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CO_2 Capture, Transport and Storage

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Nils A. Røkke and Hanna Knuutila

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FREE ENERGIES OF ACTIVATION FOR VISCOUS FLOW OF DIFFERENT AMINE MIXTURES IN POST COMBUSTION CO_2 CAPTURE

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Abstract

The viscosity of ternary mixtures of N-methyldiethanol amine (MDEA) + monoethanol amine (MEA) + H_2O, N-methyldiethanol amine (MDEA) + diethanol amine (DEA) + H_2O and 2-amino-2-methyl-1-propanol (AMP) + diethanol amine (DEA) + H_2O were correlated using Eyring's viscosity model based on absolute rate theory. The correlations were capable of representing viscosity data within AARD 1.9%, 1.4% and 2.1% for the mixtures MDEA + MEA + H_2O, MDEA + DEA + H_2O and AMP + DEA + H_2O respectively. These accuracies are acceptable in engineering calculations. The excess properties of volume V^E, viscosity η^E and free energy of activation for viscous flow ΔG^{E*} were studied to understand the intermolecular interactions in the mixtures. The study shows that all mixtures have a negative sign for V^E, η^E and a positive sign for ΔG^{E*}. This indicates weak intermolecular interactions in mixtures compared to the pure liquids and strong molecular attractions like H-bonds in the mixtures.

Keywords: *Viscosity, Excess free activation energy, Eyring's viscosity model, Amines*

1. Introduction

The applicability of different amine mixtures to capture CO_2 in a post-combustion absorption and desorption process has gained interest during past years. The combined effect of higher equilibrium capacities of tertiary and sterically hindered amines with the fast reaction rates of primary and secondary amines make this technology more feasible for large scale implementations. MEA (monoethanol amine) is regarded as the benchmark solvent in Post Combustion Capture (PCC) as it shows high CO_2 absorption rate, is relatively cheap and is less harmful to the environment compared to other commercial amines in PCC [1]. The main disadvantage of MEA is the high energy demand for regeneration and that limits the use of MEA + H_2O mixture as a solvent. DEA (diethanol amine) is a secondary amine that exhibits high absorption rate [2]. The irreversible side reactions and the formation of corrosive products are the disadvantages of using DEA. A tertiary amine like MDEA (N-methyldiethanol amine) has a relatively low absorption rate and high absorption capacity compared to MEA. The reaction between MDEA and CO_2 has a low heat of reaction and reduces the energy penalty of the amine regeneration. AMP (2-amino-2-methyl-1-propanol) is a sterically hindered primary amine that has both acceptable absorption capacity, absorption rate and regeneration energy demand, which is suitable for PCC.

Some studies of blends of aqueous alkanolamines as solvents in acid gas treating have been reported in the literature. As the studies reveal, those blends can enhance the physicochemical properties compared to amine and water mixtures with one amine [3,4]. There, the primary or secondary amine is mixed with a tertiary amine and water. Primary and secondary amines enhance the absorption rate while tertiary amines increase the absorption capacity and reduce the regeneration energy.

Densities and viscosities are important for the design of process equipment due to the influence on flow behavior, typically in pumps and pipes. Densities and viscosities are also influencing the heat and mass transfer performance in heat exchangers, absorbers and stripper units. Especially the density and viscosity appear in correlations for estimating heat and mass transfer coefficients and interfacial areas in random and structured packings. Reduction of the uncertainty in estimation methods for the density and viscosity in mixtures will improve design methods considerably. Several studies have been reported in the literature regarding density and viscosity measurements for the mixtures of (MEA + MDEA + H_2O), (DEA + MDEA + H_2O) and (DEA + AMP + H_2O) under different amine concentrations and temperatures [5].

In this study, the Eyring's [6] absolute rate theory approach on dynamic viscosity of Newtonian fluids was considered to evaluate the free energy of activation for viscous flow of different amine solutions based on available density and viscosity data that have been published by Mandal *et al.* [5]. The excess volume and the excess viscosity were determined to analyze the intermolecular attractions of the mixtures.

