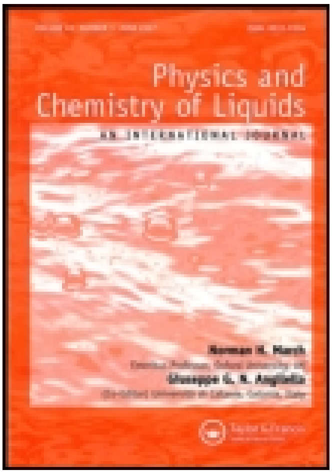

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# Viscosity and surface tension of glycerol + *N*-methyl-2-pyrrolidone mixtures from 293 to 323 K

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The viscosity and surface tension of binary mixtures of glycerol (1) + *N*-methyl-2-pyrrolidone (2) at $T$ = 293, 298, 303, 308, 313, 318, and 323 K and at atmospheric pressure are reported. The Jouyban–Acree model was used for mathematical correlation of measured data. The average percentage deviation was used as an accuracy criterion. Derived properties from measured thermophysical properties, including deviations in viscosity and surface tension; the enthalpy, entropy and Gibbs energy of flow activation; standard enthalpy and entropy of surface formation data were reported.

**Keywords:** viscosity; surface tension; Jouyban–Acree model; glycerol; *N*-methyl-2-pyrrolidone

## 1. Introduction

Glycerol is a pharmaceutical cosolvent [1] with the chemical systematic name of 1,2,3-propanetriol. Glycerol is a component of triglycerides and could be produced from their hydrolysis. Nitroglycerin is its trinitrated derivative and it is an explosive with vasodilation effects on vascular system. In addition to pharmaceutical applications of glycerol, it has many other applications in the chemical industry. *N*-methyl-2-pyrrolidone or 1-methyl-2-pyrrolidone is another pharmaceutical cosolvent. [1] *N*-methyl-2-pyrrolidone is used in the pharmaceutical products, and its applications in the pharmaceutical area have been reviewed. [2] Glycerol and *N*-methyl-2-pyrrolidone were added to water for enhancing the solubility of low water-soluble pharmaceuticals and also used as their individual or mixtures for various purposes. [3–11] The viscosity and/or surface tension of glycerol [12–27] and *N*-methyl-2-pyrrolidone [28–33] have been reported in the literature. To the best of our knowledge, there is no report on the properties of glycerol + *N*-methyl-2-pyrrolidone mixtures in the literature. Figure 1 shows the chemical structures of the investigated solvents.

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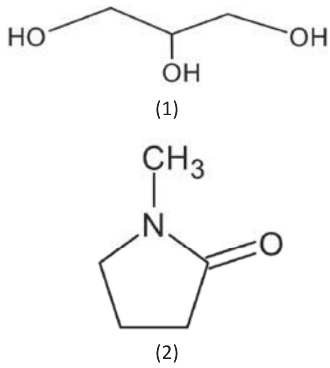

Figure 1. Chemical structures of glycerol (1) and *N*-methyl-2-pyrrolidone (2).

Viscosity and surface tension of liquids are crucial physicochemical properties (PCPs) affecting mass and heat transfer processes in the solutions. Therefore, these data points could be used in the designing and engineering of such processes. However, in most cases, different blends of solvents are used, and it has been observed that PCPs of solvent mixtures show deviation from ideal mixing. Because of this fact finding an appropriate solvent mixture for a desired amount of PCP requires some measurements or accurate predictions. One of the most accurate equations for calculating the PCPs of solvent mixtures is the Jouyban–Acree model, which is expressed as follows for the correlation of PCPs of the binary solvent mixtures [34–37]:

$$\begin{aligned}\ln PCP_{m,T} &= x_1 \cdot \ln PCP_{1,T} + x_2 \cdot \ln PCP_{2,T} \\ &\quad + J_0\left[\frac{x_1 \cdot x_2}{T}\right] + J_1\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] + J_2\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}\right]\end{aligned} \tag{1a}$$

where $PCP_{m,\mathrm{T}}$, $PCP_{1,T}$, $PCP_{2,T}$ are the PCPs under study of the mixture, solvent 1 and solvent 2, respectively; $x_1$ and $x_2$ denote mole fractions of the solvents 1 and 2, respectively; $T$ is the absolute temperature; and $J_\mathrm{i}$ terms are the coefficients of the model which are computable using a no-intercept regression of the experimental data as follows [38]:

$$\begin{aligned}\ln PCP_{m,T} - x_1 \cdot \ln PCP_{1,T} - x_2 \cdot \ln PCP_{2,T} &= J_0\left[\frac{x_1 \cdot x_2}{T}\right] \\ &\quad + J_1\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] + J_2\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}\right]\end{aligned} \tag{1b}$$

