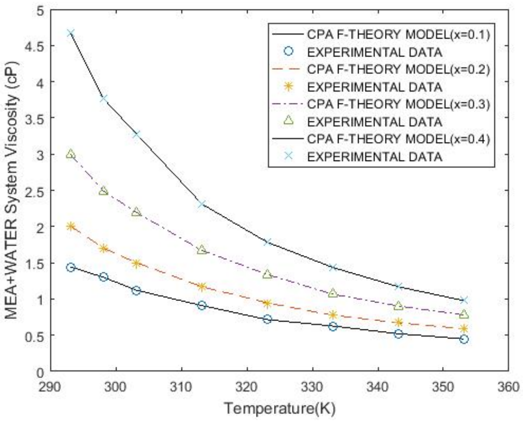

Fig. 1. Viscosity versus temperature for the MEA + WATER system in the mole fractions of 0.1-0.4. **[14,41]**

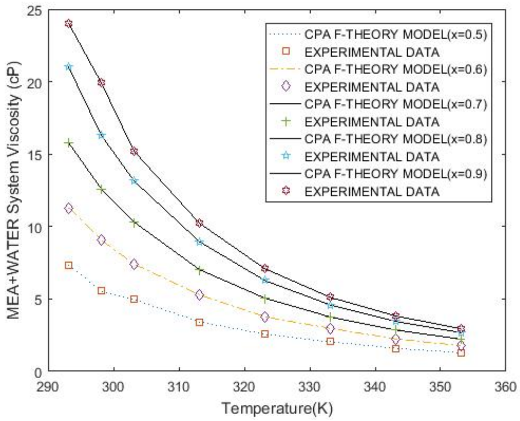

Fig. 2. Viscosity versus temperature for the MEA + WATER system in the mole fractions of 0.5-0.9 **[14,44]**

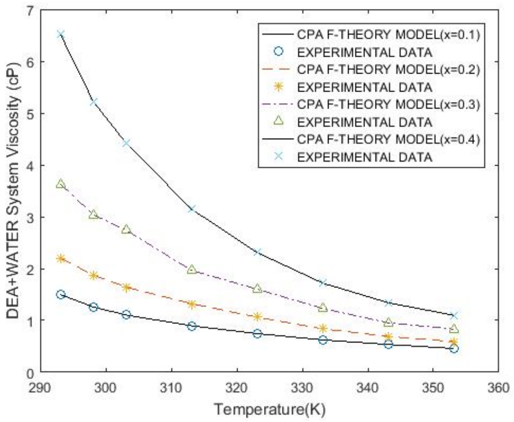

Fig. 3. Viscosity versus temperature for the DEA + WATER system in the mole fractions of 0.1-0.4 **[40-43]**

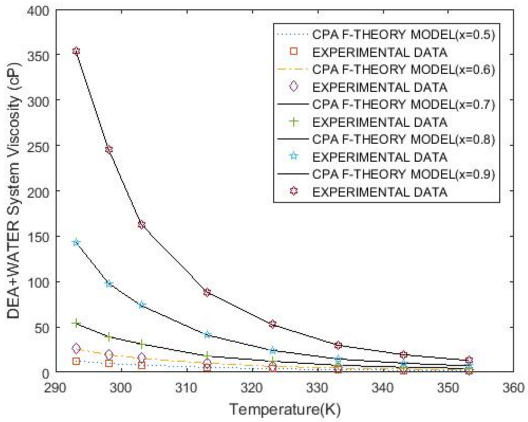

Fig. 4. Viscosity versus temperature for the DEA + WATER system in the mole fractions of 0.5-0.9 **[40-43]**

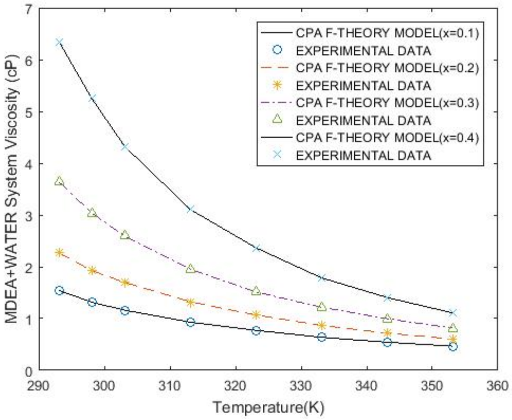

Fig. 5. Viscosity versus temperature for the MDEA + WATER system in the mole fractions of 0.1-0.4 **[14,41-43]**

