

# The density, dynamic viscosity and kinematic viscosity of protic and aprotic polar solvents (pure and mixed) systems: An experimental and theoretical insight of thermophysical properties

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## ABSTRACT:

We report herein the thermophysical properties of pure protic and aprotic polar solvents (water, dimethyl sulfoxide and *N,N*-dimethyl formamide) and their mixed (binary and ternary) systems. The experimental density ( $\rho$ ) and viscosity ( $\eta$ ) were determined for these systems by varying temperature range from 293.15 K to 343.15 K. The excess properties (*viz.*, excess molar volumes ( $V^E$ ), excess thermal expansion coefficient ( $\alpha_E$ ), viscosity deviation ( $\Delta\eta$ ) and excess Gibb's free energy ( $\Delta G^E$  for viscous flow) were computed for mixed systems from experimental values. The excess properties illustrate non ideal behavior of the mixed systems. Especially, a mixture of water with aprotic polar component show better thermophysical properties than pure systems. The pure DMF system shows increasing dynamic viscosity values at higher temperature region. Both binary as well as ternary mixed systems shows maximum activation energy ( $E_a$ ) in the presence of water depicts its thermal stability of the mixtures. The thermal expansion coefficient ( $\alpha$ ) also revealed increase in thermal behaviour for protic-aprotic systems. In addition to that, thermodynamic parameters are also evaluated for better understanding of thermal stability.

**Keywords:** Thermophysical properties, Density, Dynamic Viscosity, Excess properties, Arrhenius equation and Thermodynamics.

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## 1. INTRODUCTION

In experimental as well as theoretical point of view to understand the molecular interaction and molecular packing of the mixed systems, density and viscosity are the significant physical properties [1-5]. Particularly, viscosity of fluids and their mixtures provide as quantitative data to construct chemical engineering process including mass transfer, heat transfer and fluid flow mechanism [6, 7]. The unique thermophysical characteristics of the mixed system of various composition ranges find wide application in industrial processes [8-10]. Generally, transport properties of solvents mixtures with different temperature range are well known and reported [11-13]. However, the better comparison of thermophysical properties for protic and aprotic polar solvent mixture systems is insufficient which need to be addressed for indepth knowledge [14-18].

The mixture of protic and aprotic polar solvents strongly attracts to industrial application as cosolvents in ionic liquids, dissolution, polymerisation medium, etc., because of its unmatched benefits [19-24]. Particularly, dimethyl sulfoxide (DMSO) and *N,N*-dimethyl formamide (DMF) are aprotic polar solvents where it has many industrial and research application. The molecular aggregates formed by the interaction between mixed systems are believed to be responsible for the variation in thermophysical properties with the mole fraction adjustment [25, 26]. In case of DMSO and water, undergo self-associates in pure form, and by mixing the association is susceptible to rupture by the action of proton-acceptor solvents [27, 28].

As a result, unlike molecules interact upon mixing. The interaction between unlike varies from strong to weak based on its chemical characteristics (*viz.*, hydrogen bond, charge transfer, dispersion force, etc). Sometimes, molecular clusters or aggregates also form by the

interaction between unlike molecules are believed to have an ordered geometry and to be responsible for the strongly non ideal behaviour on mixing [29].

The objective of this research is to find the variation in thermophysical and thermodynamic properties of mixed systems. Particularly, the physical behaviour of protic-aprotic and aprotic-aprotic mixed system on various temperature conditions can provide fundamental understanding. This give better understanding of solvent mixture to formulate any dispersion system or chemical processing for various application [30].

In our previous study, we have studied thermophysical properties of protic polar solvents and their mixtures [25]. In the present study, we have investigated thermodynamic and thermophysical properties of pure and mixed system of protic (water) and aprotic polar (dimethyl sulfoxide (DMSO) and *N,N*-dimethyl formamide (DMF)) systems. The experimental density, dynamic viscosity and kinematic viscosity were measured for the pure systems: water, DMSO and DMF, binary systems: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) and ternary system: water (1) + DMSO (2) + DMF (3) at the temperature range from 293.15 K to 343.15 K in interval of 5 K. The obtained experimental data is used to evaluate excess properties [31-33]: excess molar volumes, thermal expansion coefficient, excess thermal expansion coefficient, viscosity deviation and excess Gibb's free energy [34] for activation of viscous flow for the mixed systems. The excess properties of binary systems were correlated with Redlich-Kister type polynomial equation [35, 36] . To understand the single, binary and ternary systems better way, we find out the thermodynamic properties of mixed systems, dynamic viscosity values is computed with Eyring transition state equation [37, 38]. The temperature dependent behavior of viscosities were also correlated with modified Arrhenius equation for Newtonian classic solvents [39, 40].

## 2. MATERIALS AND METHODS.

**2.1. Materials.** Dimethyl sulfoxide (CAS No. 67-68-5) of purity 99.9% and *N,N*-dimethyl formamide (CAS No. 68-12-2) of the purity 99.8% were purchased from Merck Specialities Private Limited, India and water were used for experiment from Milli-Q water which is double distilled and deionised (specific conductivity 1–2  $\mu\text{S cm}^{-1}$  at 298.15 K). The measured values of pure systems were compared with the literature values [5, 41-45] (see Table 1) and deviation of experimental values with the literature values have standard error in the order of  $10^{-3}$  and this error can be due to impurity in the chemicals and uncertainty in the experiment.

**2.2. Density Measurements.** Densities ( $\rho$ ) of protic and aprotic polar solvents were measured by using the U-tube digital density meter (Laboratory Density Meter (model no.: DMA 5000 M), Anton Paar GmbH) with a repeatability of  $1 \times 10^{-5} \text{ g/cm}^3$  and 0.01 K, at atmospheric pressure. For all (the densities of pure water, DMF and DMSO and their binary as well as ternary mixture) systems, the measurements were investigated at the temperature range of 293.15 to 343.15 K with a temperature interval of 5 K at constant atmospheric pressure. The measuring range of temperature (from 293.15 to 343.15 K) was controlled by a built-in Peltier thermostat (PT-100) with an accuracy of  $\pm 0.01$  K. A standard deviation of  $1.4 \times 10^{-6} \text{ g/cm}^3$  was used to measure the density.

**2.3. Viscosity Measurements.** The viscosity of pure, binary and ternary liquid mixture was measured at a temperature range from 293.15 to 343.15 K with a temperature interval of 5 K by using the Microviscometer (model no.: LOVIS 2000 ME) Microviscometer Module, Anton Paar GmbH). The measuring temperature (from 293.15 to 343.15 K) was controlled by a built-in Peltier thermostat with an accuracy of  $\pm 0.01$  K. It can measure the viscosity in the range of 0.3 mPa·s to 10000 mPa·s and the viscosity accuracy is 0.5%. The nominal uncertainty of the measured was estimated to be better than  $\pm 0.5$  % and reproducibility 0.1 %. The quantity of liquid required (approximately 1 mL) was transferred to the apparatus by a syringe; special

care was taken not to introduce bubbles in the tube to prevent errors in the measurements. In this range of viscosity values the diameter of the capillary was 1.59 mm, and the error for this capillary was < 0.01 %.

### 3. RESULT AND DISCUSSION

**3.1. Pure systems.** Table 1 represents thermophysical properties of pure components (water, DMSO & DMF). For all pure systems, the values of density decrease with increase in temperature. Comparatively, the water system has intermediate density values than the aprotic systems. It can be seen that DMSO has higher density values possibly due to the presence of balanced hydrophobic and hydrophilic part which may play a major role in compact molecular packing [46, 47]. In spite of structural integrity, DMF molecular system shows less density values due to deficit molecular interaction for compact packing (see Supporting Information (SI) Fig. S1 (A)) [48]. The steepest decrease of density values was observed for DMSO and DMF with respect to increase in temperature compared to water. Thermal expansion coefficient for pure systems was also calculated using equation (3). The values are greater for DMSO and DMF than water and predicted that of the water has stronger intermolecular interaction on varying thermal stress than other pure system. As increase of temperature, thermal expansion coefficient decreases for all pure systems as given in Table 1.

The kinematic viscosity ( $\nu$ ) of the systems was calculated using equation (1):

$$\nu = \frac{\eta}{\rho} \quad (1)$$

where  $\eta$  is dynamic viscosity and  $\rho$  is density of the fluid systems.

The dynamic and kinematic viscosity values decrease for pure system (water and DMSO) as increase of temperature. However, for DMF system viscosity values decrease up to 328.15 K and above that increasing viscosity values were observed. This result contradicts reported data of AK Nain et al where there is no increase in viscosity values as increase of temperature [42]. It can be predicted that influence of molecular mechanism of DMF to the rheological behaviour

is so different at lower and higher temperature condition. The increasing of viscosity for DMF system above a certain temperature (323.15 K) can be provided molecular insight of DMF for better understanding of viscosity relation to the molecular interaction on varying thermal stress. In case of water and DMSO, the trends were observed concave for temperature dependence of viscosity by the reflection of molecular events (see SI Fig. S1 (B and C)).

### 3.2. Binary systems.

**Density.** Figure 1 shows density values of all binary molecular mixtures. Density values were logarithmically decreasing as mole fraction of water increasing for water (1) + DMSO (2) as seen in Fig. 1(A). Figure 1(B) represents linear decrease of density values as increase of temperature and the mixed system (mole fraction 0.5 and 0.75 of water) has intermediate density values of pure component. In contradiction to that, mixture with mole fraction 0.25 of water shows almost similar values at low temperature and at higher temperature, values increase further. It can be predicted that DMSO with low mole fraction of water form compact molecular packing possibly by involving hydrogen bonding between water-water and water-DMSO. In the case of water (1) + DMF (2), logarithmic increase of density values as mole fraction of water increase shown in Fig. 1(C). It can be observed that linear decrease of values as function of temperature for pure systems as well as 0.75 mole fraction of water with DMF. At certain mole fraction (0.25 and 0.5 water) of water (1) + DMF (2) shows molecular aggregation characteristic which can be observed by deviation in linearity of density values as function of temperature as seen in Fig. 1 (D). Density values for the system (DMSO (1) + DMF (2)) are linearly increasing as increase of mole fraction of DMSO (Fig. 1 (E & F)). Linearity in the values illustrates ideal behaviour of aprotic-aprotic mixed system where original molecular packing is not much disrupted by unlike molecule. Figure 2 depicts the density values for all the binary mixture at 298.15 K. As mole fraction of water increases, logarithmic increase and decrease for water (1) + DMSO (2) and water (1) + DMF (2) were observed. On other hand, a

linear deviation was observed for the system DMSO (1) + DMF (2). By increasing temperature, density values decrease for all binary mixtures and there is no significant change in trend by varying the mole fraction (see SI, Fig. S2 (A to J)).