2. Methodology

The excess free energies of activation were calculated and correlated according to a Redlich-Kister [7] type polynomial. Eyring's viscosity model for Newtonian fluids is given in Eq (1).

$$\eta = \frac{hN}{V} exp\left(\frac{\Delta G^*}{RT}\right) \tag{1}$$

Where, $\eta, V, \Delta G^*, h, N, R$ and T are dynamic viscosity, molar volume, free energy of activation for viscous flow, Planck's constant, Avogadro's number, gas constant and temperature respectively. In order to compare with ideal solutions and calculate the excess activation energy properties following Eq (2) and (3) are obtained by using Eq (1).

$$ln(\eta V) = ln(\eta V)_{ideal} + \frac{\Delta G^{E*}}{RT} \quad (2)$$

$$ln(\eta V) = \sum_i x_i ln(\eta_i V_i^o) + \frac{\Delta G^{E*}}{RT} \quad (3)$$

The excess free energy of activation for viscous flow ΔG^{E*} was determined from the density and viscosity data reported by Mandal *et al.* [5] as shown in Eq (3). A Redlich-Kister type polynomial with temperature dependency as given in Eq (5) and Eq (6) is proposed to correlate ΔG^{E*} of the amine mixtures according to the Eq (4). Here η_i and V_i^o are viscosity and molar volume of the pure liquids.

$$\frac{\Delta G^{E*}}{RT} = \Delta G_{12}^{E*} + \Delta G_{13}^{E*} + \Delta G_{23}^{E*} \quad (4)$$

$$\Delta G_{jk}^{E*} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i \quad (5)$$

$$A_i = a + b(T) + c(T)^2 \quad (6)$$

There are several ways to determine ideal viscosity contribution in a mixture [8-10]. The excess viscosity η^E as given in Eq (7) provides a quantitative approach to determine the deviation of the viscosity of a real mixture from its ideal conditions. The sign of η^E signifies the molecular interactions between unlike molecules in the mixture [11]. The molecular interaction between molecules in the mixture has a significant effect on viscosity. Heric and Brewer [12] explained, $\Delta G^{E*} > 0$ for a mixture that exhibits higher viscosities in the real mixtures than that of the ideal mixture.

$$\eta^E = \eta - \sum_{i=1}^{n} x_i \eta_i \quad (7)$$

A positive sign of η^E indicates that the mixture exhibits strong intermolecular interaction and negative sign specify weak interaction among the unlike molecules [13]. Intermolecular interaction is not the only aspect that influences the viscosity deviation of liquid mixtures [14, 15]. The excess molar volume of mixtures also can reveal intermolecular attractions in a liquid mixture [16]. The excess molar volume V^E represent the molar volume variation of a real mixture compared to its ideal condition. Three characteristics have been discussed in the literature that contribute to determining the sign of V^E. The mixtures having specific or chemical interactions including charge transfer, forming of H-bonds and other complex forming interactions provide a negative contribution for V^E. The molecules with different shape and size can rearrange within vacant spaces by giving a negative contribution to V^E [17].

$$V^E = V - \sum_{i=1}^{n} x_i V_i \quad (8)$$

The V^E gets a positive contribution where the mixtures have interactions owing to the dispersion forces or weak dipole-dipole interaction.

The Eyring's viscosity model enables to analyze viscosity data from a thermodynamic point of view to extract further information about liquid mixtures. Meyer *et al.* [18] reported the possibilities of using ΔG^{E*} to examine molecular interactions as the viscosity deviation. A positive deviation ΔG^{E*} signifies strong specific interactions between unlike molecules and classifiable as dispersion forces show negative deviation as suggested by authors in the references [18, 19]. All these parameters of ΔG^{E*}, η^E and V^E help to understand the nature of molecular interactions, size and shape of the molecules. Further, they are useful to correlate measured density and viscosity data of liquid mixtures.