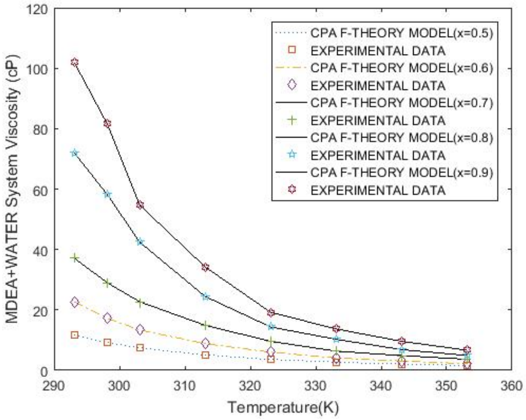

Fig. 6. Viscosity versus temperature for the MDEA + WATER system in the mole fractions of 0.5-0.9 **[14,41-43]**

In the next step, the effect of pressure changes (non-atmospheric pressure) on the results of the CPA ƒ-theory model for MEA and MDEA aqueous solutions was investigated. In this case, according to the above explanation, the ε and k_{ij} values must be calculated. The parameter ε was adjusted/optimized in the range of 0-1, as mentioned earlier. The performance of the CPA ƒ-theory model at ε = 0.6 was appropriate for MEA and the ε parameter for MDEA was suitable in all ranges from 0-1 except at 0.03, 0.4 and 0.7 mole fractions. Also, the k_{ij} values for the (MEA + WATER) and (MDEA + WATER) systems were considered -0.165 and -0.087, respectively. This study shows that the changes in the viscosity of the systems with the pressure are not significant, but the viscosity of these systems decreases with increasing temperature. Table 4 provides the friction constants calculated using the least-squares method for MEA and MDEA.

Table 4. The friction constants of the CPA ƒ-theory model for aqueous solutions of alkanolamines at different pressures

System	MEA + Water	MDEA + Water
k_a^c	17.98953128	32.23386025
$k_{a,0,0}$	-43.25813863	-140.2273141
$k_{a,1,0}$	27.16576377	94.93671101
$k_{a,1,1}$	-28.95624187	-7.708693164
$k_{a,2,0}$	-38.71496197	-0.046404238
$k_{a,2,1}$	198.3588555	-38.30048458
$k_{a,2,2}$	-184.74153	23.78973245
k_r^c	-565.811612	-180.3036954
$k_{r,0,0}$	3438.771214	1013.460406
$k_{r,1,0}$	-2462.226454	-783.6772805
$k_{r,1,1}$	-683.1837122	-225.9207697
$k_{r,2,0}$	135.1267175	163.674309
$k_{r,2,1}$	606.0455178	-356.7969721
$k_{r,2,2}$	-360.8958355	261.299556
k_{rr}^c	0.359880668	0.569953451
$k_{rr,2,1}$	-0.032800178	-0.021516944

Figures 7-10 show the performance of the CPA ƒ-theory model for aqueous solutions of MEA and MDEA. These diagrams show the effect of pressure on the estimation of viscosity, which indicate an excellent agreement in a wide range of temperature and pressure. Examination of Figures shows that the CPA ƒ-theory model can make an extraordinary estimation for aqueous solutions of alkanolamines. This model can be used for aqueous solutions of the studied alkanolamines in wide ranges of temperatures and pressures.

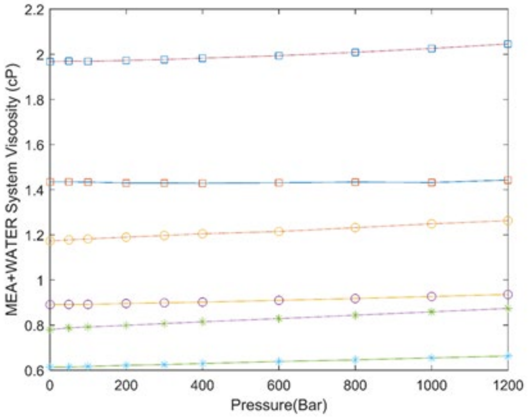

Fig. 7. Viscosity versus pressure for the MEA + WATER system in the mole fractions = 0.1 (solid line), 0.2 (dotted line) and temperature = 293 (square), 313 (circle), 333 (star), 353 (cross) **[14,41]**

