Densities, Viscosities and Excess Properties for Dimethyl Sulfoxide with Diethylene Glycol and Methyldiethanolamine at Different Temperatures

Maria Magdalena Budeanu * and Vasile Dumitrescu

Chemistry Department, Petroleum and Gas University of Ploiesti, 100680 Ploiesti, Romania; vdui@upg-ploiesti.ro
* Correspondence: maria.budeanu@upg-ploiesti.ro

Abstract: Densities and viscosities of the binary systems dimethylsulfoxide with diethylene glycol and methyldiethanolamine were measured at temperatures ranging from 293.15 to 313.15 K, at atmospheric pressure and over the entire composition range. The experimental density data was correlated as a function of composition using Belda’s and Herraez’s equations, and as a function of temperature and composition using the models of Emmerling et al. and Gonzalez-Olmos-Iglesias. The viscosity results were fitted to the Grunberg-Nissan, Heric-Brewer, Wilson, Noda, and Ishida and Eyring-NRTL equations. The values of viscosity deviation ($\Delta \eta$), excess molar volume ($V_E$), partial molar volumes ($V_1$ and $V_2$) and apparent molar volume ($V_{\phi,1}$ and $V_{\phi,2}$) were determined. The excess functions of the binary systems were fitted to the polynomial equations. The values of thermodynamic functions of activation of viscous flow were calculated and discussed.

Keywords: density; viscosity; excess molar volume; viscosity deviation; thermodynamic functions of activation

1. Introduction

The removal of acidic gases or liquids such as carbon dioxide (CO$_2$), sulfur dioxide (SO$_2$), hydrogen sulfide (H$_2$S), carbonyl sulfide (COS) and carbon disulfide (CS$_2$) from natural settings, refineries, synthesis gas streams and petrochemicals are of increasing importance as environmental protection becomes more and more serious [1]. It is a significant operation in gas processing to eliminate acid compounds by means of various processes, among which is gas absorption by chemical solutions such as alkanolamines (monoethanolamine, diethanolamine, diisopropanolamine, or methyldiethanolamine) [2]. The importance of basic physicochemical properties for the density and viscosity data is an indispensable requirement over a broad range of temperatures for the absorption and desorption processes of SO$_2$ [3].

Dimethyl sulfoxide (DMSO) was used intensively in SO$_2$ absorption because of its low volatility and good affinity with SO$_2$ [4,5]. The physicochemical properties of solutions of glycols are useful, since such solutions are used in several processes in the pharmaceutical, petroleum, cosmetic, oil and food industries [6]. Binary solution of DMSO with glycols may attract attention due to the possible intermolecular interplay of S=O group in DMSO with –OH group in glycol [7].

In this work, the densities and viscosities of binary systems of dimethyl sulfoxide (DMSO) + diethylene glycol (DEG) or methyldiethanolamine (MDEA) were measured at temperatures between 293.15 and 313.15 K, over the entire composition range and at atmospheric pressure. Investigations into the literature have shown that these systems have been examined but not in the same conditions. Tsierkezos et al. [6] reported the values of densities for diethylene glycol with dimethylsulfoxide at 298.15 K and Naidu et al. [8] investigated the densities and viscosities of diethylene glycol with dimethyl sulfoxide at 308.15 K. Wang et al. [1] studied densities of binary mixtures of dimethyl
sulfoxide with methyldiethanolamine at atmospheric pressure with temperatures ranging from 293.15 to 363.15 K. Wang et al. [9] studied the densities and viscosities of diethylene glycol + dimethyl sulfoxide solutions in the temperature range 298.15–313.15 K.

The present work was mainly focused on investigating density and viscosity data of binary solutions of DEG + DMSO and MDEA + DMSO at T = 293.15, 298.15, 303.15, 308.15, and 313.15 K for the whole composition range. From our experimental data, excess molar volumes and viscosity deviations were calculated and correlated with the polynomial equations. The thermodynamic functions of activation of viscous flow have been estimated from the experimental values. Five equations were tested to correlate viscosity of the binary mixtures.

2. Materials and Methods

2.1. Materials

The chemical DMSO (mass ≥99.5%, CAS 67-68-5, water content ≤0.05%) was obtained from Merck, DEG (mass ≥99%, CAS 111-46-6, water content ≤1%) was supplied by Chemical Company and MDEA (mass ≥99%, CAS 105-59-9, water content ≤1%) was purchased from Chemical Company. In order to reduce the influence of water on the experiment, the chemicals DEG and MDEA were dried over molecular sieves (Fluka type 4 Å), and their effective component content was determined by means of gas chromatography. All specification of chemical samples is listed in Table 1. The measurements were made at atmospheric pressure, \( p = 0.1 \) MPa, which was measured in our laboratory by a mercury barometer with an uncertainty of ±0.002 MPa.

| Chemical Name | Chemical Formula | Source | Mass Fraction Purity | Isolation Method |
|---------------|-----------------|--------|----------------------|------------------|
| DMSO          | \( \text{C}_2\text{H}_6\text{OS} \) | Merck  | 99.5%                | None             |
| DEG           | \( \text{C}_4\text{H}_{10}\text{O}_3 \) | Chemical Company | ≥99.3% | Desiccation \(^a\) and Degasification \(^b\) |
| MDEA          | \( \text{C}_5\text{H}_{13}\text{NO}_2 \) | Chemical Company | ≥99.3% | Desiccation \(^a\) and Degasification \(^b\) |

\(^a\) Molecular sieve type 4Å. \(^b\) Ultrasound.

2.2. Measurements and Method Analysis

The binary solutions were prepared by weighing using an analytical balance (Adventurer Pro AV 264CM model) at atmospheric pressure and ambient temperature with a precision of ±10\(^{-4}\) g. The uncertainty for the mixtures’ mole fraction was less than 0.0006.

Densities of pure liquids and their mixtures were determined with an Anton Paar digital vibrating U-tube densimeter (model DMA 500). The temperature was determined with an integrated Pt100 platinum thermometer together with a Peltier element. The stated repeatability for density and temperature measurements by the manufacturer was 0.0002 g·cm\(^{-3}\) and 0.1 K respectively. The densimeter was calibrated with bidistilled and degassed water before and after each of the density measurements. The combined expanded uncertainty of the densities is estimated within 0.0015 g·cm\(^{-3}\) with a 0.95 level of confidence for the present work. Expanded uncertainty of the excess volume is estimated to be 0.04 cm\(^3\)·mol\(^{-1}\) (0.95 confidence level).

Viscosities of the pure compounds and of the binary solutions were determined with an Ubbelohde kinematic, viscosity measuring unit ViscoClock (Schott-Gerate GmbH) that was kept in a vertical position in a thermostatic bath (U-10 Freital). The temperature was controlled with a precision of ±0.05 K.

The kinematic viscosity was calculated using the equation:

\[
\nu = At - \left( \frac{B}{t} \right)
\]

where \( \nu \) is the kinematic viscosity and \( t \) is the flow time, \( A \) and \( B \) are characteristic constants of the viscometer. The constants \( A \) and \( B \) were determined by taking doubly distilled water
and benzene (Merck, mole fraction purity ≥ 0.995) as the calibrating liquids. The accuracy of time measurement is ±0.01 s. The dynamic viscosity was determined using the equation:

\[ \eta = \nu \rho \]  

(2)

where \( \rho \) is the density of the liquid.

Each value of the viscosity was the average of five measurements (the measurements refer to the uncertainty values within ±0.02 s). The combined relative expanded uncertainty of the dynamic viscosity was estimated to be 5%. Expanded uncertainties in the deviation viscosity was estimated to be 0.06 mPa·s (0.95 of confidence).

3. Results

3.1. Density and Viscosity

The experimental densities and viscosities for pure DMSO, DEG and MDEA in the temperature range from 293.15 to 313.15 K are found to be in good agreement with reported values in the literature and comparison of these values is reported in Table 2.

DMSO density values reported in the literature [1,6,9–16] differ from our experimental data with a maximum of 0.2% and viscosity values reported in the literature [3,12,17–21] differ with a maximum of 2.5%. For DEG, density values found in the literature [1,9,22–28] differ with a maximum of 0.2% and for MDEA [1,29–34] they differ by less than 0.07%. Viscosity values reported in the literature differ from our results by a maximum of 2.3% for DEG [26,28] and a maximum of 2.1% for MDEA [29–31,35–37]. Viscosity values of DMSO and DEG reported by Wang et al. [9] differ by more than 30% compared with our results. These differences can be attributed to the different purity of the reagents used.