To continue our previous studies on PCPs of mixed solvents, [39–42] the aims of this work are to report experimental viscosity and surface tension values of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures at different temperatures at atmospheric pressure, to give predictive models for viscosity, and surface tension values of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures at different temperatures, and to compare the accuracies of available models from the literature to represent the generated data. Although the accuracy of the Jouyban–Acree model for representing viscosity [35] and surface tension [36] of various binary solvents has been evaluated, yet the model was not tested on glycerol + *N*-methyl-2-pyrrolidone mixtures, which are investigated in this work. The results were also compared with those of similar algorithms from the literature.

## 2. Experimental

### 2.1. *Reagents*

Glycerol A.R. grade (mass fraction purity of 99.4%) and *N*-methyl-2-pyrrolidone anhydrous A.R. grade (mass fraction purity of 99.5%) were purchased from S.D. Fine Chemicals (India) and Sigma-Aldrich (USA), respectively. Chemicals were used as received from the companies.

### 2.2. *Apparatus and procedures*

Appropriate proportions of the solvents were mixed with each other volumetrically (with uncertainty of 0.1 mL) to produce different blends of glycerol and *N*-methyl-2-pyrrolidone mixtures by intervals of 0.10 in volume fraction. The prepared solvent mixtures were placed on a shaking water bath (Memmert, Germany) with a set temperature accuracy of 0.1 K (fluctuation ±0.1 K) at 293, 298, 303, 308, 313, 318, and 323 K temperatures. After assurance of equilibrium, the samples were analysed. The viscosity of the solvent mixtures was measured using an Ostwald U-tube glass viscometer (Union Scientific Appliances, India) suspended in a constant temperature water bath. Surface tension of the prepared blends of solvents was determined by drop number method. The accuracies for reported data are 0.01 (mPa s) and 0.01 (mN $m^{-1}$) for viscosity and surface tension, respectively. The standard deviations of the repeated measurements for viscosity are 0.02, 2.36 and with the overall relative standard deviation of 0.71%. The corresponding values are 0.01, 1.75 and 0.80% for surface tension measurements, respectively. All measurements were done at least in triplicates.

### 2.3. *Computational methods*

The Jouyban–Acree model is used as an algorithm to calculate the studied PCPs in solvent mixtures. For each property, it is trained with experimental data using a no-intercept regression analysis. For some mixtures, ideal mixing behaviour could also be used to provide simpler prediction method. The model could be represented as:

$$\ln PCP_{m,T} = x_1 \cdot \ln PCP_{1,T} + x_2 \cdot \ln PCP_{2,T} \tag{2}$$

The average percentage deviation (*APD*) was used as an error criterion which is defined as:

$$APD = \frac{100}{NDP} \sum \frac{\left|PCP_{Exp} - PCP_{Cal}\right|}{PCP_{Exp}} \tag{3}$$

where *NDP* is the number of data points in each set; $PCP_{\mathrm{Exp}}$ and $PCP_{\mathrm{Cal}}$ are the experimental and calculated PCPs under investigation.

The common models from the literature are used to represent the data, and their accuracies are compared with the Jouyban–Acree model.

Arrhenius proposed the following equation for calculating viscosity of the ideal liquid mixtures [43]:

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 \tag{4}$$

where $\eta_m$ is the viscosity of the mixture, $\eta_1$ and $\eta_2$ are the viscosities of neat liquids 1 and 2 at a given $T$, respectively. The viscosity of real mixtures shows deviations from ideal values predicted by Equation (4). In order to cover the deviations, Grunberg and Nissan [44] added one more variable to Equation (4) and obtained:

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 + d x_1 x_2 \tag{5}$$

where $d$ is a curve-fit parameter. They found that $d$ is positive for liquid mixtures showing negative deviation from Raoult's law, and it is negative for systems exhibiting positive deviation from Raoult's law. [44] Equation (5) was used to correlate the viscosity of mixtures at isothermal condition, but it could be rewritten as