3. Results and Discussion

The proposed correlation able to predict excess free energy of activation for viscous flow of the amine mixture with below 2% average absolute relative deviation (AARD %) for all mixtures. Table 1-3 give the calculated parameters of the correlation given in Eq 4-6 for the mixtures. Viscosity predictions were compared with measured data. Figure 1 shows the comparison between measured and correlated data for MDEA + MEA + H_2O mixtures. The correlation was able to fit the data within AARD 1.9% and maximum deviation (AMD) of 0.1 mPa·s.

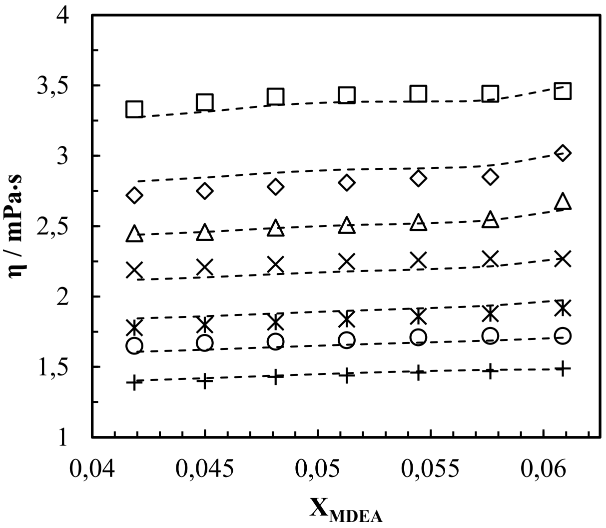

Figure 1: Viscosity variation of MDEA + MEA + H_2O mixtures with MDEA mole fraction and temperature: Experimental data from ref [5]; 293.15 K, '□'; 298.15 K, '◇'; 303.15 K, '△'; 308.15 K, 'x'; 313.15 K, 'ж'; 318.15 K, '○'; 323.15 K, '+'. The dotted lines represent the correlation in this work.

A comparison between measured and correlated viscosities was performed for the MDEA + DEA + H_2O mixtures as shown in Figure 2. It was found that the AARD and AMD for this mixture were 1.4% and 0.14 mPa·s respectively. Figure 3 illustrates the measured data with correlation for AMP + DEA + H_2O mixtures in which AARD and AMD were determined as 2.1% and 0.15 mPa·s respectively. The parameters of the correlation for the mixtures are given in Table 1 and the accuracies of the data fit are acceptable for engineering calculations.

A qualitative analysis was performed to investigate the intermolecular attractions in the mixtures. The summary of the excess properties of the amine mixtures is given in Table 4. The η^E was calculated as shown in the Eq (7). The calculated η^E is negative for all mixtures at considered concentrations and temperatures. It emphasizes weak intermolecular interactions between the unlike molecules compared to pure liquids. The pure MDEA, DEA and AMP are able to form strong H-bonds due to the presence of O-H in the molecule [20]. MDEA and DEA have two O-H groups while AMP has only one to contribute for H-bonds. During the mixing of amines with water, the breaking of H-bonds may result in a negative sign in η^E. The temperature has an effect of η^E in such a way that η^Ebecomes less negative with the increase of temperature. The strength of molecular interactions may get weaker due to the increase of thermal energy of the molecules.

Table 1: Binary parameters A_o, A_1 and A_2 of the equation $\Delta G_{jk}^{E*} = x_j x_k \sum_{i=0}^{n} A_i \left(x_j - x_k\right)^i$ for the excess free energy of activation for viscous flow for MDEA + MEA + H_2O