Table 2. Experimental and literature values of density (\( \rho \)) and viscosity (\( \eta \)) of the pure components in the temperature range from 293.15 to 313.15 K.

| Component | T/(K)   | \( \rho/(g\,cm^{-3}) \) | \( \eta/(mPa\cdot s) \) |
|-----------|---------|-----------------|------------------|
|           | This Work | Lit. Value | This Work | Lit. Value |
| DMSO      | 293.15  | 1.1002 [10] | 2.245 [17] |
|           |         | 1.10053 [11]|             |
|           |         | 1.10865 [1] |             |
|           |         | 1.0955 [12] |             |
|           |         | 1.0954 [13] |             |
|           |         | 1.09530 [6] |             |
|           |         | 1.0946 [9]  | 1.40 [9]     |
|           | 298.15  | 1.0904 [12] | 1.0900 [14]  |
|           |         | 1.0888 [9]  | 1.090812 [1] |
|           |         | 1.0854 [12] | 2.021         |
|           |         | 1.08573 [15]|             |
|           |         | 1.0831 [9]  | 1.0807 [12]  |
|           |         | 1.08024 [16]|             |
|           |         | 1.0785 [9]  | 1.843         |
|           |         | 1.080770 [1]|             |
|           | 303.15  | 1.0902      | 1.867         |
|           |         | 1.0853      | 1.867         |
|           | 308.15  | 1.0803      | 1.525         |
|           | 313.15  | 1.0803      | 1.525         |
Table 2. Cont.

| Component | T/(K) | \(\rho/(g \cdot cm^{-3})\) | \(\eta/(mPa \cdot s)\) |
|-----------|-------|-----------------|-----------------|
|           | This Work | Lit. Value | This Work | Lit. Value |
| DEG       | 293.15 | 1.1105 [22] | 33.270 | - |
|           | 298.15 | 1.11583 [23] | 26.865 | 27.15 [26] |
|           | 303.15 | 1.11351 [25] | 21.280 | 21.754 [26] |
|           | 308.15 | 1.1128 [9] | 12.4 [9] |
|           | 313.15 | 1.11260 [1] | 17.00 [9] |
|           |        | 1.1098 [26] | 12.7 [26] |
|           |        | 1.11098 [27] | 16.9 [26] |
|           |        | 1.1093 [9] | 10.4 [9] |
| MDEA      | 293.15 | 1.0406 [29] | 1.0409 | 102.7 [31] |
|           | 298.15 | 1.03966 [30] | 1.03556 [31] | 74.81 [35] |
|           | 303.15 | 1.037863 [32] | 1.0338 [29] | 57.57 [36] |
|           | 308.15 | 1.03213 [30] | 1.02834 [31] | 44.21 [37] |
|           | 313.15 | 1.03017 [1] | 1.025401 [1] | 34.89 [29] |
|           |        | 1.0331 [29] | 45.129 | 44.62 [29] |
|           |        | 1.0303 [30] | 34.78 [36] |
|           |        | 1.0255 [29] | 34.89 [29] |
|           |        | 1.0250 [34] | 35.00 [37] |
|           |        | 1.0250 [34] | 34.833 | 35.00 [37] |
|           |        | 1.0250 [34] | 34.833 | 35.00 [37] |

Standard uncertainties: \(u(\rho) = 0.002 \text{ MPa}, \) Expanded uncertainties: \(U(\eta) = 5\%\) and \(U(\rho) = 0.0015 \text{ g} \cdot \text{cm}^{-3}(0.95\% \text{ of confidence})\).

The experimental densities and viscosities for the binary systems DEG (1) + DMSO (2) and MDEA (1) + DMSO (2) are listed in Tables 3 and 4.

The density of binary system DEG (1) + DMSO (2) increases with the increase in DEG concentration, and for the system MDEA (1) + DMSO (2), density increases with the increase in DMSO concentration. Viscosity of binary system DEG (1) + DMSO (2) increases with the increase in DEG concentration, while for the system MDEA (1) + DMSO (2), it increases with the increase in MDEA concentration.

Table 3. Density values \(\rho/(g \cdot cm^{-3})\) as a functions of mole fraction in the temperature range from 293.15 to 313.15 K and at atmospheric pressure.

| \(T/(K)\) | \(x_1\) | DEG (1) + DMSO (2) | MDEA (1) + DMSO (2) |
|-----------|-------|------------------|------------------|
| 293.15    | 0.1062 | 1.09988 | 1.09393 | 1.08899 | 1.08400 |
| 298.15    | 0.2007 | 1.10199 | 1.10700 | 1.09721 | 1.08732 |
| 303.15    | 0.3034 | 1.10489 | 1.10900 | 1.09512 | 1.09041 |
| 308.15    | 0.4110 | 1.10759 | 1.11026 | 1.09778 | 1.09321 |
| 313.15    | 0.5108 | 1.11059 | 1.11058 | 1.10121 | 1.09682 |
|           | 0.5972 | 1.11449 | 1.11071 | 1.10251 | 1.09821 |
|           | 0.6955 | 1.11149 | 1.11086 | 1.10411 | 1.09981 |
|           | 0.8096 | 1.11149 | 1.11094 | 1.10491 | 1.10071 |
|           | 0.8819 | 1.11149 | 1.11094 | 1.10491 | 1.10071 |
Table 3. Cont.

| $T$/(K) | $x_1$ | MDEA (1) + DMSO (2) | DEG (1) + DMSO (2) | MDEA (1) + DMSO (2) |
|---------|-------|---------------------|---------------------|---------------------|
|         | 293.15 | 298.15 | 303.15 | 308.15 | 313.15 |
| 0.1003  | 1.0899 | 1.0850 | 1.0803 | 1.0758 | 1.0712 |
| 0.1967  | 1.0808 | 1.0762 | 1.0718 | 1.0675 | 1.0631 |
| 0.2997  | 1.0725 | 1.0678 | 1.0635 | 1.0592 | 1.0550 |
| 0.4006  | 1.0654 | 1.0607 | 1.0565 | 1.0525 | 1.0483 |
| 0.5001  | 1.0595 | 1.0549 | 1.0509 | 1.0469 | 1.0428 |
| 0.6020  | 1.0544 | 1.0498 | 1.0459 | 1.0419 | 1.0379 |
| 0.6937  | 1.0504 | 1.0458 | 1.0419 | 1.0380 | 1.0341 |
| 0.7983  | 1.0465 | 1.0419 | 1.0381 | 1.0342 | 1.0303 |
| 0.8967  | 1.0436 | 1.0390 | 1.0351 | 1.0315 | 1.0277 |

Standard uncertainties: $u(x_1) = 6 \times 10^{-4}$, $u(p) = 0.002$ MPa, $u(T) = 0.1$ K; Expanded uncertainties: $U(p) = 0.0015$ g cm$^{-3}$ (0.95 of confidence).

Table 4. Viscosity values $\eta/$(mPa-s) as a functions of mole fraction in the temperature range from 293.15 to 313.15 K and at atmospheric pressure.

| $T$/(K) | $x_1$ | DEG (1) + DMSO (2) | MDEA (1) + DMSO (2) |
|---------|-------|---------------------|---------------------|
|         | 293.15 | 298.15 | 303.15 | 308.15 | 313.15 |
| 0.1062  | 3.110  | 2.877  | 2.520  | 2.281  | 2.049  |
| 0.2007  | 4.306  | 3.799  | 3.404  | 3.012  | 2.677  |
| 0.3034  | 5.792  | 5.115  | 4.334  | 3.883  | 3.383  |
| 0.4110  | 7.884  | 6.762  | 5.702  | 5.082  | 4.361  |
| 0.5108  | 10.198 | 8.630  | 7.072  | 6.337  | 5.394  |
| 0.5972  | 12.843 | 10.717 | 8.605  | 7.647  | 6.428  |
| 0.6955  | 16.372 | 13.356 | 10.796 | 9.502  | 7.951  |
| 0.8096  | 21.286 | 17.154 | 13.923 | 11.854 | 9.912  |
| 0.8819  | 24.874 | 20.097 | 16.220 | 13.703 | 11.431 |

Standard uncertainties: $u(x_1) = 6 \times 10^{-4}$, $u(p) = 0.002$ MPa, $u(T) = 0.05$ K; Expanded uncertainties: $U(\eta) = 5\%$.