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + d' \left( \frac{x_1 x_2}{T} \right) \tag{6}$$

where $d'$ is a model constant. Equation (6) is a simplified version of our proposed model and a theoretical justification could be found for this extension. [35]

The available models for correlating surface tension of mixed solvents with respect to solvent composition and temperature from the literature are the combined Connors–Wright–Hoke–Patton [35] and Lee models. [45] Connors and Wright [46] proposed Equation (7) for correlating the surface tension as a function of solvent composition

$$\sigma_m = \sigma_1 - \left[ 1 + \frac{b x_1}{1 - a x_1} \right] x_2 (\sigma_1 - \sigma_2) \tag{7}$$

where $a$ and $b$ are the model constants. Hoke and Patton [47] correlated the values of $a$ and $b$ with temperature

$$a = H_0 + H_1 T + H_2 T^2 \tag{8}$$

$$b = P_0 + P_1 T + P_2 T^2 \tag{9}$$

replacing $a$ and $b$ values from Equations (8) and (9) into Equation (7) yields [35]:

$$\sigma_{m,T} = \sigma_{1,T} - \left[ 1 + \frac{x_1 (P_0 + P_1 T + P_2 T^2)}{1 - x_1 (H_0 + H_1 T + H_2 T^2)} \right] x_2 \left( \sigma_{1,T} - \sigma_{2,T} \right) \tag{10}$$

The Lee model [45] is as follows:

$$\sigma_{m,T} = L_0 + L_1 x_1 + L_2 x_1^2 + L_3 T + L_4 x_1 T + L_5 x_1^2 T + L_6 T^2 + L_7 x_1 T^2 + L_8 x_1^2 T^2 \tag{11}$$

where $L$ terms are the model constants.

## 3. Results and discussion

Experimental values of viscosity and surface tension are tabulated in Table 1. Available experimental data of viscosity and surface tension of glycerol and *N*-methyl-2-pyrrolidone at different temperatures were compared with the reported data in Table 2. Relatively higher deviations were observed for the reported data by Leron *et al.* [19]. There are generally good agreements between generated viscosity data with those taken from the literature. The only

Table 1. Volume fraction ($\varphi_2$) and mole fraction ($\boldsymbol{x_2}$) of *N*-methyl-2-pyrrolidone, viscosity ($\eta$) and surface tension ($\sigma$) of the glycerol (1) + *N*-methyl-2-pyrrolidone (2) binary mixtures and their standard deviations **(SD)** at different temperatures ($T$)[a].