The excess volumetric properties for binary mixtures are calculated using experimental density values which is given by

$$V^E = \frac{\sum_{i=1}^n x_i M_i}{\rho} - \sum_{i=1}^n \frac{x_i M_i}{\rho_i} \quad (2)$$

where  $x_i$  is mole fraction,  $M_i$  is molar mass of pure component,  $\rho_i$  is the density of pure component,  $\rho$  is the density of mixture respectively. The excess molar volumes of all binary mixture at 298.15 K are given in Fig. 3. The molecular mixed system of water and DMSO has sigmoidal type deviation. Negative deviation is observed up to 0.5 mole fraction of water and beyond that positive deviation is predicted. It can be clearly understand that the presence of water molecules at lower mole fraction influence the hydration of DMSO via multiple hydrogen bonding. In case of water (1) + DMF (2) negative deviation was observed for throughout the molecular mixture which indicates stronger interaction between unlike molecules like intermolecular dipolar interaction, geometrical fitting. The mixed system of aprotic-aprotic shows positive deviation up to mole fraction of DMSO (0.5) with DMF and above that ideal behaviour was observed. Excess molar volumes of binary mixtures show similar trends as function of mole fraction at different temperature (see SI, Fig. S3 (A to J)).

The thermal expansion coefficient of binary systems were calculated based on temperature dependence of density using equation (3)

$$\alpha = \frac{-1}{\rho} \left( \frac{\partial \rho}{\partial T} \right)_P \quad (3)$$

$$\alpha_E = \alpha_m - \sum_{i=1}^n \phi_i \alpha_i \quad (4)$$

where  $\phi_i = \sum_{j=1}^n \phi_j$  and  $\phi_j = \frac{x_j V_j}{\sum_{i=1}^n x_i V_i}$ ;  $\alpha_m$  is coefficient of thermal expansion of mixture,  $\alpha_i$  coefficient of thermal expansion of pure component and  $\phi_i$  is the volume fraction of the mixed system, respectively. The calculated excess thermal expansion coefficients by using equation (3) were recorded in Table 2 and 3. Figure 4 shows the excess thermal expansion coefficient for all binary mixed systems at 298.15 K. The values were found negative for water (1) + DMSO and water (1) + DMF (2) indicates presence of water in aprotic molecular systems enhance the intermolecular interaction and extend thermal stability. By the observation, mixture of water and DMF shows stronger interaction over molecular mixture of water with DMSO. The mixture DMSO (1) + DMF (2) shows almost no much deviation, have values nearly at ideal axis indicates weaker interaction between unlike molecules. Excess thermal expansion coefficient of binary mixed systems at different temperature follows similar trends (see SI, Fig. S4 (A to J)).

**Viscosity.** Figures 5 to 8 show dynamic and kinematic viscosities data of all binary mixed systems. Figure 5(A & B) represents dynamic viscosity for water (1) + DMSO (2). The distribution in values predict that dynamic viscosity of mixed system is higher than pure component (DMSO). It can be clearly seen that viscosity values of mixed system decrease as mole fraction of water increase which possibly due to disruption of hydrophobic interaction between DMSO-DMSO. Decreasing values of dynamic viscosity as increase of temperature is not observed for all the cases. At mole fraction 0.75 of water shows increase in viscosity values at 333.15 K and further decrease on higher temperature possibly due to hydration shell formation over DMSO. A molecular mixture, water (1) + DMF (2) has viscosity values higher than pure components as shown in Fig. 5(C). Figure 5(D) shows that on elevated temperature at 333.15 K viscosity increases for pure molecular system (DMF). As increase of temperature, viscosity values for mixed system (water (1) + DMF (2)) of water mole fraction (0.25 and 0.50) behaves almost similar. In the case of mixture, (DMSO (1) + DMF (2)) has relatively linear



deviation of dynamic viscosity at lower temperature and non linear deviation at higher temperature as represented in Fig. 5(E). The reason can be the formation of aggregates by unlike molecules via stronger intermolecular interaction like charge transfer. Predominantly, the viscosity values of molecular mixture lie between the pure system. For the mixture DMSO (0.25) and DMF (0.75), by increasing temperature above 313.15 K slant increase in values were observed and sharp increase over 333.15 K exhibit in Fig. 5(F). Figure 6 shows dynamic viscosities for all binary system at 298.15 K. Parabolic trend were observed at decreasing y-axis for water (1) + DMSO (2) and water (1) + DMF (2) and in the case of DMSO (1) + DMF (2) concave trend was observed. As increase of temperature, dynamic viscosity values decrease for binary mixed systems as dependence of mole fraction. The mixture of DMSO (1) + DMF (2) shows deviation in ideal behaviour at higher temperature (see SI, Fig. S5 (A to J)).

Figure 7(A to F) represents the kinematic viscosity of binary mixed systems. Kinematic viscosity values decrease as increase of temperature for the system water (1) + DMSO (2) shown in Fig. 7(A and B). But, for the mixtures with presence of DMF show increase in viscosity values at higher temperature represented in Fig. 7(C to F). Figure 8 shows the kinematic viscosity of all mixed system at 298.13 K. Deviation follows as similar as dynamic viscosity values. Kinematic viscosity decreases for binary mixed system as increase of temperature except that DMSO (1) + DMF (2) show increase in kinematic viscosity at higher temperature (see SI, Fig. S6 (A to J)).

The viscosity deviation for mixed system can be calculated using equation (5):

$$\Delta\eta = \eta - \sum_{i=1}^n x_i\eta_i \quad (5)$$

where  $\eta$  is the dynamic viscosity of mixture,  $x_i$  is the mole fraction and  $\eta_i$  is the viscosity of pure components. Figure 9 represents viscosity deviation of all binary mixed systems at 298.15 K. The mixture of water (1) + DMSO (2) and water (1) + DMF (2) has positive viscosity deviation for throughout composition range. Whereas, mixed system of DMSO (1) + DMF (2)

has negative deviation nearly at ideal axis which illustrates that it could be weaker interaction between unlike molecules such as hydrogen bond rupture and dispersion force. The maximum viscosity deviation is observed for the mixture of water (1) + DMSO (2), indicates stronger intermolecular interaction between unlike molecules like hydrogen bonding, charge transfer, etc., The data illustrates that protic-aprotic mixture has a stronger molecular interaction, whereas aprotic-aprotic mixture possesses weaker interaction. The rheological property of mixed system also behaves differently based on the combination of protic and aprotic molecular mixture. The protic-aprotic polar molecular system shows increased viscosity values by involving stronger intermolecular interaction. In the case of aprotic-aprotic system change in viscosity values is not significant indicating ideal behaviour. Viscosity deviation of binary mixed systems decrease as increase of temperature and particularly for the mixture DMSO (1) + DMF (2) trends shifted from ideal axis to sigmoidal deviation at higher temperature region (see SI, Fig. S7 (A to J)).

The excess Gibb's free energy for activation of viscous flow for mixed systems were calculated from:

$$\Delta G^E = RT \left[ \ln \eta V - \sum_{i=1}^n \eta_i V_i \right] \quad (6)$$

where  $\eta$ ,  $\eta_i$ ,  $V$ ,  $V_i$ ,  $R$  and  $T$  are viscosity of mixture and pure components, molar volume of mixture and pure components, gas constant and absolute temperature, respectively. The excess Gibb's free energy for activation of viscous flow for the binary mixed systems represented in Fig. 10. The positive values of excess Gibb's free energy were observed for binary system; water (1) + DMSO (2) and water (1) + DMF (2). The mixture of DMSO and DMF shows negative excess Gibb's free energy at nearly ideal axis indicates ideal behaviour of the mixture. The theoretical investigation represent protic-aprotic molecular mixture have more thermodynamica stability than aprotic-aprotic molecular mixture. In which, water-DMF shows extended stability compared to other two binary mixtures. Excess Gibb's free energy for

DMSO (1) + DMF (2) deviated from ideal axis at higher temperature and show sigmoidal deviations (see SI, Fig. S8 (A to J)).

The calculated excess properties (*viz.*, excess molar volumes, excess thermal expansion coefficient, viscosity deviation and excess Gibb's free energy) of binary mixed system were fitted using Redlich-Kister type polynomial equation and represented in Figs. 3, 4, 9 & 10 as dotted lines.

$$Y^E = x_1 x_2 \sum_{j=1}^n A_j (x_1 - x_2)^j \quad (7)$$

where  $x_1$  and  $x_2$  are mole fraction of components,  $A_j$  is interaction parameter and  $Y^E$  is excess properties. The interaction parameters were evaluated by fitting equation using nonlinear least square regression method. The evaluated fitted parameters were recorded in Table 4. To find the standard deviation of experimental values from fitted values equation (7) were used:

$$\sigma(Y) = \left[ \frac{\sum (Y_{expt} - Y_{calc})^2}{N - n} \right]^{\frac{1}{2}} \quad (8)$$

where  $N$  is the number of data and  $n$  is the number of interaction parameters.