The densities of binary solutions were represented as a function on composition by the following Belda [38] (Equation (3)) and Herraez [39] (Equation (4)) equations, and with composition and temperature using the Emmerling et al. [40] (Equation (5)) and Gonzalez-Olmos Iglesias [41] (Equation (6)) equations:

\[
\rho = \rho_2 + (\rho_1 - \rho_2)x_1 \left( \frac{1 + m_1(1 - x_1)}{1 + m_2(1 - x_1)} \right) 
\]

(3)

\[
\rho = \rho_2 + (\rho_1 - \rho_2)x_1^{A + Bx_1 + Cx_1^2} 
\]

(4)

\[
\rho = x_1\rho_1 + x_2\rho_2 + x_1x_2 \left[ P_1 + P_2T + P_3T^2 + (P_4 + P_5T + P_6T^2)(x_1 - x_2) + \right] (P_7 + P_8T + P_9T^2)(x_1 - x_2) 
\]

(5)

\[
\rho = 2 \sum_{i=0}^{2} A_i x_i^i 
\]

(6)
The temperature dependence of the densities ($\rho_i$) of each pure substance $i$ involved in Equation (5) is expressed using the equation:

$$\rho_i = A_i + B_iT + C_iT^2 \quad (i = 1, 2)$$

$$A_i = \sum_{j=1}^{2} A_{ij}T^i$$

The adjustable parameters of these equations ($m_1$, $m_2$, $A$, $B$, $C$, $P_1$, $P_2$, $A_i$, $B_i$, $C_i$ and $A_{ij}$) were estimated using the experimental data and a nonlinear regression analysis employing the Levenberg-Marquardt algorithm [42]. Tables A1 and A2 in the Appendix A show the fitting parameters along with the standard deviation calculated with the equation:

$$\sigma = \left[ \frac{\sum(X_{\text{exp}}-X_{\text{calc}})^2}{m-n} \right]^{1/2}$$

where $X$ is the value of the analyzed property, $m$ is the number of data points, and $n$ is the number of estimated parameters. Data presented in Tables A1 and A2 show that Herraez's equation offers the best results for correlating the density with composition, while Emmerling et al.'s equations the best for correlating the density of the binary solutions with composition and temperature.

In this paper, the one-parameter Grunberg–Nissan [43] and two-parameter Eyring–Brewer [44], Wilson [45], Noda and Ishida [46] and Eyring-NRTL [47] models were used to represent the dependence of viscosity on the concentration of components in binary systems. Grunberg and Nissan [43] proposed an equation based on a parameter:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d$$

The Eyring–Brewer [44] equation with two parameters is:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 [\alpha_{12} + \alpha_{21} (x_1 - x_2)]$$

By the application of the Wilson [45] equation, viscosity of the binary mixtures can be expressed as:

$$\ln(\eta V) = x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + x_1 \ln \left(x_1 + x_2 \frac{V_2}{V_1} \exp \left(-\frac{\lambda_{12}}{RT} \right) \right) + x_2 \ln \left(x_2 + x_1 \frac{V_1}{V_2} \exp \left(-\frac{\lambda_{21}}{RT} \right) \right)$$

Noda and Ishida [46] proposed the following semi-empirical equation:

$$\ln(\eta V) = x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + x_1 x_2 \left[ \frac{w_{12}}{x_2 + x_1 \exp \left(-\frac{w_{12}}{RT} \right)} + \frac{w_{21}}{x_1 + x_2 \exp \left(-\frac{w_{21}}{RT} \right)} \right]$$

The Eyring-NRTL [47] correlative model is given by the relation:

$$\ln(\eta V) = x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + x_1 x_2 \left[ \frac{\tau_{12} \exp(-\alpha \tau_{21})}{x_1 + x_2 \exp(-\alpha \tau_{21})} + \frac{\tau_{12} \exp(-\alpha \tau_{12})}{x_2 + x_3 \exp(-\alpha \tau_{12})} \right]$$

In these equations $\eta$, $\eta_1$, $\eta_2$ are the dynamic viscosities of the liquid mixtures and of the pure components 1 and 2, $x_1$, $x_2$ are the mole fractions, $M_1$, $M_2$ are the molecular masses, $V$ is the molar volume of the mixtures, $V_1$ and $V_2$ are the respective molar volumes of the pure components, $T$ is the temperature, $R$ is the gas constant; $d$, $\alpha_{12}$, $\alpha_{21}$, $\lambda_{12}$, $\lambda_{21}$, $w_{12}$, $w_{21}$, $\tau_{12}$ and $\tau_{12}$ are interaction parameters (viscosity coefficients) and reflect the non-ideality of the systems. The Eyring-NRTL equation has three parameters, including $\alpha$, which is a measure of non-ideality of the systems, considered here to be fixed at 0.20 [48].

The parameters were estimated using the experimental viscosity data and a non-linear regression analysis employing the Levenberg-Marquardt algorithm [42].
The ability of these models to correlate viscosity data was tested by calculating the average absolute deviation (ADD%), between the experimental and calculated values, using the equation:

$$ADD\% = \frac{100}{m} \sum_{i=1}^{m} \left| \frac{\eta_{exp} - \eta_{cal}}{\eta_{exp}} \right|$$  \hspace{1cm} (15)

where $n$ is the number of experimental data points.

The presented data in Table A3 show that, for the DEG + DMSO system, ADD% values of maximum 2% are obtained for the Grunberg–Nissan and Noda–Ishida equations, and for the MDEA + DMSO system, the ADD% values for the two equations are very high (10%). For the Heric–Brewer equation, ADD% values of maximum 1.5% are obtained for the DEG + DMSO system and of maximum 6.0% in the case of the system formed by MDEA and DMSO. Approximately the same values are obtained for the Wilson equation. The Eyring–NRTL equation presents the best results, with ADD% values of maximum 0.8% for the DEG + DMSO system and maximum 3.3% for the MDEA + DMSO system. The higher ADD% in the MDEA + DMSO system than in the DEG + DMSO system can be attributed to higher deviation of the system from ideality. The obtained ADD% values lower than 5% are regarded to be acceptable for engineering calculations [49].

3.2. Excess Properties

3.2.1. Excess Molar Volume

The excess molar volumes have been calculated from the experimental densities data using the following equation:

$$V^E = \left[ \frac{x_1 M_1 + x_2 M_2}{\rho} \right] - \left[ \frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right]$$  \hspace{1cm} (16)

where $x_1$ and $x_2$ are the mole fractions of the components, $M_1$ and $M_2$ are the molar masses of components 1 and 2, $\rho$, $\rho_1$, and $\rho_2$ are the respective densities of the solution and of the pure components. The results of excess molar volumes are illustrated in Figures 1 and 2.

![Figure 1](image1.png)

**Figure 1.** Excess molar volumes ($V^E$) with mole fraction for DEG (1) + DMSO (2) system at various temperatures: ■ 293.15 K; ● 298.15 K; ▲ 303.15 K; ▼ 308.15 K; ♦ 313.15 K.
bonding systems [52,53]. In addition, DMSO can provide an S=O group and the hydroxylamines can provide OH or C-H groups for interactions [54].

The positive values for the MDEA + DMSO system indicate that there were no strong intermolecular interactions. The positive values are due to expansion of the solution volume due to mixing caused by the hydrogen bond rupture and dispersive interactions between unlike molecules [55]. These positive values of excess volume for the MDEA + DMSO system can be explained by the fact that DMSO forms a strong associative structure and by the self-association of MDEA molecules. The effect of temperature on the excess volumes shows a systematic decrease with rising temperature.

The experimental excess molar volumes are negative for the DEG + DMSO system and positive for the MDEA + DMSO binary system in the whole composition range at all temperatures.