Parameter		Binary pair		
		MDEA + MEA	MEA + H_2O	MDEA + H_2O
A_0	a	5.3631 x 10^{4}	1.7589 x 10^{4}	-7.2961 x 10^{3}
	b	-1.6978 x 10^{2}	1.0270 x 10^{1}	9.8700
	c	2.1304 x10^{-1}	1.5794 x 10^{-1}	3.4820 x10^{-1}
A_1	a	1.1958 x 10^{5}	2.5656 x 10^{4}	-3.979 x 10^{3}
	b	8.4487 x 10^{2}	-1.4979 x 10^{1}	1.9979 x 10^{1}
	c	-4.6806	5.9708 x 10^{-1}	6.3296 x 10^{-1}
A_2	a	1.3708 x 10^{6}	3.4239 x 10^{3}	4.6638 x 10^{3}
	b	-1.8188 x 10^{4}	-2.0450 x 10^{1}	1.1958 x 10^{1}
	c	4.2780 x 10^{1}	4.6224 x 10^{-1}	2.6581 x 10^{-1}

Table 2: Binary parameters A_o, A_1 and A_2 of the equation $\Delta G_{jk}^{E*} = x_j x_k \sum_{i=0}^{n} A_i \left(x_j - x_k\right)^i$ for the excess free energy of activation for viscous flow for MDEA + DEA + H_2O

Parameter		Binary pair		
		MDEA + DEA	DEA + H_2O	MDEA + H_2O
A_0	a	1.0027 x 10^{8}	4.1277 x 10^{7}	-5.0908 x 10^{6}
	b	3.1414 x 10^{5}	1.1918 x 10^{5}	-1.7794 x10^{4}
	c	9.5620 x10^{2}	3.7002 x 10^{2}	-6.3678 x 10^{1}
A_1	a	4.3279 x 10^{7}	4.5805 x 10^{6}	-2.5435 x 10^{5}
	b	1.3742 x 10^{5}	-1.1254 x 10^{4}	5.5912 x 10^{2}
	c	4.7546 x 10^{2}	-3.3341 x 10^{1}	-3.5906 x 10^{-3}
A_2	a	1.4787 x 10^{7}	-5.0089 x 10^{7}	6.3095 x 10^{6}
	b	-4.8893 x10^{4}	-1.7303 x10^{5}	2.3706 x10^{4}
	c	4.8911 x 10^{1}	-5.3515 x 10^{2}	8.2541 x 10^{1}

Table 3: Binary parameters A_o, A_1 and A_2 of the equation $\Delta G_{jk}^{E*} = x_j x_k \sum_{i=0}^{n} A_i \left(x_j - x_k\right)^i$ for the excess free energy of activation for viscous flow for AMP + DEA + H_2O

Parameter		Binary pair		
		AMP + DEA	DEA + H_2O	AMP + H_2O
A_0	a	1.1157 x 10^{4}	2.7551 x 10^{3}	1.5131 x 10^{3}
	b	8.4643 x 10^{1}	2.0589 x 10^{1}	2.3656
	c	-9.8481 x 10^{-1}	5.2882 x 10^{-1}	-1.0803 x 10^{-1}
A_1	a	-3.0081 x 10^{5}	7.6788 x 10^{3}	4.2936 x 10^{2}
	b	1.0653 x 10^{3}	-1.7424 x 10^{1}	5.1912
	c	2.0884 x 10^{-1}	1.1355	3.6564 x 10^{-1}
A_2	a	4.4827 x 10^{6}	4.7115 x 10^{3}	-1.5476 x 10^{3}
	b	-2.5146 x 10^{4}	-4.9228 x 10^{1}	2.6677
	c	3.3641 x 10^{1}	6.1636 x 10^{-1}	5.8774 x 10^{-1}