### 3.3. Ternary systems.

Figure 11 represents the excess thermophysical properties of the ternary molecular mixed system. Excess molar volumes of ternary system, water (1) + DMSO (2) + DMF (3) were determined and shown in Fig. 11(A). The least negative values were observed at water and DMF molecular rich region and intermediate values were observed at equimolar region of water (1) + DMSO (2) + DMF (3). Particularly at water and DMSO rich region, positive values were found which indicates that domination of weaker interaction. The data illustrates that the interaction of water with DMF is stronger than DMSO. Figure 11(B) represents excess thermal expansion coefficient of ternary mixed system. The colour plot shows that the values are larger negative for water and DMF rich region, indicates the stronger interaction between unlike molecules (protic-aprotic). The rich regions of aprotic-aprotic polar solvent system have higher

excess thermal expansion coefficient values show lesser thermal stability in that region of ternary system. The intermediate values were found at equimolar mixture.

The viscosity deviation of ternary mixed system represented in Fig. 11(C). The maximum positive values were observed for water and DMSO rich region, illustrates the compact geometrical fitting of unlike molecules. Least values were found for DMSO and DMF rich region of the system indicates weaker interaction of aprotic-aprotic region and all other regions have intermediate values. Figure 11(D) shows the excess Gibb's free energy for activation of viscous flow for mixed system. The stable regions were found for DMSO and DMF with water rich region by observing the maximum values. Whereas, DMSO and DMF rich region has least values shows thermally fewer stable.

### 3.4. Theoretical insight of temperature dependence of viscosity.

The modified equation based on Eyring transition state theory was taken to correlate the temperature dependence of viscosity for mixed systems. The thermodynamic parameters: change of enthalpy, entropy and Gibb's free energy for activation of viscous flow for pure and their mixtures were also evaluated using equation (10):

$$\eta = \frac{hN_A}{V} \exp\left(\frac{\Delta G^*}{RT}\right)_P \quad (9)$$

where

$$\left(\frac{\Delta G^*}{RT}\right)_P = \ln\left(\frac{\eta V}{hN_A}\right) \quad (10)$$

and  $\eta$  is the experimental dynamic viscosity,  $V$  is the molar volume,  $h$  is the Planck's constant and  $N_A$  is the Avogadro number. A straight line was observed by plotting  $\ln(\eta V/hN_A)$  as dependence of  $1/T$ .

$$\Delta G^* = \Delta H^* - T\Delta S^* \quad (11)$$

$$\ln\left(\frac{\eta V}{hN_A}\right) = \frac{\Delta H^*}{RT} - \frac{\Delta S^*}{R} = \frac{\Delta G^*}{RT} \quad (12)$$

From the plotted graphs, the enthalpy change and entropy change for viscous flow were found as slope and intercept. Table 5 and 6 show evaluated values for molecular mixture of protic and aprotic polar solvent. Gibb's free energy for viscous flow were calculated for studied temperature based on contributions of enthalpy and entropy changes represented in Table 1 to 3. The free energy change for viscous flow values increase as increase of temperature for all mixed systems.

To understand the thermal stability of mixed system, Arrhenius type equation for Newtonian classic solvents is the best one to correlate the temperature dependence of dynamic viscosity by modifying to logarithmic form:

$$\ln(\eta) = \ln(A_s) + \frac{E_a}{R} [1/T] \quad (13)$$

$$T^* = \frac{E_a}{R} \quad (14)$$

where  $A_s$  is pre-exponential entropic factor related to viscosity at infinite temperature,  $E_a$  is activation energy for transition state,  $R$  is gas constant and  $T$  is absolute temperature. The linear Arrhenius behavior were observed by fitting to the Andrade equation. Using linear least square regression method, the equation is fitted with data where slope and intercept found as  $E_a/R$  and  $\ln(A_s)$ . In addition, activation temperature ( $T^*$ ) were determined from linear Arrhenius equation.

$$T_A = \frac{-E_a}{R \ln(A_s)} \quad (15)$$

The Arrhenius temperature ( $T_A$ ) were ascertained from the intercept with abscissa axis [49] to modify the equation of the form:

$$\ln(\eta) = \frac{E_a}{RT} \left( \frac{1}{T} - \frac{1}{T_A} \right) \quad (16)$$

where pure and mixture systems have linear Arrhenius behavior having activation energy for viscous flow of range ( $2 < E_a < 20 \text{ kJ.mol}^{-1}$ ) and preexponential factor of range ( $-7 < \ln(A_s) <$

-1 mPa.s) reported in Table 5 and 6. The binary mixed systems have the activation energy in the order of (water (1) + DMSO (2)) > (water (1) + DMF (2)) > (DMSO (1) + DMF (2)). The presence of water in the aprotic mixed system increase the activation energy and make system more stable. By observing the influence of temperature on physical properties of pure and molecular mixture of protic and aprotic polar solvents, useful knowledge on the molecular interaction and organisation have been predicted. Moreover, the theoretical attempts to predict the critical parameters through simple experimental methods have been carried out. To the best of our knowledge, there is no key literature available to support the significant variation observed in studied physical properties of pure as well as molecular mixtures. So, the present investigation will provide good amount of information for future work.

#### **4. CONCLUSIONS**

The pure and their mixing behaviour of protic (water) and aprotic (dimethyl sulfoxide and *N,N*-dimethyl formamide) polar solvents have been studied for water, DMSO and DMF. Density, dynamic viscosity and kinematic viscosity have been experimentally measured. Density as well as viscosity decrease as increase of temperature except pure DMF and mixture with DMF where values increase above certain temperature. Excess molar volumes, thermal expansion coefficient, viscosity deviation, excess Gibb's free energy were evaluated from experimental density and viscosity values. The Redlich-Kister type polynomial equation was fitted to excess properties through least square regression method. Viscosity deviation is positive for mixture of water with aprotic polar systems which indicates strong interaction between unlike molecules. The mixtures of aprotic-aprotic have almost behaved ideal observed by deviation of viscosity at ideal axis. By perceiving of all experimental and calculated data, water with aprotic system have very stronger interaction than aprotic-aprotic mixture which have ideal interaction. The temperature dependence of viscosities were studied by using mathematical expression of Eyring state transition equation and Arrhenius type equation for Newtonian classic solvents. The present investigation has provided an insight in to the molecular mixture of binary and ternary system with protic and aprotic polar molecules. The variation in excess properties of molecular mixture form a basis to understand the molecular process of mixing and it influence in physical behaviour change on different thermal stress.

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## Figure Captions:

**Fig. 1.** Density ( $\rho$ ) of binary mixed systems for water (1) + DMSO (2) as a function of mole fraction (A) and temperature (B), water (1) + DMF (2) as a function of mole fraction (C) and temperature (D), DMSO (1) + DMF (2) as a function of mole fraction (E) and temperature (F).

**Fig. 2.** Density ( $\rho$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 3.** Excess molar volume ( $V^E$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 4.** Excess thermal expansion coefficient ( $\alpha_E$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 5.** Dynamic viscosity ( $\eta$ ) of binary mixed systems for water (1) + DMSO (2) as a function of mole fraction (A) and temperature (B), water (1) + DMF (2) as a function of mole fraction (C) and temperature (D), DMSO (1) + DMF (2) as a function of mole fraction (E) and temperature (F).

**Fig. 6.** Dynamic viscosity ( $\eta$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 7.** Kinematic viscosity ( $\nu$ ) of binary mixed systems for water (1) + DMSO (2) as a function of mole fraction (A) and temperature (B), water (1) + DMF (2) as a function of mole fraction (C) and temperature (D), DMSO (1) + DMF (2) as a function of mole fraction (E) and temperature (F).

**Fig. 8.** Kinematic viscosity ( $\nu$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

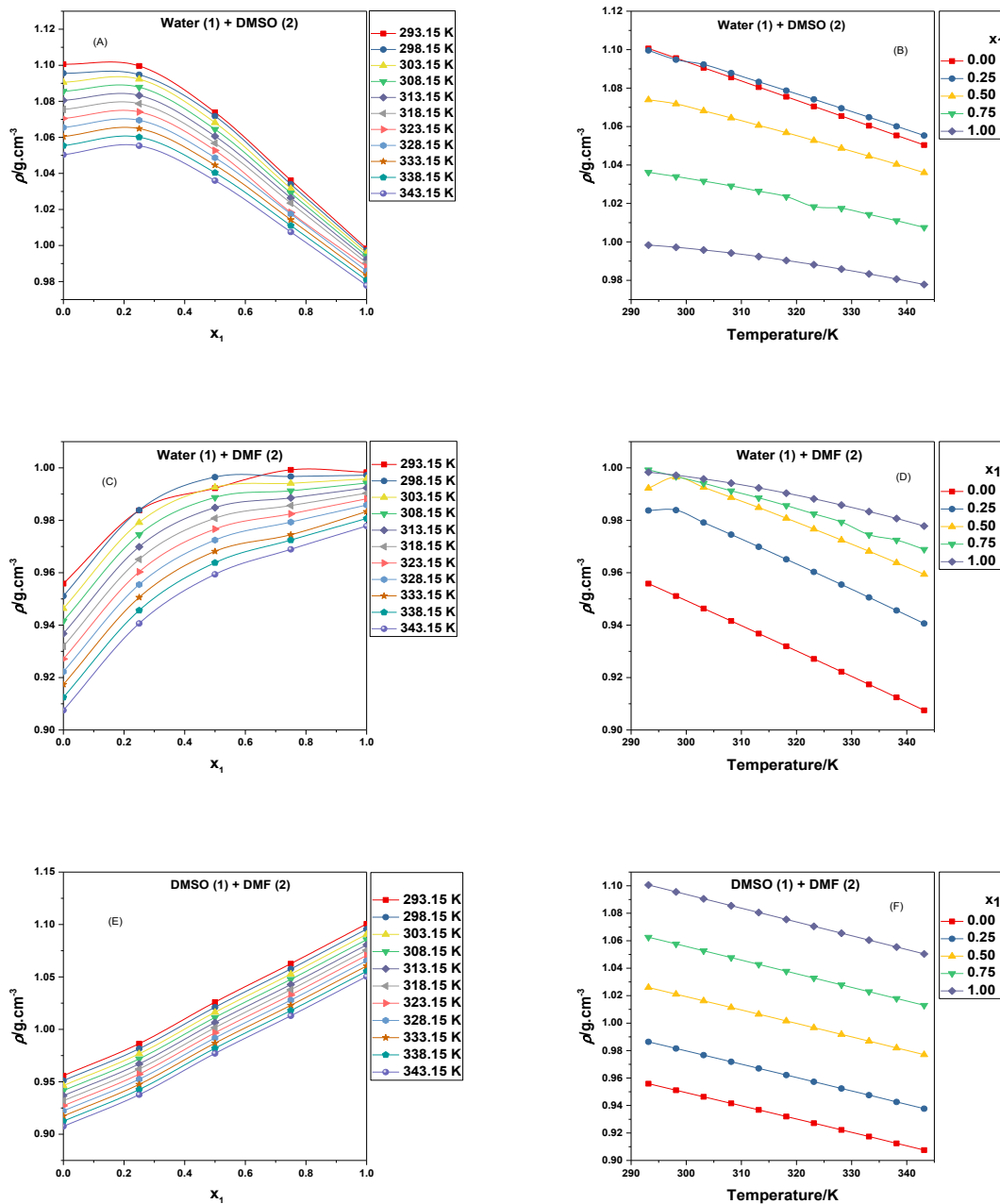
**Fig. 9.** Viscosity deviation ( $\Delta\eta$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 10.** Excess Gibb's free energy for activation of viscous flow ( $\Delta G^E$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