The negative values are a consequence of the following effects: (1) strong intermolecular interactions due to the charge-transfer complex, dipole-dipole and dipole-induced dipole interactions, and H-bonding between unlike molecules finally leading to more efficient packing in the mixture than in the pure liquids; (2) structural effects which arise from suitable interstitial accommodation giving a more compact structure of solutions [50].

The negative excess volume values for the DEG + DMSO system indicated that the volume of the mixture was less than the sum of the volumes of the pure components, possibly due to contraction of the mixing volume caused by structural effects and strong intermolecular interactions between DEG and DMSO. Similar behavior was observed by Qiao et al. [12] for the binary system tri-ethylene glycol + dimethyl sulfoxide.

The value became less negative with increasing temperature and arrived at the minimum around molar fraction 0.40 for DEG at all temperatures. These values indicate that there is a maximum volume contraction on mixing DEG with DMSO at a rate of 2:3.

Dimethyl sulfoxide is a highly polar solvent, not forming H-bond networks and tending toward self-association [1]. The molecular dynamics simulations demonstrated that in liquid DMSO, the H-bonds C–H...O=S are formed [51]. Amines are moderately polar but not as polar as alcohols of comparable molecular weights, and the polar nature of N–H results in the formation of hydrogen bonds with other amine molecules, or other H-bonding systems [52,53]. In addition, DMSO can provide an S=O group and the hydroxylamines can provide OH or C-H groups for interactions [54].

The positive values for the MDEA + DMSO system indicate that there were no strong intermolecular interactions. The positive values are due to expansion of the solution volume due to mixing caused by the hydrogen bond rupture and dispersive interactions between unlike molecules [55]. These positive values of excess volume for the MDEA + DMSO system can be explained by the fact that DMSO forms a strong associative structure and by the self-association of MDEA molecules. The effect of temperature on the excess volumes shows a systematic decrease with rising temperature.
3.2.2. Viscosity Deviation

The viscosity deviation ($\Delta \eta$) values were calculated from the experimental data of viscosity using the equation:

$$\Delta \eta = \eta - (x_1 \eta_1 + x_2 \eta_2)$$

(17)

where $\eta$ is the dynamic viscosity of the mixture, $x_1$, $x_2$ and $\eta_1$, $\eta_2$ are the mole fractions and the dynamic viscosities of pure components 1 and 2, respectively.

The $\Delta \eta$ values are shown in Figures 3 and 4. The viscosity deviation values are negative at all investigated temperatures for both systems. The viscosity deviations may be generally explained by considering the following factors: (1) the difference in size and shape of the component molecules and the loss of dipolar association to a decrease in viscosity; (2) specific interactions between unlike molecules, such as H-bond formation, and charge transfer complexes may cause an increase in the viscosity of mixtures rather than in pure components. The former effect produces negative excess viscosity, and the latter effect produces positive excess viscosity [56]. For the DEG + DMSO system, the negative values of viscosity deviation indicate that the strength of specific interactions is not the only factor influencing the deviation in viscosity. The molecular size and shape of the components also play an important role [57]. For these systems the negative values of viscosity deviation indicate that the molecular size and shape of the components is a more important factor than the strength of specific interactions for determining the viscosity deviation.

The negative values of viscosity deviation for MDEA + DMSO corroborated with positive $V^E$ values demonstrate that there were no strong molecular interactions.

The values of viscosity deviation decrease with an increase in temperature. An increase in temperature decreases self-association as well as the association between unlike components because of the increase in thermal energy [58].

![Figure 3. Viscosity deviation ($\Delta \eta$) with mole fraction for DEG (1) + DMSO (2) system at various temperatures: ■ 293.15 K; ● 298.15 K; ▲ 303.15 K; ▼ 308.15 K; ♦ 313.15 K.](image-url)
3.2.3. Apparent Molar Volume

The apparent molar volumes and viscosity deviation of the binary systems can be represented by the Redlich–Kister [59] (Equation (18)) and Hwang [60] (Equation (19)) equations:

$$X^E = x_1 x_2 \sum_{k=0}^{3} a_k (2x_1 - 1)^k$$  \hspace{1cm} (18)

$$X^E = x_1 x_2 \left( A_0 + A_1 x_1^2 + A_2 x_2^2 \right)$$  \hspace{1cm} (19)

where $X^E$ represents either of the following properties: $V^E$, $\Delta \eta$; $x_1, x_2$ are the mole fractions of the components 1 and 2, respectively, and $a_k, A_0, A_1, A_2$ denote the polynomial coefficients.

In addition, the excess molar volumes were also correlated with the Myers and Scott [61] equation:

$$X^E = x_1 x_2 \frac{\sum_{k=0}^{p} B_k z_{12}^k}{1 + \sum_{l=1}^{m} C_l z_{12}^l}$$  \hspace{1cm} (20)

where $X^E$ is $V^E$ and $z_{12} = x_1 - x_2$. $B_k$ and $C_l$ are polynomial coefficients.

The values of polynomial coefficients are given in Table A4 along with the standard deviation, $\sigma$, calculated with Equation (9). From the presented data it can be seen that, for both systems, the excess molar volume is best correlated using the Myers and Scott equation. The Redlich–Kister equation shows better results than the Hwang equation for correlating the viscosity deviation for both systems.

3.2.3. Apparent Molar Volume

The apparent molar volumes $V_{\phi,1}$ and $V_{\phi,2}$ of the binary systems were calculated with the equations [62]:

$$V_{\phi,1} = \frac{x_2 M_2 \rho_2 - \rho_m}{x_1} + \frac{M_2}{\rho_m}$$  \hspace{1cm} (21)

$$V_{\phi,2} = \frac{x_1 M_1 \rho_1 - \rho_m}{x_2} + \frac{M_2}{\rho_m}$$  \hspace{1cm} (22)

The values obtained in the temperature range from 293.15 to 313.15 K are listed in Tables A5 and A6.
3.2.4. Partial Molar Volumes

Partial molar volumes were calculated using the following equations:

\[
V_1 = V^E + V_1^0 + (1 - x_1) \left( \frac{\partial V^E}{\partial x_1} \right)_{p,T} 
\]

\[
V_2 = V^E + V_2^0 - x_1 \left( \frac{\partial V^E}{\partial x_1} \right)_{p,T} 
\]

where \( V_1^0 \) and \( V_2^0 \) are the molar volumes of pure components. The derivative \( \left( \frac{\partial V^E}{\partial x_1} \right)_{p,T} \) in Equations (23) and (24) was obtained by differentiation of Equation (18), which leads to the following equations:

\[
V_1 = V_1^0 + x_2^3 \sum_{k=0}^{3} a_k (2x_1 - 1)^k - 2x_1 x_2^3 \sum_{k=1}^{3} a_k (2x_1 - 1)^{k-1} 
\]

\[
V_2 = V_2^0 + x_1^3 \sum_{k=0}^{3} a_k (2x_1 - 1)^k + 2x_1^2 x_2^3 \sum_{k=1}^{3} a_k (2x_1 - 1)^{k-1} 
\]

The calculated values of partial molar volumes are listed in Tables A7 and A8.

Our results show that for the DEG + DMSO system, the decrease in the DMSO concentration leads to the increase in the values of the apparent molar volumes and partial molar volumes of DEG, and the decrease in the values of the apparent molar volumes and partial molar volumes of DMSO. For the MDEA + DMSO system the decrease in the DMSO concentration leads to the decrease in the values of the apparent molar volumes and partial molar volumes of MDEA and the increase in the values of the apparent molar volumes and partial molar volumes of DMSO.

3.3. Thermodynamic Functions of Activation

The activation energy of viscous flow was calculated with the equations [63]:

\[
\eta = \frac{hN}{V} \exp \left( \frac{\Delta G^\neq}{RT} \right) 
\]

\[
\Delta G^\neq = \Delta H^\neq - T \Delta S^\neq 
\]

where \( \eta \) is the viscosity of a liquid solution, \( h \) is Planck’s constant, \( N \) is Avogadro’s number, \( V \) is the molar volume of the solution, \( R \) is the universal gas constant, \( T \) is temperature, \( \Delta G^\neq \), \( \Delta H^\neq \) and \( \Delta S^\neq \) are the molar Gibbs energy, enthalpy and entropy of activation of viscous flow. The plots of \( \ln(\eta V/hN) \) versus \( 1/T \) are linear in the temperature range 293.15 to 313.15 K and the values of enthalpy of activation of viscous flow (\( \Delta H^\neq \)) and entropy of activation viscous flow (\( \Delta S^\neq \)) were obtained from the corresponding slopes and intercept. The values of \( \Delta G^\neq \) were also calculated. The values of thermodynamic functions of activation of viscous flow are listed in Table 5. The values of \( \Delta G^\neq \) and \( \Delta H^\neq \) are positive for both binary systems and increase with the decrease in DMSO concentration in the solution at a constant temperature. The values of \( \Delta G^\neq \) at constant concentration decrease if the temperature increases, except for the pure DMSO.