| $\varphi_2$ | $x_2$ | $\eta$/mPa s | $\sigma$/mN m$^{-1}$ |
|---|---|---|---|
| $T$ = 293 K | | | |
| 0.000 | 0.000 | 1410.97 (0.74) | 64.00 (0.93) |
| 0.100 | 0.078 | 619.80 (0.22) | 63.76 (0.84) |
| 0.200 | 0.160 | 409.49 (0.27) | 62.19 (0.94) |
| 0.400 | 0.337 | 105.71 (0.21) | 57.81 (1.46) |
| 0.500 | 0.433 | 46.89 (0.15) | 55.07 (0.12) |
| 0.600 | 0.534 | 25.34 (0.09) | 53.17 (1.49) |
| 0.700 | 0.640 | 12.41 (0.09) | 50.36 (1.22) |
| 0.800 | 0.753 | 4.63 (0.04) | 48.42 (0.67) |
| 0.900 | 0.873 | 2.70 (0.02) | 44.82 (0.01) |
| 1.000 | 1.000 | 1.70 (0.02) | 42.38 (0.56) |
| $T$ = 298 K | | | |
| 0.000 | 0.000 | 946.53 (0.57) | 62.23 (1.69) |
| 0.100 | 0.078 | 418.36 (0.10) | 60.90 (1.13) |
| 0.200 | 0.160 | 269.42 (0.23) | 59.10 (1.72) |
| 0.400 | 0.338 | 72.84 (0.85) | 55.22 (0.75) |
| 0.500 | 0.433 | 38.94 (0.03) | 53.15 (1.50) |
| 0.600 | 0.534 | 20.42 (0.09) | 51.18 (0.08) |
| 0.700 | 0.641 | 11.12 (0.07) | 48.55 (0.66) |
| 0.800 | 0.754 | 4.23 (0.04) | 45.97 (0.60) |
| 0.900 | 0.873 | 2.44 (0.04) | 43.21 (1.14) |
| 1.000 | 1.000 | 1.61 (0.02) | 41.58 (0.55) |
| $T$ = 303 K | | | |
| 0.000 | 0.000 | 611.35(0.94) | 60.94 (1.75) |
| 0.100 | 0.078 | 289.94 (0.17) | 58.96 (0.53) |
| 0.200 | 0.160 | 160.52 (0.12) | 57.32 (1.41) |
| 0.400 | 0.336 | 55.93 (0.07) | 53.37 (0.69) |
| 0.500 | 0.432 | 30.16 (0.07) | 51.55 (0.08) |
| 0.600 | 0.533 | 17.83 (0.14) | 49.67 (0.68) |
| 0.700 | 0.640 | 9.48 (0.11) | 47.41 (0.34) |
| 0.800 | 0.753 | 3.75 (0.04) | 44.52 (0.01) |
| 0.900 | 0.872 | 2.26 (0.04) | 42.65 (0.33) |
| 1.000 | 1.000 | 1.50 (0.02) | 41.02 (0.01) |
| $T$ = 308 K | | | |
| 0.000 | 0.000 | 413.90 (0.46) | 59.57 (1.47) |
| 0.100 | 0.077 | 211.32 (0.07) | 57.52 (0.86) |
| 0.200 | 0.159 | 118.25 (0.09) | 55.72 (0.84) |
| 0.400 | 0.335 | 43.40 (0.05) | 51.55 (0.73) |
| 0.500 | 0.430 | 25.79 (0.11) | 50.45 (0.57) |
| 0.600 | 0.531 | 15.16 (0.09) | 48.53 (0.37) |

(*continued*)

Table 1. (Continued).

| $\varphi_2$ | $x_2$ | $\eta$/mPa s | $\sigma$/mN m$^{-1}$ |
|---|---|---|---|
| 0.700 | 0.638 | 7.70 (0.09) | 46.27 (0.53) |
| 0.800 | 0.751 | 3.27 (0.04) | 43.25 (0.32) |
| 0.900 | 0.872 | 2.02 (0.02) | 41.86 (0.21) |
| 1.000 | 1.000 | 1.39 (0.01) | 40.39 (0.53) |
| $T$ = 313 K | | | |
| 0.000 | 0.000 | 285.47 (0.47) | 58.92 (0.48) |
| 0.100 | 0.077 | 143.39 (0.10) | 56.86 (0.80) |
| 0.200 | 0.158 | 87.14 (0.07) | 54.55 (0.79) |
| 0.400 | 0.334 | 35.17 (0.05) | 51.18 (0.46) |
| 0.500 | 0.430 | 20.73 (0.06) | 49.10 (0.68) |
| 0.600 | 0.530 | 10.45 (0.09) | 47.28 (0.83) |
| 0.700 | 0.637 | 6.13 (0.11) | 45.71 (0.80) |
| 0.800 | 0.751 | 2.91 (0.02) | 42.65 (0.59) |
| 0.900 | 0.871 | 1.88 (0.04) | 41.52 (0.54) |
| 1.000 | 1.000 | 1.29 (0.02) | 40.27 (0.42) |
| $T$ = 318 K | | | |
| 0.000 | 0.000 | 213.68 (0.92) | 58.02 (0.79) |
| 0.100 | 0.077 | 106.05 (0.12) | 56.14 (0.72) |
| 0.200 | 0.158 | 54.08 (0.21) | 53.92 (0.74) |
| 0.400 | 0.334 | 25.00 (0.07) | 49.90 (1.15) |
| 0.500 | 0.429 | 16.10 (0.09) | 48.34 (0.26) |
| 0.600 | 0.530 | 9.03 (0.07) | 46.36 (0.61) |
| 0.700 | 0.637 | 5.17 (0.09) | 44.75 (1.01) |
| 0.800 | 0.750 | 2.68 (0.02) | 42.41 (0.40) |
| 0.900 | 0.871 | 1.72 (0.02) | 40.99 (0.52) |
| 1.000 | 1.000 | 1.22 (0.02) | 40.19 (0.83) |
| $T$ = 323 K | | | |
| 0.000 | 0.000 | 140.97 (0.58) | 57.68 (0.46) |
| 0.100 | 0.077 | 83.45 (0.14) | 55.91 (0.77) |
| 0.200 | 0.158 | 50.16 (0.09) | 53.34 (1.43) |
| 0.400 | 0.334 | 21.33 (0.38) | 49.57 (1.14) |
| 0.500 | 0.429 | 13.43 (0.09) | 47.69 (0.04) |
| 0.600 | 0.530 | 7.11 (0.11) | 45.93 (1.18) |
| 0.700 | 0.637 | 4.64 (0.09) | 44.69 (1.01) |
| 0.800 | 0.750 | 2.46 (0.02) | 42.17 (0.53) |
| 0.900 | 0.871 | 1.60 (0.02) | 40.90 (0.52) |
| 1.000 | 1.000 | 1.12 (0.04) | 40.08 (0.07) |