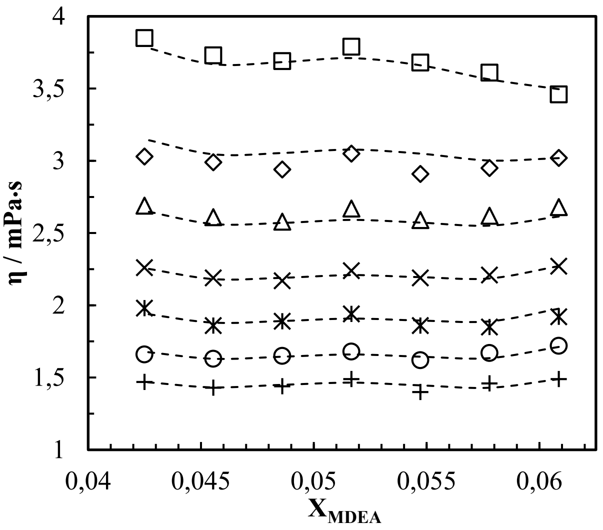

Figure 2. Viscosity variation of MDEA + DEA + H_2O mixtures with MDEA mole fraction and temperature: Experimental data from ref [5]; 293.15 K, '□'; 298.15 K, '◇'; 303.15 K, '△'; 308.15 K, 'x'; 313.15 K, 'ж'; 318.15 K, 'o'; 323.15 K, '+'. The dotted lines represent the correlation in this work.

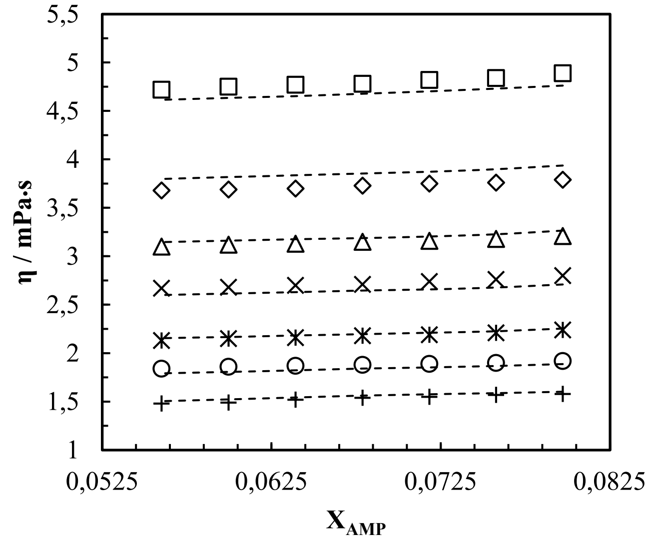

Figure 3. Viscosity variation of AMP + DEA + H_2O mixtures with AMP mole fraction and temperature: Experimental data from ref [5]; 293.15 K, '□'; 298.15 K, '◇'; 303.15 K, '△'; 308.15 K, 'x'; 313.15 K, 'ж'; 318.15 K, 'o'; 323.15 K, '+'. The dotted lines represent the correlation in this work.

As shown by Kauzmann and Eyring [21], the viscosity of a mixture highly depends on the entropy of the mixture that is related to the molecular structure, interaction between molecules and enthalpy of the mixture [22, 23]. The negative deviation of V^E for all mixtures in their different compositions and temperatures indicates strong intermolecular interactions among the unlike molecules. Further, V^E gets a negative contribution by arranging molecules within vacant spaces in each other's structure due to the different size and shape of the molecules. This also affects the viscosity of a mixture as intermolecular interactions.

As shown in Figure 4, the calculated ΔG^{E*} from Eyring's viscosity representation is positive for the considered mixture compositions and temperatures indicating that the mixtures have strong intermolecular interactions. This is supported by excess volume property giving a negative sign for V^E. The formation of new H-bond between unlike molecules can result in a positive deviation in ΔG^{E*}. MEA has the potential to form H-bonds with other amines and H_2O due to the presence of hydroxyl and amino functional groups. For (MDEA + MEA + H_2O) mixtures at 293.15 K, highest excess activation energy is shown at the mixture composition of 30 mass% MDEA + 0 mass% MEA + 70 mass% H_2O mixture and it gradually decreased with the decrease of MDEA and increase of MEA concentrations under constant weight percent of H_2O. The decrease of ΔG^{E*} with the increase of MEA mole fraction shows that intermolecular attractions have been weakened by MEA.