The values of \( \Delta S^\neq \) are positive for all compounds and binary mixtures except DMSO. The positive \( \Delta H^\neq \) values decrease with increasing DMSO concentration, indicating that the viscous flow in DMSO is easier than in binary mixtures (DEG + DMSO, MDEA + DMSO) or in DEG. The \( \Delta S^\neq \) values decrease with increasing DMSO concentration for both analyzed systems, which reveals that the viscous flow is more ordered processing DMSO than in binary mixtures or in DEG.
Table 5. Values of $\Delta G^\neq$, $\Delta H^\neq$, $\Delta S^\neq$ for the binary mixtures.

| $x_1$ | $\Delta H^\neq$ (kJ/mol) | $\Delta S^\neq$ (J/mol K) | $\Delta G^\neq$ (kJ/mol) T/(K) |
|-------|---------------------------|---------------------------|--------------------------------|
|       |                           |                           | 293.15 | 298.15 | 303.15 | 308.15 | 313.15 |
| DEG (1) + DMSO (2) |                           |                           |       |        |        |        |        |
| 0.0000 | 14.02                     | −2.01                     | 14.61  | 14.62  | 14.63  | 14.64  | 14.65  |
| 0.1062 | 15.59                     | 0.26                      | 15.51  | 15.51  | 15.51  | 15.50  | 15.50  |
| 0.2007 | 17.40                     | 3.49                      | 16.34  | 16.33  | 16.31  | 16.29  | 16.27  |
| 0.3034 | 19.96                     | 9.58                      | 17.15  | 17.10  | 17.05  | 17.01  | 16.96  |
| 0.4110 | 21.80                     | 13.10                     | 17.96  | 17.80  | 17.83  | 17.77  | 17.70  |
| 0.5108 | 23.55                     | 16.72                     | 18.65  | 18.57  | 18.48  | 18.40  | 18.32  |
| 0.5972 | 25.69                     | 21.91                     | 19.27  | 19.16  | 19.05  | 18.94  | 18.83  |
| 0.6955 | 26.68                     | 23.06                     | 19.91  | 19.80  | 19.68  | 19.57  | 19.45  |
| 0.8096 | 28.41                     | 26.52                     | 20.64  | 20.50  | 20.37  | 20.24  | 20.11  |
| 0.8819 | 29.02                     | 27.12                     | 21.07  | 20.93  | 20.80  | 20.66  | 20.53  |
| 1.0000 | 32.34                     | 35.69                     | 21.88  | 21.71  | 21.53  | 21.35  | 21.17  |
| MDEA (1) + DMSO (2) |                           |                           |       |        |        |        |        |
| 0.0000 | 14.02                     | −2.01                     | 14.61  | 14.62  | 14.63  | 14.64  | 14.65  |
| 0.1003 | 21.72                     | 16.02                     | 17.02  | 16.94  | 16.86  | 16.78  | 16.70  |
| 0.1967 | 23.69                     | 19.72                     | 17.91  | 17.81  | 17.71  | 17.61  | 17.52  |
| 0.2997 | 25.93                     | 24.08                     | 18.87  | 18.75  | 18.63  | 18.51  | 18.39  |
| 0.4006 | 28.08                     | 28.27                     | 19.79  | 19.65  | 19.51  | 19.37  | 19.23  |
| 0.5001 | 29.52                     | 30.13                     | 20.69  | 20.54  | 20.39  | 20.24  | 20.09  |
| 0.6020 | 32.32                     | 36.46                     | 21.63  | 21.45  | 21.26  | 21.08  | 20.90  |
| 0.6937 | 34.31                     | 40.39                     | 22.47  | 22.26  | 22.06  | 21.86  | 21.66  |
| 0.7983 | 36.57                     | 45.18                     | 23.32  | 23.10  | 22.87  | 22.64  | 22.42  |
| 0.8967 | 38.16                     | 48.10                     | 24.06  | 23.82  | 23.58  | 23.34  | 23.10  |
| 1.0000 | 39.56                     | 49.62                     | 25.01  | 24.76  | 24.51  | 24.27  | 24.02  |

4. Conclusions

Density and viscosity of the binary systems DEG (1) + DMSO (2) and MDEA (1) + DMSO (2) were determined at temperatures between 293.15 to 313.15 K and atmospheric pressure. The calculated $V^E$ values are negative for the DEG (1) + DMSO (2) system and positive for the MDEA (1) + DMSO (2) system, while the calculated $\Delta \eta$ were negative for both systems. Models from Grunberg–Nissan, Heric–Brewer, Wilson, Noda–Ishida and Eyring–NRTL have been used to calculate viscosity coefficients and were compared with experimental data. The results showed that the Eyring–NRTL model is adequate to describe the viscosities of the binary mixtures. The activation energies of viscous flow were calculated. The values of $\Delta G^\neq$ and $\Delta H^\neq$ are positive for both binary systems and the values of $\Delta S^\neq$ are positive for all compounds and binary mixtures except DMSO.

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Appendix A

Table A1. Parameters for the Belda and Herraez equations and standard deviations at temperature range from (293.15 to 313.15) K.

| Model                      | Parameters and \( \sigma/(g \text{ cm}^{-3}) \) | T/(K)     |
|----------------------------|-----------------------------------------------|-----------|
|                            | \( m_1 \)                                      | 293.15    |
|                            | 0.0732                                        | 0.0781    |
|                            | 0.0914                                        | 0.1248    |
|                            | 0.0734                                        |           |
| Belda                     | \( m_2 \)                                      | 298.15    |
|                            | -0.5208                                       | -0.4861   |
|                            | -0.4518                                       | -0.4087   |
|                            | -0.4267                                       |           |
|                            | \( 10^4 \sigma \)                             | 1.94      |
|                            | 1.81                                           | 1.97      |
|                            | 2.12                                           |           |
|                            | \( A \)                                        | 0.8147    |
|                            | 0.8549                                         | 0.8652    |
|                            | 0.8883                                         | 0.8994    |
|                            | \( B \)                                        | -1.0314   |
|                            | -1.1068                                       | -1.0394   |
|                            | -1.0627                                       | -1.0737   |
|                            | \( C \)                                        | 0.7901    |
|                            | 0.8852                                         | 0.8077    |
|                            | 0.8238                                         | 0.8712    |
|                            | \( 10^5 \sigma \)                             | 7.84      |
|                            | 7.49                                           | 4.70      |
|                            | 4.95                                           | 2.80      |
| MDEA (1) + DMSO (2)        | \( m_1 \)                                      | 0.3401    |
|                            | 0.3454                                         | 0.3586    |
|                            | 0.3935                                         | 0.4319    |
| Belda                     | \( m_2 \)                                      | -0.2987   |
|                            | -0.2827                                       | -0.2690   |
|                            | -0.2366                                       | -0.2016   |
|                            | \( 10^4 \sigma \)                             | 1.13      |
|                            | 1.24                                           | 1.53      |
|                            | 1.77                                           | 2.09      |
|                            | \( A \)                                        | 0.8238    |
|                            | 0.8289                                         | 0.8346    |
|                            | 0.8448                                         | 0.8576    |
|                            | \( B \)                                        | -0.7358   |
|                            | -0.7382                                       | -0.7492   |
|                            | -0.7638                                       | -0.7930   |
|                            | \( C \)                                        | 0.3382    |
|                            | 0.3382                                         | 0.3481    |
|                            | 0.3557                                         | 0.3788    |
|                            | \( 10^4 \sigma \)                             | 1.17      |
|                            | 0.99                                           | 1.13      |
|                            | 1.27                                           | 1.20      |