**Fig. 11.** Excess molar volumes ( $V^E$ ) (A), excess thermal expansion coefficient ( $\alpha_E$ ) (B), viscosity deviation ( $\Delta\eta$ ) (C) and excess Gibb's free energy ( $\Delta G^{*E}$ ) for activation of viscous flow (D) of ternary mixture systems for water (1) + DMSO (2) + DMF (3) as a function of mole fraction at 298.15 K.

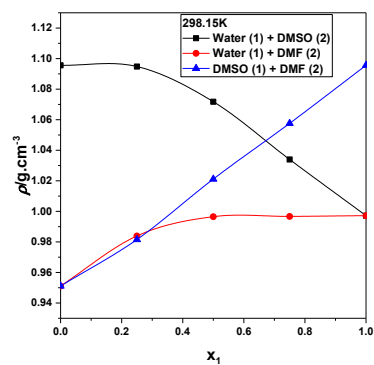


Figure(s)



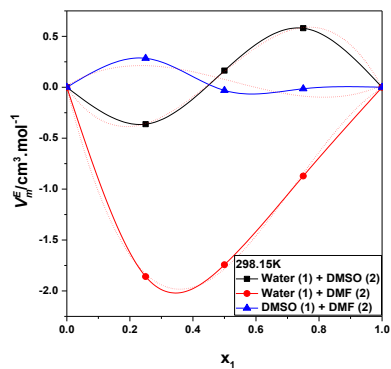
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Figure(s)



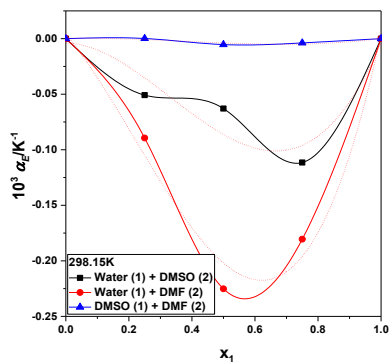
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Figure(s)



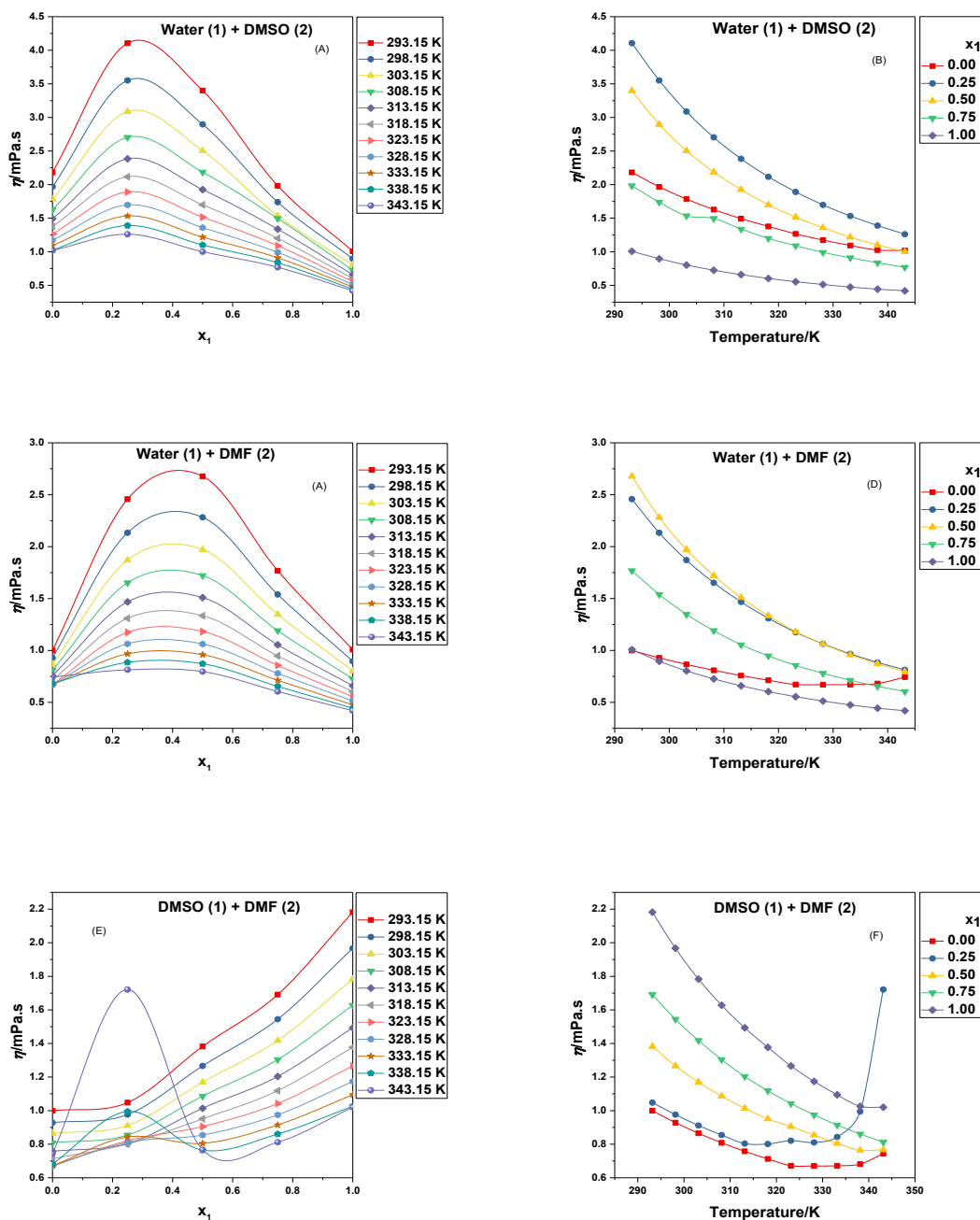
**Fig. 3.** Excess molar volume ( $V_m^E$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

Figure(s)



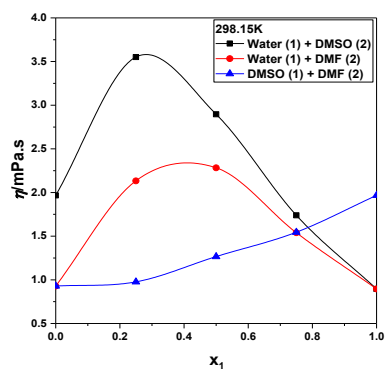
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Figure(s)



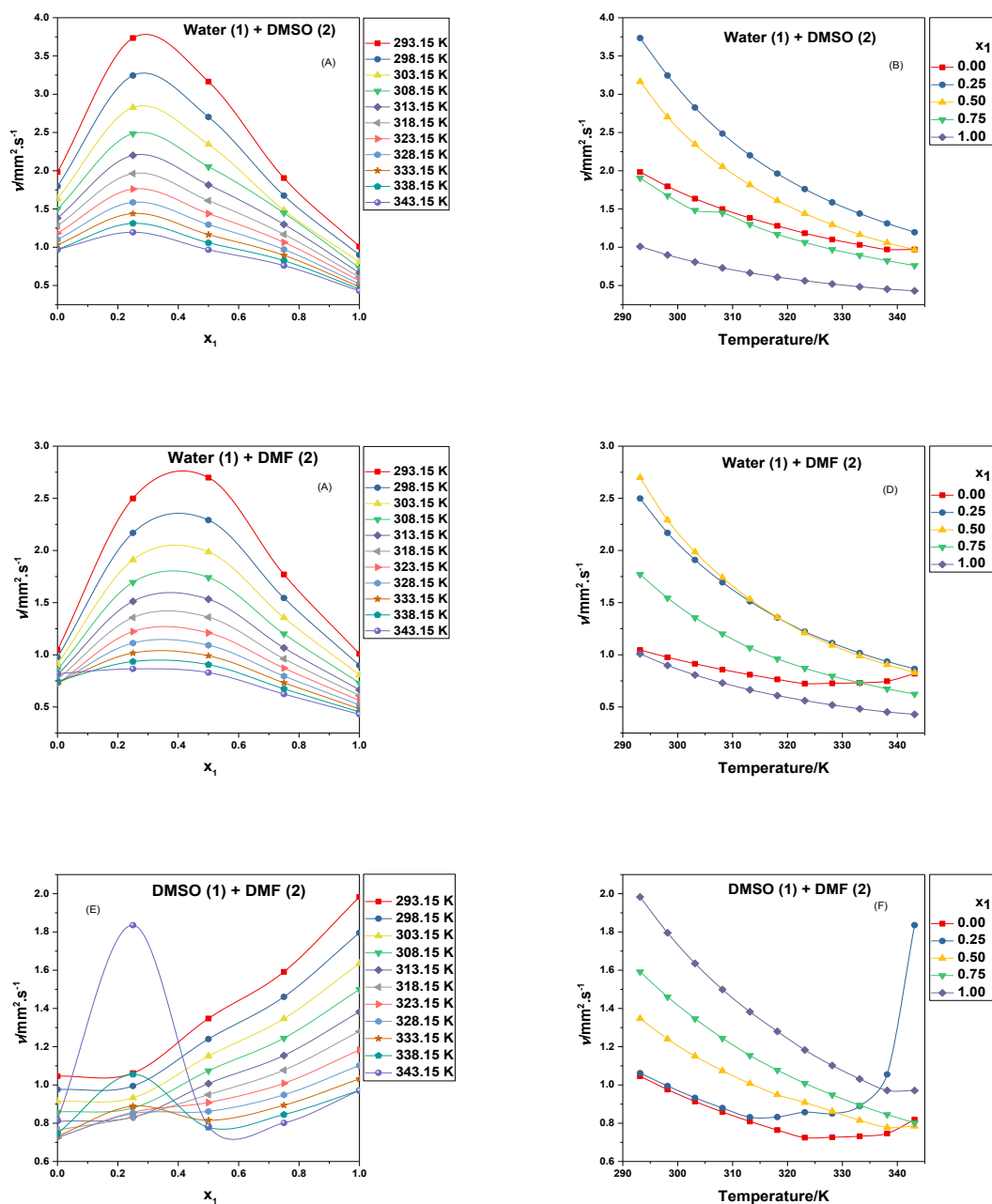
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Figure(s)