Table 4: Excess properties of ΔG^{E*}, η^E and V^E of the amine mixtures.

Mixture	ΔG^{E*}	η^E	V^E
MDEA + MEA + H_2O	> 0	< 0	< 0
MDEA + DEA + H_2O	> 0	< 0	< 0
AMP + DEA + H_2O	> 0	< 0	< 0

For the AMP + DEA + H_2O mixtures at 293.15 K, highest excess free activation energy is observed at the mixture composition of 30 mass% AMP + 0 mass% DEA + 70 mass% H_2O mixture. The increase of DEA mole fraction decreases the ΔG^{E*} indicating weak intermolecular interactions compared to the mixture of 30 mass% AMP + 0 mass% DEA + 70 mass% H_2O.

The excess free energy of activation for viscous flow of MDEA + DEA + H_2O mixtures shows a peak at the mixture composition of 25.5 mass% MDEA + 4.5 mass% DEA + 70 mass% H_2O at 293.15 K. This indicates that the intermolecular interactions are stronger at that particular composition than the other amine composition at 293.15 K.

The free energy of activation for viscous flow was determined by using Eq (1). Figure 5 illustrates the variation of $\Delta G^*$with amine mole fraction of the mixtures at 293.15 K. The AMP + DEA + H_2O mixtures show the highest free energy among the considered mixtures while MDEA + MEA + H_2O has the lowest free energy.

The increase in temperature decreases both ΔG^{E*} and ΔG^*. Figure 6 shows the influence of temperature on ΔG^* for the mixtures with 24 mass% AMINE (1) + 6 mass% AMINE (2) + 70 mass% H_2O. The increase in molecular energy has weakened the strength of H-bonds and has enhanced the movements of the molecules. The decrease in ΔG^{E*} indicates that solution characteristics change toward the ideal conditions with the increase of temperature.

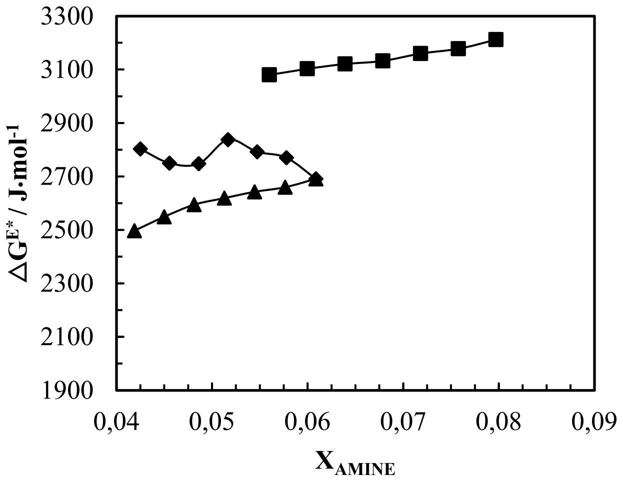

Figure 4. Variation of ΔG^{E*} with amine mole fractions at temperature of 293.15 K. X_{AMP} in AMP + DEA + H_2O '■'; X_{MDEA} in MDEA + DEA + H_2O '◆'; X_{MDEA} in MDEA + MEA + H_2O '▲'.