Table A2. Values of parameters in the range 293.15–313.15 K for the Emmerling et al. and Gonzales-Olmos-Iglesias models and standard deviations.

| Model                      | Parameters                         | Units:       |
|----------------------------|------------------------------------|--------------|
|                            | \( A_0 \) = 1.0860                 | \( A_{01} = -0.0014 \) |
|                            | \( A_1 = 1.4180 \)                 | \( A_{02} = 6.9155 \times 10^{-7} \) |
|                            | \( P_1 = -0.0368 \)               | \( A_{11} = 0.0028 \) |
|                            | \( P_2 = -0.0364 \)               | \( A_{10} = -0.4239 \) |
|                            | \( P_3 = 0.4773 \)                | \( A_{20} = 0.0383 \) |
| Emmerling et al.           | \( B_1 = 9.7611 \times 10^{-4} \) | \( C_1 = -2.9500 \times 10^{-6} \) |
|                            | \( B_2 = -0.0002 \)               | \( C_2 = 2.9207 \times 10^{-7} \) |
|                            | \( P_4 = 3.6037 \times 10^{-4} \) | \( P_5 = -6.2313 \times 10^{-7} \) |
|                            | \( P_6 = 0.0003 \)                | \( P_5 = -6.3487 \times 10^{-6} \) |
|                            | \( P_7 = 1.09 \)                   | \( P_5 = 4.6316 \times 10^{-6} \) |
| Gonzalez-Olmos-Iglesias   | \( A_{00} = 1.4584 \)              | \( A_{01} = -0.0014 \) |
|                            | \( A_{10} = -0.4239 \)             | \( A_{11} = 0.0028 \) |
|                            | \( A_{20} = 0.0383 \)              | \( A_{21} = -3.6947 \times 10^{-4} \) |
|                            | \( 10^4 \sigma = 1.09 \)          | \( 10^4 \sigma = 3.14 \) |
| MDEA (1) + DMSO (2)        | \( A_0 = 0.0034 \)                 | \( A_{01} = -0.0015 \) |
|                            | \( A_1 = 1.6658 \)                 | \( A_{02} = 8.2965 \times 10^{-7} \) |
|                            | \( A_2 = 1.4324 \)                 | \( C_1 = 4.2989 \times 10^{-6} \) |
|                            | \( B_1 = -0.0003 \)               | \( C_2 = 4.6600 \times 10^{-7} \) |
|                            | \( B_2 = 0.0006 \)                | \( P_3 = 3.0708 \times 10^{-6} \) |
|                            | \( P_4 = -0.2610 \)               | \( P_5 = -3.4634 \times 10^{-6} \) |
|                            | \( P_7 = 0.3710 \)                | \( P_9 = 4.5659 \times 10^{-6} \) |
| Emmerling et al.           | \( B_1 = 0.0034 \)                 | \( G_{01} = -0.0015 \) |
|                            | \( B_2 = -0.0003 \)               | \( G_{02} = 6.3730 \times 10^{-7} \) |
|                            | \( P_4 = 0.0006 \)                | \( P_5 = 0.0016 \) |
|                            | \( P_7 = 0.3710 \)                | \( P_9 = 3.0582 \times 10^{-6} \) |

1 Units: \( A_i, P_i, P_7, A_{00}, A_{10}, A_{20}, g \cdot cm^{-3}; B_i, P_5, P_9, A_{01}, A_{11}, A_{21}; g \cdot cm^{-3} \cdot K^{-1}; C_i, P_3, P_6, A_{02}, A_{12}, A_{22}; g \cdot cm^{-3} \cdot K^{-2}. \)
### Table A3. Values of parameters for the relations of Grunberg–Nissan, Heric–Brewer, Wilson, Noda–Ishida and Eyring–NRTL and average absolute deviation in the temperature range from 293.15 to 313.15 K.

| Model                          | Parameters and ADD% | T/(K)       |
|-------------------------------|---------------------|-------------|
|                               |                     | 293.15      | 298.15 | 303.15 | 308.15 | 313.15 |
| DEG (1) + DMSO (2)            |                     |             |        |        |        |        |
| Grunberg–Nissan               | \(d\)               | 0.528       | 0.533  | 0.426  | 0.520  | 0.531  |
|                               | ADD%                | 1.03        | 1.91   | 2.00   | 0.84   | 0.96   |
|                               | \(a_{12}\)          | 0.575       | 0.582  | 0.475  | 0.568  | 0.579  |
| Heric–Brewer                  | \(\lambda_{12}\)    | -481.19     | 1985.47| 1754.04| -287.47| 724.45 |
| Wilson                        | \(\lambda_{21}\)    | -681.61     | -1986.45| -1754.39| -868.51| -1501.24|
| Noda and Ishida               | \(w_{12}\)          | -30.23      | -31.10 | -28.29 | -30.77 | -31.31 |
|                               | \(w_{21}\)          | 30.42       | -31.28 | 28.43  | 30.96  | 31.50  |
|                               | ADD%                | 1.02        | 1.90   | 1.97   | 0.85   | 0.93   |
|                               | \(\tau_{12}\)       | -0.455      | -1.227 | -1.380 | -0.544 | -0.780 |
| Eyring-NRTL \(^a\)           | \(\tau_{12}\)       | 1.172       | 2.586  | 2.753  | 1.297  | 1.681  |
|                               | ADD%                | 0.80        | 0.58   | 0.66   | 0.54   | 0.50   |
| MDEA (1) + DMSO (2)           |                     |             |        |        |        |        |
| Grunberg–Nissan               | \(d\)               | 1.627       | 1.517  | 1.383  | 1.224  | 1.250  |
|                               | ADD%                | 10.41       | 10.68  | 9.60   | 8.84   | 8.47   |
|                               | \(a_{12}\)          | 1.719       | 1.610  | 1.474  | 1.316  | 1.342  |
| Heric–Brewer                  | \(\lambda_{12}\)    | 3811.93     | 3720.99| 3538.15| 3393.73| 3422.86|
| Wilson                        | \(\lambda_{21}\)    | -3812.09    | -3727.36| -3555.24| -3399.01| -3425.11|
| Noda and Ishida               | \(w_{12}\)          | -53.26      | -52.00 | -50.55 | -48.21 | -48.71 |
|                               | \(w_{21}\)          | 53.84       | 52.55  | 51.04  | 48.65  | 49.17  |
|                               | ADD%                | 10.43       | 10.68  | 9.62   | 8.88   | 8.49   |
|                               | \(\tau_{12}\)       | 0.014       | -0.136 | -0.049 | -1.329 | -1.362 |
| Eyring-NRTL \(^a\)           | \(\tau_{12}\)       | 14.593      | 14.381 | 15.582 | 8.453  | 7.5085 |
|                               | ADD%                | 2.18        | 1.61   | 1.74   | 3.30   | 3.30   |

\(^a\) Eyring-NRTL as two-parameter model (\(a = 0.20\)).

### Table A4. Polynomial coefficients \(^1\) and standard deviations (\(\sigma\)) for the binary systems in the temperature range from 293.15 to 313.15 K.

| Model                          | Parameters and \(\sigma\) | T/(K)       |
|-------------------------------|-----------------------------|-------------|
|                               |                             | 293.15      | 298.15 | 303.15 | 308.15 | 313.15 |
| DEG (1) + DMSO (2)            | \(V^E/(cm^3\cdot mol^{-1})\) |             |        |        |        |        |
| Redlich-Kister                | \(a_0\)                     | -0.736      | -0.726 | -0.701 | -0.679 | -0.668 |
|                               | \(a_1\)                     | 0.420       | 0.411  | 0.339  | 0.287  | 0.280  |
|                               | \(a_2\)                     | 0.157       | 0.296  | 0.319  | 0.379  | 0.453  |
|                               | \(a_3\)                     | -0.384      | -0.422 | -0.328 | -0.307 | -0.198 |
|                               | \(10^3\cdot \sigma\)        | 3.0         | 3.5    | 2.7    | 3.7    | 2.4    |
| Hwang                         | \(A_0\)                     | -0.785      | -0.821 | -0.805 | -0.803 | -0.817 |
|                               | \(A_1\)                     | 0.545       | 0.703  | 0.689  | 0.717  | 0.853  |
|                               | \(A_2\)                     | -0.139      | 0.071  | 0.150  | 0.285  | 0.348  |
|                               | \(10^3\cdot \sigma\)        | 7.4         | 8.0    | 6.3    | 6.3    | 4.4    |
### Table A4. Cont.