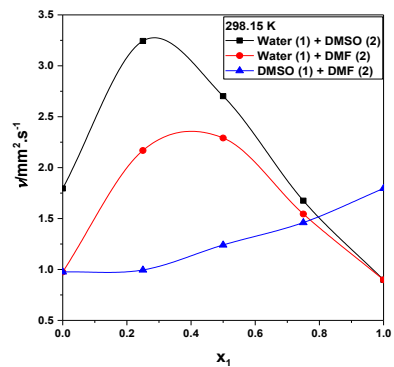
**Fig. 6.** Dynamic viscosity ( $\eta$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

Figure(s)



**Fig. 7.** Kinematic viscosity ( $\nu$ ) of binary mixed systems for water (1) + DMSO (2) as a function of mole fraction (A) and temperature (B), water (1) + DMF (2) as a function of mole fraction (C) and temperature (D), DMSO (1) + DMF (2) as a function of mole fraction (E) and temperature (F).

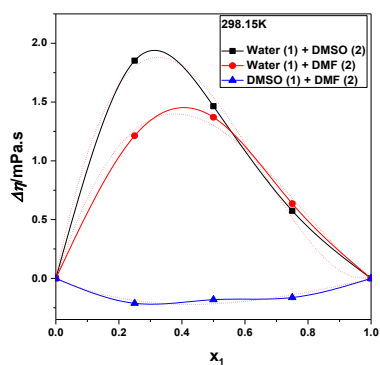
Figure(s)



**Fig. 8.** Kinematic viscosity ( $\nu$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

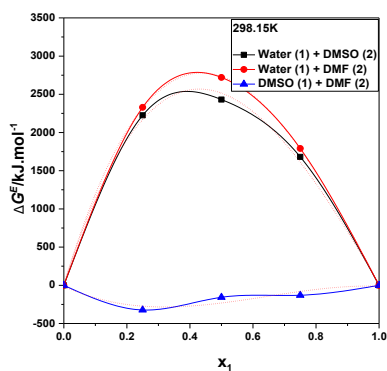


Figure(s)



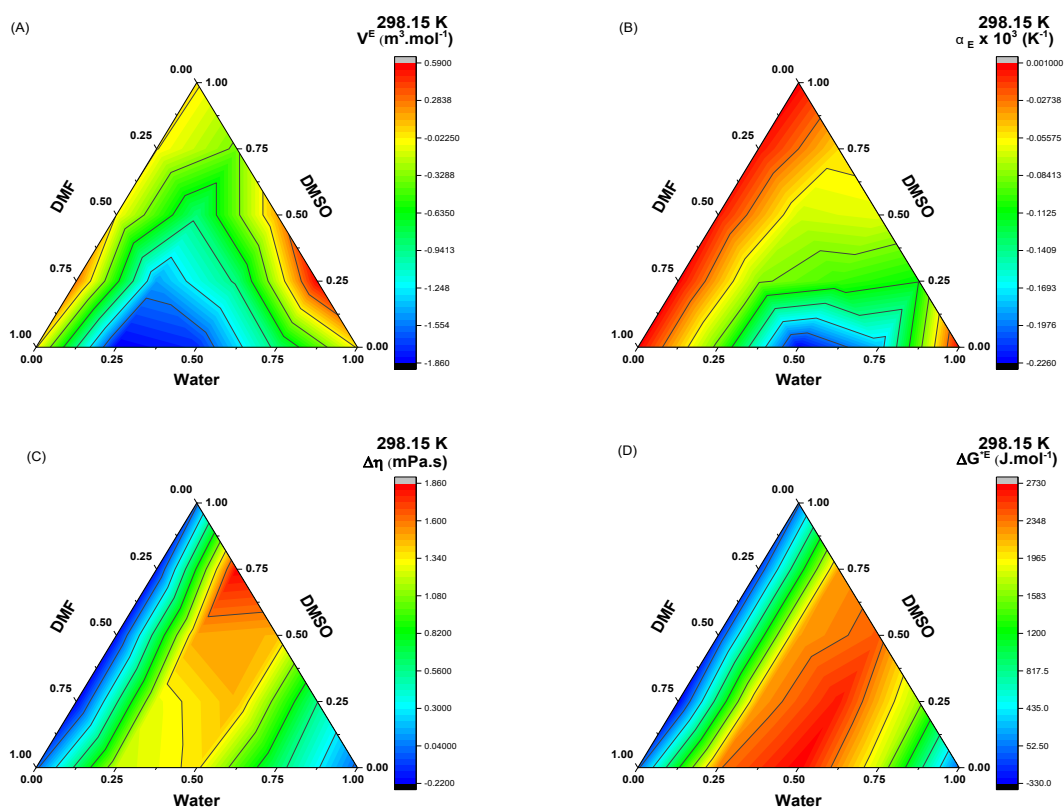
**Fig. 9.** Viscosity deviation ( $\Delta\eta$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

Figure(s)



**Fig. 10.** Excess Gibb's free energy for activation of viscous flow ( $\Delta G^E$ ) of binary mixed systems for water (1) + DMSO (2), water (1) + DMF (2), DMSO (1) + DMF (2) as a function of mole fraction at 298.15 K.

Figure(s)



**Fig. 11.** Excess molar volumes ( $V^E$ ) (A), excess thermal expansion coefficient ( $\alpha_E$ ) (B), viscosity deviation ( $\Delta\eta$ ) (C) and excess Gibb's free energy ( $\Delta G^{*E}$ ) for activation of viscous flow (D) of ternary mixture systems for water (1) + DMSO (2) + DMF (3) as a function of mole fraction at 298.15 K.

**Table 1.** Experimental Density ( $\rho$ ), Dynamic Viscosity ( $\eta$ ), Kinematic Viscosity ( $\nu$ ), Thermal Expansion Coefficient ( $\alpha$ ) and for Pure Water, DMSO and DMF

$T$ /K	$10^{-3}\rho$ /Kg.m <sup>-3</sup>		$\eta$ /mPa.s		$\nu$ /mm <sup>2</sup> .s <sup>-1</sup>	$10^3\alpha$ /K <sup>-1</sup>
	Exp	Lit	Exp	Lit		
			<u>Water</u>			
293.15	0.99833	1.00000 [45]	1.007	1.000 [45]	1.009	0.401
298.15	0.99718		0.896		0.899	0.401
303.15	0.99578	0.99800 [45]	0.803	0.805 [45]	0.806	0.402
308.15	0.99416		0.726		0.730	0.402
313.15	0.99234	0.99430 [45]	0.659	0.651 [45]	0.664	0.403
318.15	0.99034		0.603		0.609	0.404
323.15	0.98816	0.99020 [45]	0.555	0.544 [45]	0.561	0.405
328.15	0.98581		0.512		0.520	0.406
333.15	0.98331	0.98520 [45]	0.475	0.472 [45]	0.483	0.407
338.15	0.98066		0.443		0.452	0.408
343.15	0.97782	0.97960 [45]	0.419	0.417 [45]	0.429	0.409
			<u>DMSO</u>			
293.15	1.10059	1.1012 [5]	2.182	2.26 [5]	1.983	0.909
298.15	1.09557		1.967		1.796	0.913
303.15	1.09055	1.0912 [5]	1.783	1.85 [5]	1.635	0.917
308.15	1.08553		1.628		1.499	0.921
313.15	1.08051	1.0811 [5]	1.493	1.56 [5]	1.382	0.925
318.15	1.07549		1.377		1.280	0.930
323.15	1.07047	1.0711 [5]	1.266	1.33 [5]	1.183	0.934
328.15	1.06545		1.174		1.102	0.939
333.15	1.06042	1.0612 [5]	1.094	1.16 [5]	1.032	0.943
338.15	1.05538		1.025		0.972	0.948
343.15	1.05034	1.0513 [5]	1.020	1.02 [5]	0.971	0.952
			<u>DMF</u>			
293.15	0.95584	0.94910 [42]	0.999	0.8685 [42]	1.046	1.046
298.15	0.95110	0.94460 [42]	0.928	0.8123 [42]	0.976	1.051
303.15	0.94634	0.94010 [42]	0.865	0.7647 [42]	0.914	1.057
308.15	0.94156	0.93560 [42]	0.808	0.7189 [42]	0.859	1.062
313.15	0.93676	0.93110 [42]	0.758	0.6851 [42]	0.809	1.068
318.15	0.93194	0.92660 [42]	0.712	0.6642 [42]	0.764	1.073
323.15	0.92710		0.672		0.724	1.079
328.15	0.92224		0.670		0.727	1.084
333.15	0.91735		0.671		0.732	1.090
338.15	0.91244		0.681		0.747	1.096
343.15	0.90751		0.743		0.819	1.102

**Table 2.** Experimental Density ( $\rho$ ), Dynamic Viscosity ( $\eta$ ), Kinematic Viscosity ( $\nu$ ), Excess Molar Volume ( $V^E$ ), Viscosity Deviation ( $\Delta\eta$ ), Thermal Expansion Coefficient ( $\alpha$ ), Excess Thermal Expansion Coefficient ( $\alpha_E$ ), Gibb's Free Energy ( $\Delta G^*$ ) and Excess Gibb's Free Energy of Activation of Viscous Flow ( $\Delta G^E$ ) for Binary Mixture of Water (1) + DMSO (2), Water (1) + DMF (2) and DMSO (1) + DMF (2).