Note: [a]Standard uncertainties $u$ are $u(T)$ = 0.1 K, $u(\varphi_2) = 0.001$, $u(x_2) = 0.001$, $u(\eta) = 0.001$ mPa s and $u(\sigma) = 0.01$ mN m$^{-1}$.

point is less viscosity of *N*-methyl-2-pyrrolidone at 293 K. Good agreements were observed among surface tension data.

The resulted equations for viscosity and surface tension calculations using the Jouyban–Acree model after excluding non-significant ($p > 0.10$) constants are:

$$\ln \eta_{m,T} = x_1 \cdot \ln \eta_{1,T} + x_2 \cdot \ln \eta_{2,T} - 472.197\left[\frac{x_1 \cdot x_2}{T}\right] + 203.498\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] - 866.901\left[\frac{x_1 \cdot x_2 \cdot (x_2 - x_2)^2}{T}\right] \tag{12}$$

Table 2. Comparison between measured (expt) data for viscosity, and surface tension of glycerol and N-methyl-2-pyrrolidone at different temperatures with the literature data and the individual percentage deviations (IPD)[a].

| | $\eta$ (mPa s) | | $\sigma$ (mN m$^{-1}$) | | | |
|---|---|---|---|---|---|---|
| $T$ (K) | expt | Literature | IPD Glycerol | expt | literature | IPD |
| 293.15 | 1410.97 | 1410 [12] | 0.1 | 64.00 | 63.4 [12] | 0.9 |
| 293.15 | 1410.97 | 1412 [14] | −0.1 | | | |
| 298.15 | 946.53 | 938.025 [20] | 0.9 | 62.23 | 62.5 [21] | −0.4 |
| 298.15 | 946.53 | 875 [12] | 8.2 | 62.23 | 61.81 [22] | 0.7 |
| 298.15 | 946.53 | 943.8 [23] | 0.3 | | | |
| 303.15 | 611.35 | 612 [14] | −0.1 | 60.93 | 62.1 [2] | −1.9 |
| 303.15 | 611.35 | 612 [12] | −0.1 | 60.93 | 61.33 [22] | −0.7 |
| 303.15 | 611.35 | 628.9 [23] | −2.8 | | | |
| 308.15 | 413.9 | 431.9 [23] | −4.2 | 59.57 | 60.99 [22] | −2.3 |
| 313.15 | 285.47 | 284 [14] | 0.5 | 58.91 | 61.5 [2] | −4.2 |
| 313.15 | 285.47 | 284 [12] | 0.5 | | | |
| 313.15 | 285.47 | 295.9 [23] | −3.5 | | | |
| 318.15 | 213.68 | 185.8 [23] | 15.0 | | | |
| 323.15 | 140.97 | 142 [2] | −0.7 | 57.68 | 61.1 [2] | −5.6 |
| 323.15 | 140.97 | 142 [14] | −0.7 | | | |
| 323.15 | 140.97 | 151.2 [23] | −6.8 | | | |
| *N*-methyl-2-pyrrolidone | | | | | | |
| 293.15 | 1.70 | 1.899 [28] | −10.5 | 42.38 | 42.7 [28] | −0.7 |
| 293.15 | 1.70 | 1.8581 [29] | −8.5 | | | |
| 298.15 | 1.61 | 1.683 [30] | −4.3 | 41.58 | 40.7 [2] | 2.2 |
| 298.15 | 1.61 | 1.656 [31] | −2.8 | | | |
| 298.15 | 1.61 | 1.714 [32] | −6.1 | 41.58 | 40.8 [32] | 1.9 |
| 298.15 | 1.61 | 1.663 [33] | −3.2 | | | |
| 303.15 | 1.49 | 1.554 [30] | −4.1 | | | |
| 303.15 | 1.49 | 1.592 [28] | −6.4 | 41.01 | 41.5 [28] | −1.2 |
| 303.15 | 1.49 | 1.5531 [29] | −4.1 | | | |
| 313.15 | 1.39 | 1.332 [30] | 4.4 | | | |
| 313.15 | 1.39 | 1.353 [28] | 2.7 | 40.27 | 40.8 [28] | −1.3 |
| 313.15 | 1.39 | 1.3239 [29] | 5.0 | | | |
| 323.15 | 1.12 | 1.16 [30] | −3.4 | | | |
| 323.15 | 1.12 | 1.17 [28] | −4.3 | 40.08 | 40 [28] | 0.2 |
| 323.15 | 1.12 | 1.1461 [29] | −2.3 | | | |