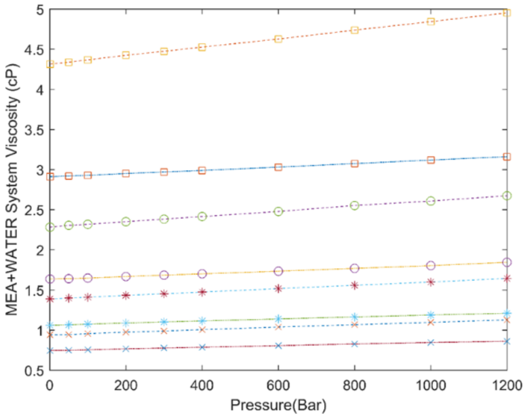

Fig. 8. Viscosity versus pressure for different the MEA + WATER system in the mole fractions = 0.3 (dashdot line), 0.4 (dashed line) and temperature = 293 (square), 313 (circle), 333 (star), 353 (cross) **[14,41]**

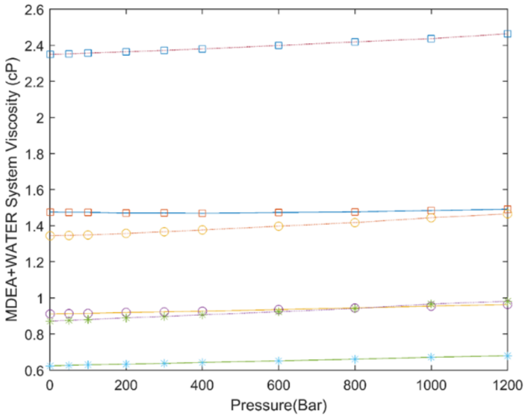

Fig. 9. Viscosity versus pressure for the MDEA + WATER system in the mole fractions = 0.1 (solid line), 0.2 (dotted line) and temperature = 293 (square), 313 (circle), 333 (star), 353 (cross) **[14,41-43]**

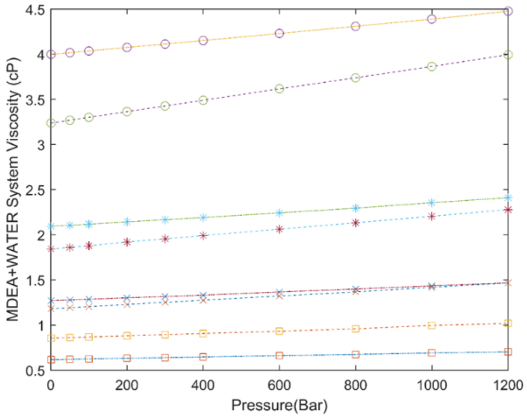

Fig. 10. Viscosity versus pressure for the MDEA + WATER system in the mole fractions = 0.3 (dashdot line), 0.4 (dashed line) and temperature = 293 (square), 313 (circle), 333 (star), 353 (cross) **[14,41-43]**

5. Conclusions

This study demonstrates the accuracy of the results of the CPA ƒ-theory model for estimating viscosity of aqueous solutions of alkanolamines in wide temperature and pressure ranges. The 4C scheme was evaluated and it was shown that it could be a suitable choice for aqueous solutions of the studied alkanolamines. With increasing temperature, the viscosity of aqueous solutions of the alkanolamines decreases, but with increasing pressure, there is no significant change in the viscosity of aqueous solutions of alkanolamines. On the other hand, it can be said that the ε value has no significant effect on estimating the viscosity of aqueous solutions of the studied alkanolamines.

Nomenclature

k_{ij}	*binary interaction parameter*
x_i	*mole fraction of component i [18]*
MM	*molecular weight (g/mol)*
M_w	*molecular weight (g/mol) [18]*
P_a	*attractive pressure term [18]*
P_r	*repulsive pressure term [18]*
P_c	*critical pressure (bar) [18]*
R	*gas constant (bar L/mol K) [18]*
T	*temperature (K) [18]*
T_c	*critical temperature (K) [18]*
K_c	*critical viscosity constant [19]*
b	*co-volume parameter (L/mol) [18]*
B_j	*site B in molecule j [18]*
g	*radial distribution function [18]*

Greek letters

η_c	*characteristic scaling viscosity defined in Eq. (7)*
ψ	*defined in Eq. (26)*
Γ	*defined in Eq. (27)*
η	*viscosity of mixture fluid*
η_0	*Mixed dilute gas viscosity [18]*
η_f	*Mixed residual friction term [18]*
ε	*mixing rule exponent*
ß	*association volume parameter [18]*
Δ	*association strength [18]*
ε^{AB}	*association energy parameter (bar L/mol) [18]*
λ	*reduced density [18]*
ρ	*molar density [18]*

Subscripts

c	*Critical [18]*
r	*Reduced [18]*

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