$x_1$	$T$	$10^{-3}\rho$	$\eta$	$\nu$	$10^6 V^E$	$\Delta\eta$	$10^3 \alpha$	$10^3 \alpha_E$	$\Delta G^*$	$\Delta G^E$
	/K	/Kg.m <sup>-3</sup>	/mPa.s	/mm <sup>2</sup> .s <sup>-1</sup>	/m <sup>3</sup> .mol <sup>-1</sup>	/mPa.s	/K <sup>-1</sup>	/K <sup>-1</sup>	/kJ.mol <sup>-1</sup>	/kJ.mol <sup>-1</sup>
<u>Water (1) + DMSO (2)</u>										
0.25	293.15	1.09958	4.105	3.734	-0.36639	2.217	0.818	-0.050	49.226	2.276
0.25	298.15	1.09476	3.551	3.244	-0.36318	1.852	0.822	-0.051	49.717	2.225
0.25	303.15	1.09229	3.087	2.826	-0.48534	1.548	0.824	-0.053	50.203	2.162
0.25	308.15	1.08781	2.703	2.485	-0.50283	1.301	0.827	-0.054	50.702	2.101
0.25	313.15	1.08324	2.384	2.201	-0.51741	1.099	0.831	-0.054	51.208	2.041
0.25	318.15	1.07870	2.117	1.962	-0.53445	0.933	0.834	-0.055	51.723	1.982
0.25	323.15	1.07412	1.890	1.760	-0.55050	0.802	0.838	-0.056	52.243	1.936
0.25	328.15	1.06949	1.697	1.587	-0.56531	0.689	0.842	-0.056	52.769	1.884
0.25	333.15	1.06482	1.533	1.439	-0.57888	0.593	0.845	-0.057	53.303	1.831
0.25	338.15	1.06010	1.390	1.311	-0.59132	0.510	0.849	-0.057	53.840	1.772
0.25	343.15	1.05534	1.262	1.195	-0.60218	0.392	0.853	-0.058	54.373	1.578
0.50	293.15	1.07391	3.399	3.165	0.24692	1.804	0.745	-0.061	48.160	2.511
0.50	298.15	1.07175	2.896	2.702	0.16379	1.464	0.746	-0.063	48.590	2.431
0.50	303.15	1.06815	2.504	2.344	0.13826	1.211	0.749	-0.064	49.047	2.370
0.50	308.15	1.06446	2.186	2.054	0.11387	1.009	0.752	-0.065	49.516	2.311
0.50	313.15	1.06060	1.925	1.815	0.09463	0.849	0.754	-0.066	49.999	2.258
0.50	318.15	1.05676	1.699	1.608	0.07227	0.709	0.757	-0.067	50.476	2.192
0.50	323.15	1.05271	1.515	1.439	0.05692	0.604	0.760	-0.068	50.971	2.146
0.50	328.15	1.04867	1.359	1.296	0.03885	0.516	0.763	-0.069	51.473	2.097
0.50	333.15	1.04457	1.217	1.165	0.02182	0.433	0.766	-0.070	51.964	2.030
0.50	338.15	1.04035	1.101	1.059	0.00764	0.367	0.769	-0.071	52.474	1.970
0.50	343.15	1.03601	1.001	0.967	-0.00274	0.282	0.772	-0.072	52.990	1.820
0.75	293.15	1.03617	1.982	1.905	0.60905	0.681	0.579	-0.110	46.057	1.693
0.75	298.15	1.03396	1.739	1.675	0.58039	0.575	0.580	-0.112	46.470	1.681
0.75	303.15	1.03160	1.535	1.483	0.55237	0.487	0.582	-0.113	46.976	1.665
0.75	308.15	1.02911	1.495	1.448	0.52508	0.544	0.583	-0.115	47.558	1.874
0.75	313.15	1.02638	1.337	1.299	0.50203	0.470	0.585	-0.116	48.083	1.860
0.75	318.15	1.02354	1.200	1.169	0.47916	0.404	0.586	-0.118	48.402	1.835
0.75	323.15	1.01830	1.090	1.064	0.52996	0.357	0.589	-0.118	48.982	1.836
0.75	328.15	1.01746	0.990	0.970	0.43825	0.313	0.590	-0.121	49.379	1.814
0.75	333.15	1.01425	0.910	0.895	0.41931	0.281	0.592	-0.122	50.486	1.816
0.75	338.15	1.01102	0.835	0.824	0.39826	0.247	0.593	-0.124	50.785	1.794
0.75	343.15	1.00750	0.769	0.761	0.38371	0.199	0.596	-0.125	51.058	1.709
<u>Water (1) + DMF (2)</u>										
0.25	293.15	0.98373	2.457	2.498	-1.55898	1.456	0.915	-0.084	48.096	2.434
0.25	298.15	0.98388	2.133	2.168	-1.85892	1.213	0.915	-0.089	48.566	2.329
0.25	303.15	0.97915	1.870	1.909	-1.86396	1.020	0.919	-0.090	49.060	2.242
0.25	308.15	0.97454	1.651	1.694	-1.87885	0.864	0.924	-0.091	49.563	2.160
0.25	313.15	0.96986	1.467	1.513	-1.89180	0.734	0.928	-0.092	50.072	2.079
0.25	318.15	0.96511	1.310	1.357	-1.90274	0.625	0.933	-0.092	50.583	1.998
0.25	323.15	0.96031	1.174	1.223	-1.91295	0.532	0.937	-0.093	51.098	1.914
0.25	328.15	0.95546	1.063	1.113	-1.92176	0.433	0.942	-0.094	51.632	1.738
0.25	333.15	0.95056	0.966	1.017	-1.92959	0.344	0.947	-0.095	52.168	1.556
0.25	338.15	0.94561	0.884	0.935	-1.93711	0.262	0.952	-0.095	52.715	1.352
0.25	343.15	0.94064	0.813	0.865	-1.94551	0.151	0.957	-0.096	53.273	0.998
0.50	293.15	0.99225	2.677	2.698	-1.34729	1.674	0.705	-0.217	47.640	2.844
0.50	298.15	0.99646	2.283	2.291	-1.74266	1.371	0.702	-0.225	48.047	2.720
0.50	303.15	0.99256	1.971	1.986	-1.76889	1.137	0.705	-0.227	48.493	2.628
0.50	308.15	0.98870	1.721	1.741	-1.80050	0.954	0.708	-0.229	48.955	2.544
0.50	313.15	0.98482	1.509	1.532	-1.83442	0.801	0.711	-0.231	49.417	2.456
0.50	318.15	0.98076	1.334	1.360	-1.86318	0.676	0.714	-0.233	49.891	2.373
0.50	323.15	0.97663	1.182	1.210	-1.89157	0.569	0.717	-0.235	50.362	2.282
0.50	328.15	0.97243	1.062	1.092	-1.91953	0.470	0.720	-0.237	50.859	2.141