Note: [a]*IPD* is calculated using: $IPD = 100 \times \left(\frac{PCP_{\text{measured}} - PCP_{\text{literature}}}{PCP_{\text{literature}}}\right)$.

$$\ln \sigma_{m,T} = x_1 \cdot \ln \sigma_{1,T} + x_2 \cdot \ln \sigma_{2,T} - 7.726\left[\frac{x_1 \cdot x_2}{T}\right] + 21.945\left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] \tag{13}$$

where $\eta$ and $\sigma$ are the indicators of viscosity and surface tension, respectively; 1, 2, and $m$ subscripts stand for glycerol, *N*-methyl-2-pyrrolidone and their mixtures, respectively. All these models correlated perfectly the investigated properties with respect to the solvent composition and temperature with the APD values of 4.7 ± 4.1%, and 1.4 ± 1.2% for viscosity and surface tension, respectively. Accurate correlation of PCP data of binary solvent mixtures at various temperatures is the main advantage of the Jouyban–Acree model, while the requirement of PCPs of the mono-solvents at each temperature of interest is its main disadvantage.

The trained version of the previous model to represent viscosity of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures is:

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} - 1.910 \left[\frac{x_1 x_2}{T}\right] \tag{14}$$

which is correlated the viscosity data with the APD of 8.6 ± 8.2%, which is significantly more than that of Equation (12) (paired *t*-test, $p < 0.0005$). The obtained APD of Equation (2) for viscosity data was 36.9 ± 23.1%, which was significantly more than those of Equations (12) and (14).

The models for representing surface tension data are:

$$\sigma_{m,T} = \sigma_{1,T} - \left[1 + \frac{x_1(728.257 + 66.564T - 1.599T^2)}{1 - x_1(-1468.066 - 28.144T + 1.130T^2)}\right] x_2\left(\sigma_{1,T} - \sigma_{2,T}\right) \tag{15}$$

and

$$\sigma_{m,T} = 49.247 + 29.711x_1 - 0.433T - 0.429x_1 T + 0.005T^2 + 0.004x_1^2 T^2 \tag{16}$$

Equations (15) and (16) back-calculated the surface tension data of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures at various temperatures with APDs of 5.4 ± 13.2% and 0.6 ± 0.5%, respectively. The APD of Equation (15) is significantly more than that of Equation (13) (paired *t*-test, $p < 0.05$), whereas the APD of Equation (16) is significantly less than that of Equation (13) (paired *t*-test, $p < 0.05$).

The thermophysical properties reported here allow us to make an estimation of the kind of interactions present among the components in the mixtures. Tables 3 and 4 show the deviations in viscosity $(\Delta\eta)$ and surface tension $(\Delta\sigma)$. These derived properties were calculated by means of Equations (17) [48] and (18) [49], respectively.