| Model                   | Parameters  | $T/(K)$               |
|-------------------------|-------------|-----------------------|
|                         | and $\sigma$| 293.15 | 298.15 | 303.15 | 308.15 | 313.15 |
| Myers and Scott         | $B_0$      | $-0.730$ | $-0.720$ | $-0.708$ | $-0.680$ | $-0.666$ |
|                         | $B_1$      | $-0.391$ | $-0.343$ | $-0.003$ | $-0.141$ | $0.004$  |
|                         | $B_2$      | $-0.240$ | $0.105$  | $-0.394$ | $0.137$  | $0.518$  |
|                         | $C_0$      | $1.122$  | $1.060$  | $0.556$  | $0.654$  | $0.410$  |
|                         | $C_1$      | $1.001$  | $0.693$  | $1.488$  | $0.680$  | $0.018$  |
|                         | $10^{3} \cdot \sigma$ | $2.9$  | $3.3$   | $2.0$    | $3.9$    | $2.6$    |
| MDEA (1) + DMSO (2)     | $a_0$      | $1.436$  | $1.403$  | $1.364$  | $1.307$  | $1.259$  |
|                         | $a_1$      | $0.164$  | $0.188$  | $0.211$  | $0.268$  | $0.315$  |
|                         | $a_2$      | $-0.274$ | $-0.307$ | $-0.359$ | $-0.411$ | $-0.520$ |
|                         | $a_3$      | $0.208$  | $0.250$  | $0.240$  | $0.264$  | $0.281$  |
|                         | $10^{3} \cdot \sigma$ | $5.1$  | $5.3$   | $8.4$    | $10.0$   | $10.4$   |
| Hwang                   | $A_0$      | $1.527$  | $1.505$  | $1.483$  | $1.444$  | $1.432$  |
|                         | $A_1$      | $-0.084$ | $-0.083$ | $-0.129$ | $-0.120$ | $-0.202$ |
|                         | $A_2$      | $-0.647$ | $-0.737$ | $-0.830$ | $-0.977$ | $-1.186$ |
|                         | $10^{3} \cdot \sigma$ | $5.2$  | $5.6$   | $8.2$    | $9.7$    | $10.1$   |
| Myers and Scott         | $B_0$      | $1.428$  | $1.389$  | $1.348$  | $1.287$  | $1.234$  |
|                         | $B_1$      | $-0.387$ | $-0.586$ | $-0.466$ | $-0.425$ | $-0.313$ |
|                         | $B_2$      | $-1.759$ | $-1.874$ | $-1.742$ | $-1.836$ | $-1.788$ |
|                         | $C_0$      | $-0.394$ | $-0.573$ | $-0.522$ | $-0.545$ | $-0.519$ |
|                         | $C_1$      | $-1.078$ | $-1.152$ | $-1.073$ | $-1.162$ | $-1.125$ |
|                         | $10^{3} \cdot \sigma$ | $3.8$  | $5.4$   | $8.4$    | $7.9$    | $8.2$    |

$\Delta \eta/(\text{mPa-s})$

| DEG (1) + DMSO (2)     | $a_0$      | $-30.94$ | $-23.81$ | $-18.47$ | $-13.15$ | $-10.22$ |
|                         | $a_1$      | $-8.67$  | $-7.84$  | $-7.25$  | $-3.29$  | $-2.78$  |
|                         | $a_2$      | $-6.08$  | $-6.20$  | $-2.65$  | $-1.35$  | $0.31$   |
|                         | $a_3$      | $-6.46$  | $6.31$   | $-0.60$  | $-0.65$  | $1.15$   |
|                         | $\sigma$   | $0.11$   | $0.05$   | $0.06$   | $0.04$   | $0.03$   |
| Hwang                   | $A_0$      | $-28.89$ | $-21.73$ | $-17.60$ | $-12.70$ | $-10.34$ |
|                         | $A_1$      | $-21.19$ | $-20.30$ | $-12.42$ | $-6.00$  | $-2.39$  |
|                         | $A_2$      | $4.89$   | $3.67$   | $5.41$   | $2.40$   | $3.29$   |
|                         | $\nu$      | $0.11$   | $0.07$   | $0.06$   | $0.03$   | $0.04$   |
| MDEA (1) + DMSO (2)     | $a_0$      | $-119.76$| $-85.34$ | $-62.82$ | $-48.12$ | $-34.08$ |
|                         | $a_1$      | $-45.73$ | $-35.93$ | $-22.03$ | $-17.44$ | $-11.65$ |
|                         | $a_2$      | $-19.54$ | $-20.99$ | $-15.89$ | $-7.35$  | $-8.74$  |
|                         | $a_3$      | $-54.82$ | $-43.35$ | $-42.66$ | $-27.20$ | $-23.83$ |
|                         | $\sigma$   | $0.63$   | $0.39$   | $0.37$   | $0.26$   | $0.19$   |
| Hwang                   | $A_0$      | $-113.24$| $-78.34$ | $-57.52$ | $-45.67$ | $-31.17$ |
|                         | $A_1$      | $-103.14$| $-88.66$ | $-64.94$ | $-41.77$ | $-35.30$ |
|                         | $A_2$      | $51.24$  | $32.85$  | $22.75$  | $22.27$  | $12.10$  |
|                         | $\nu$      | $0.80$   | $0.56$   | $0.59$   | $0.38$   | $0.32$   |

$^1$ Units: cm$^3$ mol$^{-1}$ for $V^E$ and mPa-s for $\Delta \eta$. 

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Table A5. Apparent molar volumes of DEG, $V_{\phi,1}$ (cm$^3$·mol$^{-1}$), for binary systems DEG + DMSO and apparent molar volumes of MDEA, $V_{\phi,1}$ (cm$^3$·mol$^{-1}$), for binary system MDEA + DMSO in the temperature range from 293.15 to 313.15 K.

| $x_1$         | T/(K)   |     |     |     |     |
|--------------|---------|-----|-----|-----|-----|
|              | 293.15  | 298.15 | 303.15 | 308.15 | 313.15 |
| DEG (1) + DMSO (2) |         |     |-----|-----|-----|
| 0.0000       | -       |     |     |     |     |
| 0.1062       | 94.499  | 94.922 | 95.293 | 95.770 | 96.147 |
| 0.2007       | 94.759  | 95.157 | 95.560 | 95.983 | 96.366 |
| 0.3034       | 95.077  | 95.445 | 95.861 | 96.287 | 96.679 |
| 0.4110       | 95.391  | 95.775 | 96.172 | 96.591 | 96.994 |
| 0.5108       | 95.645  | 96.035 | 96.440 | 96.858 | 97.270 |
| 0.5972       | 95.837  | 96.236 | 96.648 | 97.068 | 97.487 |
| 0.6955       | 96.023  | 96.433 | 96.853 | 97.280 | 97.707 |
| 0.8096       | 96.203  | 96.626 | 97.055 | 97.486 | 97.925 |
| 0.8819       | 96.306  | 96.736 | 97.171 | 97.605 | 98.049 |
| 1.0000       | 96.455  | 96.895 | 97.340 | 97.779 | 98.232 |
| MDEA (1) + DMSO (2) |         |     |-----|-----|-----|
| 0.0000       | -       |     |     |     |     |
| 0.1003       | 114.274 | 114.764 | 115.141 | 115.450 | 115.770 |
| 0.1967       | 113.424 | 113.859 | 114.243 | 114.619 | 115.008 |
| 0.2997       | 112.488 | 112.975 | 113.405 | 113.843 | 114.263 |
| 0.4006       | 111.671 | 112.167 | 112.618 | 113.053 | 113.511 |
| 0.5001       | 110.936 | 111.429 | 111.886 | 112.343 | 112.815 |
| 0.6020       | 110.265 | 110.760 | 111.229 | 111.700 | 112.182 |
| 0.6937       | 109.730 | 110.226 | 110.706 | 111.183 | 111.671 |
| 0.7983       | 109.186 | 109.682 | 110.169 | 110.654 | 111.151 |
| 0.8967       | 108.731 | 109.226 | 109.719 | 110.208 | 110.711 |
| 1.0000       | 108.310 | 108.805 | 109.304 | 109.797 | 110.305 |