0.50	333.15	0.96815	0.959	0.990	-1.94689	0.386	0.723	-0.239	51.364	1.999
0.50	338.15	0.96381	0.872	0.905	-1.97395	0.310	0.726	-0.241	51.881	1.844
0.50	343.15	0.95938	0.796	0.829	-2.00024	0.215	0.730	-0.243	52.400	1.573
0.75	293.15	0.99922	1.769	1.770	-0.84199	0.763	0.600	-0.178	45.735	1.837
0.75	298.15	0.99669	1.539	1.545	-0.87225	0.635	0.602	-0.180	46.178	1.791
0.75	303.15	0.99412	1.348	1.356	-0.90525	0.529	0.604	-0.183	46.623	1.740
0.75	308.15	0.99125	1.191	1.201	-0.93281	0.444	0.605	-0.185	47.082	1.691
0.75	313.15	0.98853	1.054	1.067	-0.96915	0.370	0.607	-0.187	47.537	1.634
0.75	318.15	0.98560	0.948	0.962	-1.00189	0.317	0.609	-0.190	48.022	1.598
0.75	323.15	0.98243	0.856	0.872	-1.03035	0.272	0.611	-0.192	48.512	1.561
0.75	328.15	0.97931	0.779	0.796	-1.06380	0.228	0.613	-0.194	49.014	1.494
0.75	333.15	0.97448	0.713	0.731	-1.04319	0.189	0.616	-0.195	49.527	1.433
0.75	338.15	0.97242	0.654	0.673	-1.11841	0.152	0.617	-0.198	50.037	1.349
0.75	343.15	0.96891	0.604	0.623	-1.14896	0.104	0.619	-0.201	50.559	1.201
DMSO (1) +DMF (2)										
0.25	293.15	0.98635	1.048	1.062	0.28172	-0.248	1.014	0.000	46.562	-0.343
0.25	298.15	0.98153	0.976	0.994	0.28479	-0.212	1.019	0.000	47.193	-0.323
0.25	303.15	0.97670	0.911	0.933	0.28754	-0.183	1.024	0.000	47.823	-0.307
0.25	308.15	0.97185	0.855	0.880	0.28977	-0.158	1.029	0.000	48.461	-0.288
0.25	313.15	0.96700	0.804	0.831	0.29159	-0.138	1.034	0.000	49.100	-0.271
0.25	318.15	0.96215	0.801	0.832	0.29265	-0.077	1.039	0.000	49.888	-0.111
0.25	323.15	0.95728	0.821	0.857	0.29365	0.001	1.045	0.000	50.752	0.122
0.25	328.15	0.95240	0.810	0.850	0.29402	0.014	1.050	0.000	51.515	0.144
0.25	333.15	0.94751	0.843	0.889	0.29353	0.066	1.055	0.000	52.423	0.297
0.25	338.15	0.94260	0.994	1.055	0.29232	0.227	1.061	0.000	53.690	0.771
0.25	343.15	0.93768	1.721	1.836	0.29098	0.909	1.066	0.000	56.065	2.134
0.50	293.15	1.02592	1.382	1.347	-0.02787	-0.209	0.975	-0.005	47.182	-0.157
0.50	298.15	1.02107	1.266	1.240	-0.03121	-0.181	0.979	-0.005	47.782	-0.157
0.50	303.15	1.01621	1.169	1.150	-0.03467	-0.155	0.984	-0.005	48.394	-0.149
0.50	308.15	1.01134	1.086	1.074	-0.03820	-0.132	0.989	-0.006	49.016	-0.136
0.50	313.15	1.00646	1.014	1.007	-0.04164	-0.112	0.994	-0.006	49.644	-0.122
0.50	318.15	1.00158	0.951	0.949	-0.04583	-0.094	0.998	-0.006	50.280	-0.105
0.50	323.15	0.99668	0.905	0.908	-0.04989	-0.064	1.003	-0.006	50.951	-0.049
0.50	328.15	0.99177	0.855	0.862	-0.05436	-0.067	1.008	-0.006	51.596	-0.099
0.50	333.15	0.98685	0.805	0.815	-0.05909	-0.078	1.013	-0.006	52.229	-0.171
0.50	338.15	0.98192	0.764	0.778	-0.06475	-0.089	1.018	-0.006	52.883	-0.246
0.50	343.15	0.97697	0.766	0.784	-0.06993	-0.115	1.024	-0.006	53.687	-0.356
0.75	293.15	1.06255	1.691	1.591	-0.01329	-0.196	0.941	-0.004	47.629	-0.142
0.75	298.15	1.05760	1.544	1.460	-0.01450	-0.163	0.946	-0.004	48.228	-0.131
0.75	303.15	1.05266	1.417	1.346	-0.01576	-0.136	0.950	-0.004	48.832	-0.120
0.75	308.15	1.04770	1.304	1.245	-0.01706	-0.119	0.954	-0.004	49.436	-0.116
0.75	313.15	1.04275	1.203	1.154	-0.01890	-0.106	0.959	-0.004	50.041	-0.117
0.75	318.15	1.03779	1.118	1.078	-0.02083	-0.092	0.964	-0.004	50.659	-0.111
0.75	323.15	1.03283	1.042	1.009	-0.02302	-0.076	0.968	-0.004	51.278	-0.095
0.75	328.15	1.02787	0.975	0.948	-0.02561	-0.073	0.973	-0.004	51.902	-0.122
0.75	333.15	1.02290	0.914	0.894	-0.02851	-0.074	0.978	-0.004	52.530	-0.154
0.75	338.15	1.01792	0.860	0.845	-0.03166	-0.079	0.982	-0.004	53.161	-0.201
0.75	343.15	1.01292	0.812	0.801	-0.03447	-0.139	0.987	-0.005	53.795	-0.415

**Table 3.** Experimental Density ( $\rho$ ), Dynamic Viscosity ( $\eta$ ), Kinematic Viscosity ( $\nu$ ), Excess Molar Volume ( $V^E$ ), Viscosity Deviation ( $\Delta\eta$ ), Thermal Expansion Coefficient ( $\alpha$ ), Excess Thermal Expansion Coefficient ( $\alpha_E$ ), Gibb's Free Energy ( $\Delta G^*$ ) and Excess Gibb's Free Energy of Activation of Viscous Flow ( $\Delta G^E$ ) for Ternary Mixture of Water (1) + DMSO (2) + DMF (3).

$x_1$	$x_2$	$T$	$10^{-3}\rho$	$\eta$	$\nu$	$10^6 V^E$	$\Delta\eta$	$10^3 \alpha$	$10^3 \alpha_E$	$\Delta G^*$	$\Delta G^E$
		/K	/Kg.m <sup>-3</sup>	/mPa.s	/mm <sup>2</sup> .s <sup>-1</sup>	/m <sup>3</sup> .mol <sup>-1</sup>	/mPa.s	/K <sup>-1</sup>	/K <sup>-1</sup>	/kJ.mol <sup>-1</sup>	/kJ.mol <sup>-1</sup>
Water (1) + DMSO (2) + DMF (3)											
0.25	0.25	293.15	1.02577	2.830	2.759	-1.43226	1.533	0.877	-0.080	48.390	2.300
0.25	0.25	298.15	1.02126	2.462	2.411	-1.44862	1.282	0.881	-0.081	48.881	2.227
0.25	0.25	303.15	1.01671	2.160	2.124	-1.46495	1.081	0.885	-0.082	49.381	2.157
0.25	0.25	308.15	1.01211	1.907	1.885	-1.48058	0.915	0.889	-0.083	49.889	2.088
0.25	0.25	313.15	1.00747	1.696	1.683	-1.49589	0.779	0.893	-0.084	50.404	2.021
0.25	0.25	318.15	1.00280	1.517	1.513	-1.51074	0.666	0.897	-0.084	50.927	1.957
0.25	0.25	323.15	0.99807	1.366	1.368	-1.52476	0.574	0.902	-0.085	51.457	1.899
0.25	0.25	328.15	0.99331	1.229	1.237	-1.53811	0.472	0.906	-0.086	51.978	1.755
0.25	0.25	333.15	0.98849	1.116	1.129	-1.55072	0.388	0.910	-0.087	52.517	1.618
0.25	0.25	338.15	0.98363	1.020	1.036	-1.56287	0.312	0.915	-0.087	53.065	1.467
0.25	0.25	343.15	0.97872	0.935	0.955	-1.57345	0.203	0.920	-0.088	53.616	1.169
0.25	0.50	293.15	1.06141	3.394	3.198	-0.85932	1.801	0.848	-0.066	48.799	2.280
0.25	0.50	298.15	1.05724	2.941	2.782	-0.89237	1.501	0.851	-0.067	49.286	2.213
0.25	0.50	303.15	1.05277	2.587	2.457	-0.91140	1.278	0.855	-0.068	49.800	2.167
0.25	0.50	308.15	1.04823	2.302	2.196	-0.92770	1.104	0.859	-0.069	50.333	2.131
0.25	0.50	313.15	1.04412	2.041	1.954	-0.97079	0.940	0.862	-0.070	50.847	2.070
0.25	0.50	318.15	1.03953	1.821	1.752	-0.98777	0.804	0.866	-0.071	51.370	2.013
0.25	0.50	323.15	1.03488	1.635	1.580	-1.00350	0.695	0.870	-0.072	51.898	1.965
0.25	0.50	328.15	1.03019	1.476	1.433	-1.01806	0.593	0.874	-0.072	52.435	1.879
0.25	0.50	333.15	1.02545	1.338	1.304	-1.03154	0.504	0.878	-0.073	52.974	1.788
0.25	0.50	338.15	1.02066	1.216	1.191	-1.04414	0.422	0.882	-0.074	53.514	1.679
0.25	0.50	343.15	1.01582	1.112	1.094	-1.05534	0.311	0.886	-0.074	54.063	1.441
0.50	0.25	293.15	1.04009	3.073	2.954	-0.87827	1.774	0.769	-0.097	47.928	2.704
0.50	0.25	298.15	1.03636	2.639	2.546	-0.90351	1.467	0.772	-0.098	48.377	2.632
0.50	0.25	303.15	1.03257	2.293	2.220	-0.92899	1.229	0.775	-0.100	48.843	2.570
0.50	0.25	308.15	1.02871	1.999	1.944	-0.95445	1.028	0.778	-0.101	49.307	2.498
0.50	0.25	313.15	1.02478	1.760	1.717	-0.97954	0.867	0.781	-0.103	49.785	2.432
0.50	0.25	318.15	1.02078	1.558	1.526	-1.00425	0.734	0.784	-0.104	50.268	2.365
0.50	0.25	323.15	1.01671	1.388	1.365	-1.02827	0.626	0.787	-0.105	50.758	2.304
0.50	0.25	328.15	1.01257	1.239	1.224	-1.05162	0.522	0.790	-0.107	51.246	2.197
0.50	0.25	333.15	1.00836	1.118	1.108	-1.07411	0.439	0.793	-0.108	51.752	2.101
0.50	0.25	338.15	1.00407	1.015	1.011	-1.09587	0.367	0.797	-0.109	52.270	1.998
0.50	0.25	343.15	0.99971	0.924	0.924	-1.11698	0.274	0.800	-0.110	52.787	1.786

**Table 4.** Fitting Parameters ( $A_j$ ) and Standard Deviations ( $\sigma$ ) of Excess Molar Volume ( $V^E$ ), Viscosity Deviation ( $\Delta\eta$ ) and Excess Gibb's Free Energy ( $\Delta G^E$ ) for Binary Mixture of Water (1) + DMSO (2), Water (1) + DMF (2) and DMSO (1) + DMF (2).