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{17}$$

Table 3. Deviations in viscosity (mPa s) of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures in front to the additive behaviour at several temperatures.

| $\varphi_2$ | $x_2$ | 293 K | 298 K | 303 K | 308 K | 313 K | 318 K | 323 K |
|---|---|---|---|---|---|---|---|---|
| 0.000 | 0.000 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| 0.100 | 0.078 | −681 | −454 | −274 | −170 | −120 | −91 | −47 |
| 0.200 | 0.160 | −776 | −526 | −353 | −230 | −153 | −125 | −68 |
| 0.400 | 0.337 | −830 | −555 | −350 | −231 | −155 | −117 | −73 |
| 0.500 | 0.433 | −754 | −498 | −317 | −209 | −142 | −106 | −67 |
| 0.600 | 0.534 | −633 | −422 | −268 | −178 | −123 | −91 | −59 |
| 0.700 | 0.640 | −497 | −331 | −212 | −142 | −97 | −73 | −47 |
| 0.800 | 0.753 | −345 | −231 | −148 | −100 | −69 | −51 | −33 |
| 0.900 | 0.873 | −178 | −119 | −77 | −52 | −36 | −26 | −17 |
| 1.000 | 1.000 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |

Note: $\varphi_2$ and $x_2$ are volume and mole fractions of solvent 2 in the binary mixture.

Table 4. Deviations in surface tension (mN m$^{-1}$) of glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures in front to the additive behaviour at several temperatures.

| $\varphi_2$ | $x_2$ | 293 K | 298 K | 303 K | 308 K | 313 K | 318 K | 323 K |
|---|---|---|---|---|---|---|---|---|
| 0.000 | 0.000 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| 0.100 | 0.078 | 1.45 | 0.28 | −0.43 | −0.55 | −0.61 | −0.49 | −0.40 |
| 0.200 | 0.160 | 1.65 | 0.17 | −0.43 | −0.78 | −1.39 | −1.25 | −1.52 |
| 0.400 | 0.337 | 1.10 | −0.05 | −0.86 | −1.56 | −1.45 | −2.11 | −2.18 |
| 0.500 | 0.433 | 0.43 | −0.14 | −0.76 | −0.82 | −1.74 | −1.95 | −2.37 |
| 0.600 | 0.534 | 0.72 | −0.02 | −0.63 | −0.80 | −1.68 | −2.14 | −2.35 |
| 0.700 | 0.640 | 0.20 | −0.46 | −0.78 | −1.02 | −1.27 | −1.86 | −1.73 |
| 0.800 | 0.753 | 0.70 | −0.71 | −1.42 | −1.88 | −2.23 | −2.18 | −2.26 |
| 0.900 | 0.873 | −0.31 | −0.99 | −0.90 | −0.97 | −1.12 | −1.46 | −1.42 |
| 1.000 | 1.000 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |

Note: $\varphi_2$ and $x_2$ are volume and mole fractions of solvent 2 in the binary mixture.

$$\Delta\sigma = \sigma - (x_1\sigma_1 + x_2\sigma_2) \tag{18}$$

The $\Delta\eta$ values are negative in all compositions and temperatures, the maximal deviation is found for 0.337 in mole fraction of *N*-methyl-2-pyrrolidone where there is one molecule of *N*-methyl-2-pyrrolidone for two molecules of glycerol. These values diminish with the increase in temperature. Finally, $\Delta\sigma$ values were also negative, except at 293 K, and they tend to increase as temperature increases.

On the other hand, by evaluating the variation of viscosity and surface tension the respective thermodynamic quantities of flow activation (fa) and surface formation (sf) were calculated according to the following equations [50,51]:

$$R\ln\left(\frac{\eta V}{hN}\right) = \left(\frac{\Delta_{\mathrm{fa}}H^{\ddagger}}{T}\right) - \Delta_{\mathrm{fa}}S^{\ddagger} \tag{19}$$

$$\Delta_{\mathrm{fa}}G^{\ddagger} = \Delta_{\mathrm{fa}}H^{\ddagger} - T\Delta_{\mathrm{fa}}S^{\ddagger} \tag{20}$$

$$\Delta_{\mathrm{sf}}H^{\circ} = \sigma - T\left(\frac{\partial\sigma}{T}\right)_{p,x} \tag{21}$$

$$\Delta_{\mathrm{sf}}S^{\circ} = -\left(\frac{\partial\sigma}{T}\right)_{p,x} \tag{22}$$