Table A6. Apparent molar volumes of DMSO, $V_{\phi,2}$ (cm$^3$·mol$^{-1}$), for binary systems DEG + DMSO and apparent molar volumes of DMSO, $V_{\phi,2}$ (cm$^3$·mol$^{-1}$), for binary system MDEA + DMSO in the temperature range from 293.15 to 313.15 K.

| $x_1$         | T/(K)   |     |     |     |     |
|--------------|---------|-----|-----|-----|-----|
|              | 293.15  | 298.15 | 303.15 | 308.15 | 313.15 |
| DEG (1) + DMSO (2) |         |     |-----|-----|-----|
| 0.0000       | 71.014  | 71.339 | 71.656 | 71.989 | 72.322 |
| 0.1062       | 70.966  | 71.304 | 71.633 | 71.968 | 72.304 |
| 0.2007       | 70.899  | 71.244 | 71.585 | 71.917 | 72.254 |
| 0.3034       | 70.832  | 71.166 | 71.522 | 71.862 | 72.201 |
| 0.4110       | 70.780  | 71.124 | 71.462 | 71.801 | 72.145 |
| 0.5108       | 70.732  | 71.090 | 71.432 | 71.767 | 72.114 |
| 0.5972       | 70.752  | 71.095 | 71.438 | 71.764 | 72.304 |
| 0.6955       | 70.765  | 71.120 | 71.466 | 71.808 | 72.158 |
| 0.8096       | 70.725  | 71.111 | 71.467 | 71.799 | 72.211 |
| 0.8819       | 70.792  | 71.195 | 71.533 | 71.899 | 72.327 |
| 1.0000       | -       |     |-----|-----|-----|


Table A6. Cont.

| $x_1$ | T/(K)       |
|-------|-------------|
|       | 293.15      | 298.15      | 303.15      | 308.15      | 313.15      |
| MDEA (1) + DMSO (2) |            |             |             |             |             |
| 0.0000 | 71.014      | 71.339      | 71.666      | 71.989      | 72.322      |
| 0.1003 | 71.004      | 71.332      | 71.660      | 71.972      | 72.295      |
| 0.1967 | 71.080      | 71.389      | 71.709      | 72.018      | 72.340      |
| 0.2997 | 71.188      | 71.514      | 71.842      | 72.167      | 72.482      |
| 0.4006 | 71.337      | 71.670      | 72.004      | 72.302      | 72.632      |
| 0.5001 | 71.485      | 71.807      | 72.126      | 72.434      | 72.764      |
| 0.6020 | 71.634      | 71.959      | 72.273      | 72.597      | 72.923      |
| 0.6937 | 71.794      | 72.121      | 72.455      | 72.765      | 73.076      |
| 0.7983 | 72.003      | 72.333      | 72.655      | 72.986      | 73.320      |
| 0.8967 | 72.066      | 72.397      | 72.666      | 72.944      | 73.224      |
| 1.0000 | -           | -           | -           | -           | -           |

Table A7. Partial molar volumes $\bar{V}_1$ (cm$^3$·mol$^{-1}$) for binary systems in the temperature range from 293.15 to 313.15 K.

| $x_1$ | T/(K)       |
|-------|-------------|
|       | 293.15      | 298.15      | 303.15      | 308.15      | 313.15      |
| DEG (1) + DMSO (2) |            |             |             |             |             |
| 0.0000 | 94.262      | 94.773      | 95.142      | 95.635      | 95.966      |
| 0.1062 | 94.243      | 94.679      | 95.067      | 95.533      | 95.888      |
| 0.2007 | 94.286      | 94.675      | 95.067      | 95.513      | 95.876      |
| 0.3034 | 94.375      | 94.728      | 95.115      | 95.541      | 95.904      |
| 0.4110 | 94.490      | 94.820      | 95.197      | 95.606      | 95.964      |
| 0.5108 | 94.600      | 94.918      | 95.284      | 95.681      | 96.033      |
| 0.5972 | 94.687      | 95.001      | 95.358      | 95.748      | 96.096      |
| 0.6955 | 94.770      | 95.082      | 95.431      | 95.816      | 96.161      |
| 0.8096 | 94.837      | 95.150      | 95.494      | 95.875      | 96.221      |
| 0.8819 | 94.862      | 95.176      | 95.519      | 95.900      | 96.246      |
| 1.0000 | 94.877      | 95.192      | 95.535      | 95.915      | 96.263      |
| MDEA (1) + DMSO (2) |            |             |             |             |             |
| 0.0000 | 115.271     | 115.647     | 115.955     | 116.191     | 116.400     |
| 0.1003 | 115.225     | 115.644     | 115.978     | 116.266     | 116.539     |
| 0.1967 | 115.145     | 115.593     | 115.947     | 116.274     | 116.595     |
| 0.2997 | 115.034     | 115.505     | 115.877     | 116.235     | 116.595     |
| 0.4006 | 114.915     | 115.400     | 115.786     | 116.167     | 116.555     |
| 0.5001 | 114.799     | 115.292     | 115.689     | 116.087     | 116.492     |
| 0.6020 | 114.690     | 115.190     | 115.594     | 116.003     | 116.421     |
| 0.6937 | 114.607     | 115.110     | 115.519     | 115.936     | 116.360     |
| 0.7983 | 114.536     | 115.042     | 115.453     | 115.876     | 116.303     |
| 0.8967 | 114.495     | 115.003     | 115.415     | 115.840     | 116.269     |
| 1.0000 | 114.481     | 114.989     | 115.401     | 115.827     | 116.257     |
Table A8. Partial molar volumes $\bar{V}_2$ (cm$^3$·mol$^{-1}$) for binary systems in the temperature range from 293.15 to 313.15 K.

| $x_1$ | 293.15 | 298.15 | 303.15 | 308.15 | 313.15 |
|-------|--------|--------|--------|--------|--------|
| DEG (1) + DMSO (2) |        |        |        |        |        |
| 0.0000 | 71.014 | 71.338 | 71.666 | 71.989 | 72.322 |
| 0.1062 | 71.007 | 71.329 | 71.656 | 71.979 | 72.313 |
| 0.2007 | 70.992 | 71.313 | 71.639 | 71.960 | 72.293 |
| 0.3034 | 70.974 | 71.293 | 71.616 | 71.935 | 72.267 |
| 0.4110 | 70.954 | 71.274 | 71.593 | 71.910 | 72.242 |
| 0.5108 | 70.933 | 71.258 | 71.573 | 71.889 | 72.224 |
| 0.5972 | 70.907 | 71.241 | 71.554 | 71.871 | 72.213 |
| 0.6955 | 70.863 | 71.211 | 71.527 | 71.849 | 72.206 |
| 0.8096 | 70.774 | 71.146 | 71.475 | 71.809 | 72.202 |
| 0.8819 | 70.688 | 71.078 | 71.425 | 71.770 | 72.200 |
| 1.0000 | 70.471 | 70.897 | 71.295 | 71.669 | 72.189 |
| MDEA (1) + DMSO (2) |        |        |        |        |        |
| 0.0000 | 71.014 | 71.338 | 71.666 | 71.989 | 72.322 |
| 0.1003 | 71.034 | 71.359 | 71.666 | 72.010 | 72.344 |
| 0.1967 | 71.086 | 71.411 | 71.738 | 72.064 | 72.400 |
| 0.2997 | 71.171 | 71.496 | 71.823 | 72.149 | 72.487 |
| 0.4006 | 71.281 | 71.605 | 71.931 | 72.257 | 72.593 |
| 0.5001 | 71.414 | 71.766 | 72.080 | 72.383 | 72.716 |
| 0.6020 | 71.577 | 71.896 | 72.214 | 72.534 | 72.859 |
| 0.6937 | 71.748 | 72.065 | 72.374 | 72.690 | 73.003 |
| 0.7983 | 71.977 | 72.292 | 72.587 | 72.897 | 73.192 |
| 0.8867 | 72.231 | 72.548 | 72.825 | 73.127 | 73.399 |
| 1.0000 | 72.548 | 72.872 | 73.122 | 73.417 | 73.657 |

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