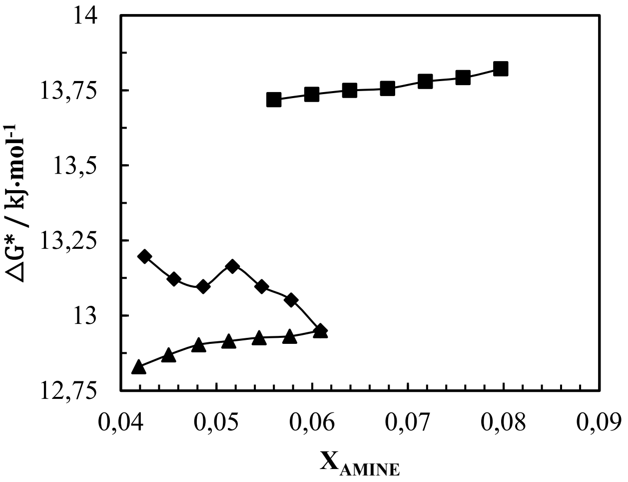

Figure 5. Variation of ΔG^{*} with amine mole fractions at temperature of 293.15 K. X_{AMP} in AMP + DEA + H_2O '■'; X_{MDEA} in MDEA + DEA + H_2O '◆'; X_{MDEA} in MDEA + MEA + H_2O '▲'.

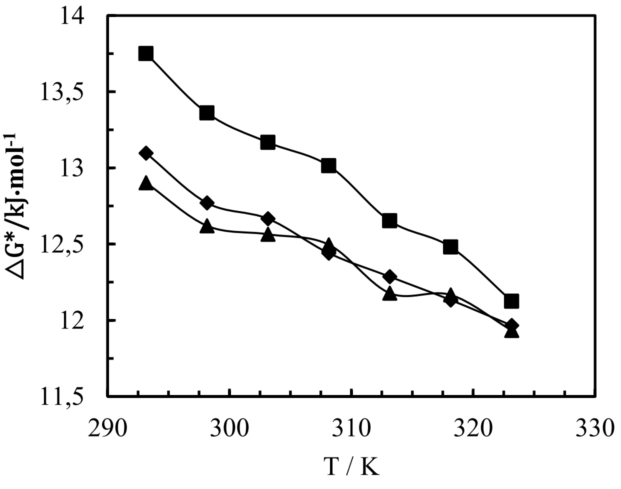

Figure 6. Variation of ΔG^{*} with temperature. AMP + DEA + H_2O '■'; MDEA + DEA + H_2O '◆'; MDEA + MEA + H_2O '▲'.

This study analyzed the density and viscosity of amine and water ternary mixtures to understand the molecular interactions. The correlation represents the viscosity data of ternary amines and water mixtures with acceptable accuracy to use them in engineering calculations. Further, the correlations can be improved to fit viscosities of CO_2 loaded amine and water mixtures as they are important in post-combustion amine based CO_2 capture.

4. Conclusions

Three aqueous amine mixtures of MDEA + MEA + H_2O, MDEA + DEA + H_2O and AMP + DEA + H_2O were analyzed based on viscosity and density data in the literature for their free energy of activation for viscous flow. The excess free energy was evaluated and correlated by a Redlich-Kister polynomial to fit the viscosity data. The proposed correlations were able to correlate the ΔG^{E*} within 2% AARD.

The same correlation was able to represent viscosities of MDEA + MEA + H_2O mixtures within AARD 1.9% and AMD of 0.1 mPa·s. For the MDEA + DEA + H_2O mixtures the viscosity data were fitted within AARD 1.4% and AMD 0.14 mPa·s. And for the AMP + DEA + H_2O mixtures that are of AARD 2.1% and 0.15 mPa·s. These accuracies are acceptable in engineering calculations.

The excess properties of molar volume, viscosity and free energy of activation for viscous flow show the presence of strong intermolecular interactions in all mixtures. The negative and positive signs for excess volume and excess free energy of activation for viscous flow indicate the presence of H-bonds between unlike molecules while negative signs of excess viscosity predict weak intermolecular interactions compared to the ideal mixture condition. This may occur due to the breaking of H-bond during the mixing. The mixtures exhibit $\Delta G^{E*}_{AMP+DEA+H_2O}$ > $\Delta G^{E*}_{MDEA+DEA+H_2O}$ > $\Delta G^{E*}_{MDEA+MEA+H_2O}$ for the considered amine concentrations. Accordingly, AMP + DEA + H_2O mixtures possess the highest intermolecular interactions among the unlike molecules and MDEA + MEA + H_2O mixtures indicate the lowest.

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