where $\Delta_{\mathrm{fa}}H^{\ddagger}$, $\Delta_{\mathrm{fa}}S^{\ddagger}$, and $\Delta_{\mathrm{fa}}G^{\ddagger}$ are the enthalpy, entropy and Gibbs energy of activation of flow, respectively; $\Delta_{\mathrm{sf}}H^{\circ}$ and $\Delta_{\mathrm{sf}}S^{\circ}$ are the standard enthalpy and entropy of surface formation, respectively and finally, $V$ is the molar volume (expressed in m$^3$ mol$^{-1}$); $h$ is the Planck constant and $N$ is the Avogadro number. Therefore, the values of enthalpy and entropy of flow activation were calculated from the slopes and intercepts of plots of $R\ln(\eta V/hN)$ as function of $1/T$ by means of linear regression analysis as described by Equation (19).

Table 5. Thermodynamic parameters of flow activation and surface formation of glycerol (1) + ***N***-methyl-2-pyrrolidone (2) mixtures at 298 *K*.

| $\varphi_2$ | $x_2$ | $\Delta_{fa}H^{\ddagger}$ (kJ mol$^{-1}$) | $\Delta_{fa}S^{\ddagger}$ (J mol$^{-1}$ K$^{-1}$) | $\Delta_{fa}G^{\ddagger}$ (kJ mol$^{-1}$) | $\Delta_{sf}H^{\circ}$ (mJ m$^{-2}$) | $\Delta_{sf}S^{\circ}$ (mJ m$^{-2}$·K$^{-1}$) |
|---|---|---|---|---|---|---|
| 0.000 | 0.000 | 59.4 | 99.3 | 29.9 | 154.7 | 0.310 |
| 0.100 | 0.078 | 52.9 | 83.6 | 27.9 | 203.0 | 0.477 |
| 0.200 | 0.160 | 56.3 | 99.0 | 26.8 | 205.4 | 0.491 |
| 0.400 | 0.337 | 41.0 | 57.5 | 23.9 | 189.6 | 0.451 |
| 0.500 | 0.433 | 32.5 | 34.4 | 22.3 | 154.4 | 0.340 |
| 0.600 | 0.534 | 32.8 | 40.3 | 20.8 | 152.9 | 0.341 |
| 0.700 | 0.640 | 26.9 | 25.7 | 19.3 | 135.2 | 0.291 |
| 0.800 | 0.753 | 16.4 | −1.8 | 17.0 | 164.1 | 0.397 |
| 0.900 | 0.873 | 12.9 | −9.5 | 15.7 | 104.6 | 0.206 |
| 1.000 | 1.000 | 10.0 | −16.1 | 14.8 | 84.6 | 0.144 |

Note: $\Delta_{fa}H^{\ddagger}$, $\Delta_{fa}S^{\ddagger}$ and $\Delta_{fa}G^{\ddagger}$, respectively, are the enthalpy, entropy and Gibbs energy of activation of flow; $\Delta_{sf}H^{\circ}$ and $\Delta_{sf}S^{\circ}$, respectively, are the standard enthalpy and entropy of surface formation.

Table 5 shows all the thermodynamic quantities of flow activation and surface formation. With regard to flow properties it is interesting to note that enthalpy and entropy diminish with the increase of *N*-methyl-2-pyrrolidone proportion in the mixtures. Negative values of entropy of activation are found for mixtures from 0.753 in mole fraction of *N*-methyl-2-pyrrolidone to pure *N*-methyl-2-pyrrolidone. According to Corradini *et al.* [48] the enthalpy of viscous flow may be regarded as a measure of the cooperation degree among the species participating in this dynamic process. Thus, in a highly structured liquid as glycerol there would be a considerable degree of order, and therefore for the cooperative movement of particles a large enthalpy of activation together with a relatively high value of entropy of activation are required for the flow process. On the other hand, both thermodynamic quantities of surface formation are positive and present two maximal values, i.e. at 0.160 and 0.753 in mole fraction of *N*-methyl-2-pyrrolidone. According to Giner *et al.* [51] symmetrical molecules only show one maximum of entropy, whereas, non-symmetrical ones, due to the different conformations that they can adopt while their surface concentration increases, may show several maximum values of this property, just as happens with glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures.

In conclusion, the measured and derived quantities could be used in the computations of the process designs employing glycerol (1) + *N*-methyl-2-pyrrolidone (2) mixtures as solvent systems.

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