$T/K$	$10^6 V^E / \text{m}^3 \cdot \text{mol}^{-1}$			$\Delta\eta / \text{mPa} \cdot \text{s}$			$10^3 \alpha_E / \text{K}^{-1}$			$\Delta G^E / \text{kJmol}^{-1}$		
	$A_0$	$A_1$	$\sigma$	$A_0$	$A_1$	$\sigma$	$A_0$	$A_1$	$\sigma$	$A_0$	$A_1$	$\sigma$
<u>Water (1) + DMSO (2)</u>												
293.15	0.842	5.202	0.035	-0.322	-0.317	0.019	7.471	-8.025	0.062	10.318	-2.907	0.067
298.15	0.623	5.032	0.008	-0.329	-0.324	0.019	6.109	-6.865	0.061	10.004	-2.974	0.068
303.15	0.392	5.534	0.039	-0.337	-0.321	0.019	5.108	-5.597	0.064	9.813	-2.542	0.080
308.15	0.286	5.482	0.041	-0.342	-0.327	0.020	4.343	-4.376	0.074	9.701	-1.784	0.110
313.15	0.199	5.437	0.043	-0.347	-0.332	0.020	3.689	-3.564	0.071	9.533	-1.356	0.121
318.15	0.102	5.406	0.045	-0.352	-0.336	0.020	3.027	-3.397	0.046	9.096	-2.074	0.079
323.15	0.107	5.762	0.029	-0.355	-0.334	0.020	2.623	-2.761	0.050	9.009	-1.502	0.102
328.15	-0.056	5.352	0.051	-0.361	-0.345	0.020	2.208	-2.541	0.035	8.586	-1.302	0.073
333.15	-0.132	5.324	0.053	-0.366	-0.349	0.020	2.101	-1.145	0.089	9.125	1.391	0.242
338.15	-0.203	5.278	0.056	-0.370	-0.354	0.020	1.725	-1.305	0.062	8.646	0.438	0.185
343.15	-0.256	5.258	0.059	-0.374	-0.358	0.021	0.617	-5.587	0.344	7.706	-0.290	0.103
<u>Water (1) + DMF (2)</u>												
293.15	-5.824	3.824	0.105	-0.797	-0.501	0.018	6.362	-3.694	0.080	11.382	-3.184	0.001
298.15	-7.105	5.262	0.032	-0.823	-0.486	0.019	5.246	-3.082	0.057	10.926	-2.867	0.011
303.15	-7.208	5.113	0.032	-0.831	-0.495	0.019	4.369	-2.619	0.043	10.556	-2.678	0.011
308.15	-7.329	5.046	0.031	-0.839	-0.502	0.019	3.675	-2.237	0.034	10.216	-2.496	0.009
313.15	-7.463	4.921	0.030	-0.848	-0.510	0.019	3.092	-1.939	0.027	9.858	-2.373	0.008
318.15	-7.579	4.805	0.030	-0.856	-0.519	0.019	2.622	-1.638	0.020	9.534	-2.130	0.010
323.15	-7.687	4.707	0.029	-0.864	-0.526	0.019	2.219	-1.383	0.014	9.187	-1.884	0.015
328.15	-7.799	4.576	0.029	-0.872	-0.535	0.019	1.830	-1.094	0.013	8.692	-1.895	0.006
333.15	-7.848	4.727	0.014	-0.879	-0.538	0.019	1.491	-0.829	0.013	7.984	-0.653	0.003
338.15	-8.004	4.366	0.026	-0.887	-0.550	0.019	1.182	-0.588	0.014	7.303	-0.015	0.018
343.15	-8.109	4.248	0.026	-0.895	-0.558	0.019	1.264	-1.289	0.139	6.110	1.084	0.044
<u>DMSO (1) + DMF (2)</u>												
293.15	0.243	-1.573	0.086	-0.016	-0.021	0.001	-0.984	0.277	0.036	-0.913	1.073	0.069
298.15	0.238	-1.596	0.088	-0.016	-0.022	0.001	-0.843	0.261	0.028	-0.876	1.026	0.060
303.15	0.231	-1.618	0.089	-0.017	-0.022	0.001	-0.720	0.250	0.024	-0.826	0.997	0.056
308.15	0.224	-1.636	0.091	-0.017	-0.022	0.001	-0.618	0.211	0.022	-0.772	0.914	0.055
313.15	0.216	-1.656	0.093	-0.017	-0.022	0.001	-0.535	0.170	0.021	-0.723	0.819	0.057
318.15	0.206	-1.672	0.094	-0.018	-0.023	0.001	-0.408	-0.079	0.008	-0.493	0.004	0.018
323.15	0.195	-1.689	0.095	-0.018	-0.023	0.001	-0.232	-0.406	0.005	-0.082	-1.156	0.028
328.15	0.183	-1.705	0.097	-0.019	-0.023	0.001	-0.222	-0.466	0.012	-0.201	-1.415	0.047
333.15	0.167	-1.718	0.098	-0.019	-0.023	0.001	-0.188	-0.745	0.030	-0.227	-2.407	0.110
338.15	0.149	-1.728	0.099	-0.020	-0.023	0.001	-0.035	-1.633	0.078	0.087	-5.182	0.259
343.15	0.133	-1.736	0.100	-0.020	-0.023	0.001	0.782	-0.252	0.487	1.151	-13.597	0.622



**Table 5.** Activation Energy of Viscous Flow ( $E_a$ ), Entropic Factor ( $\ln(A_s)$ ), Activation Temperature ( $T^*$ ), Arrhenius Temperature ( $T_A$ ), Enthalpy Change ( $\Delta H^*$ ) and Entropy Change ( $\Delta S^*$ ) for Binary Mixture of Water (1) + DMSO (2), Water (1) + DMF (2) and DMSO (1) + DMF (2).

$x_1$	$E_a/$ J.mol <sup>-1</sup>	$\ln(A_s)/$ mPa.s	$T^*/$ K	$T_A/$ K	$\Delta H^*/$ kJ.mol <sup>-1</sup>	$\Delta S^*/$ J.mol <sup>-1</sup>
<u>Water (1) + DMSO (2)</u>						
0.25	19697.79	-6.6857	2369.1	354.35	19.01	-102.92
0.50	20338.83	-7.1455	2446.2	342.34	19.73	-96.76
0.75	15945.47	-5.8779	1917.8	326.27	15.47	-104.02
<u>Water (1) + DMF (2)</u>						
0.25	18516.30	-6.7176	2227.0	331.52	17.73	-103.39
0.50	20279.80	-7.3629	2439.1	331.27	19.65	-95.20
0.75	17953.41	-6.8220	2159.3	316.52	17.43	-96.31
<u>DMSO (1) + DMF (2)</u>						
0.25	2998.44	-1.0717	360.6	336.50	-3.84	-170.23
0.50	10220.13	-3.8956	1229.2	315.54	9.40	-128.64
0.75	12279.63	-4.5229	1476.9	326.54	11.48	-123.21

**Table 6.** Activation Energy of Viscous Flow ( $E_a$ ), Entropic Factor ( $\ln(A_s)$ ), Activation Temperature ( $T^*$ ), Arrhenius Temperature ( $T_A$ ), Enthalpy change ( $\Delta H^*$ ) and Entropy change ( $\Delta S^*$ ) for Ternary Mixture of Water (1) + DMSO (2) + DMF (3).

$x_1$	$x_2$	$E_a/$ J.mol <sup>-1</sup>	$\ln(A_s)/$ mPa.s	$T^*/$ K	$T_A/$ K	$\Delta H^*/$ kJ.mol <sup>-1</sup>	$\Delta S^*/$ J.mol <sup>-1</sup>
<u>Water (1) + Methanol (2) + Ethanol (3)</u>							
0.25	0.25	18521.29	-6.5753	2227.6	338.78	17.74	-104.41
0.25	0.50	18607.76	-6.4274	2238.0	348.20	17.87	-105.35
0.50	0.25	20101.87	-7.1441	2417.7	338.42	19.44	-97.00

## Graphical Abstract

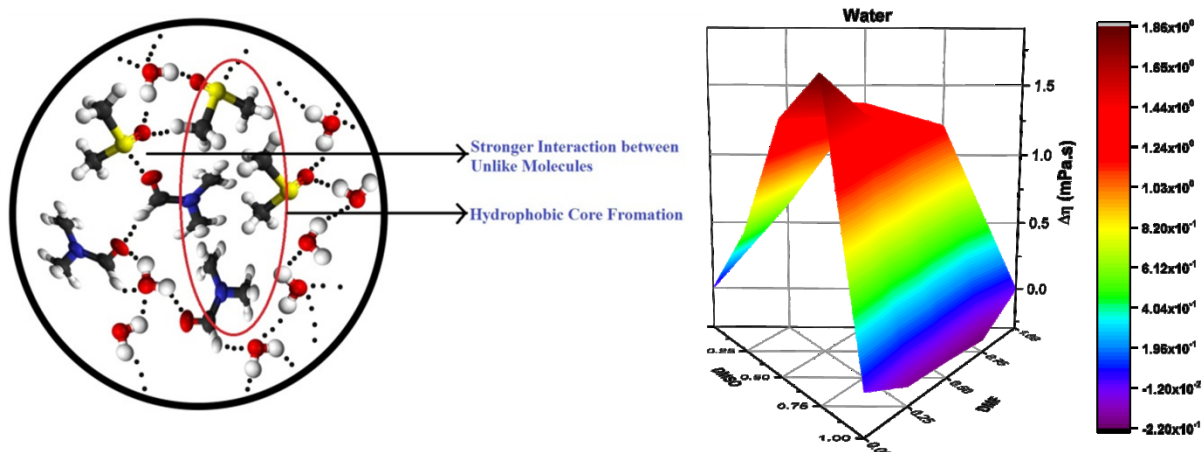
# The density, dynamic viscosity and kinematic viscosity of protic and aprotic polar solvents (pure and mixed) systems: An experimental and theoretical insight of thermophysical properties

Md. Sayem Alam<sup>a,b,\*</sup>, Baskar Ashokkumar<sup>a</sup>, A. Mohammed Siddiq<sup>a</sup>

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<sup>b</sup>Chemical Sciences, Academy of Scientific and Innovative Research (AcSIR), New Delhi-110001, India.

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[sayem@clri.res.in](mailto:sayem@clri.res.in).



Distribution of viscosity deviation for ternary mixed system of water, DMSO and DMF at 298.15 K.

## Supporting Information

# The density, dynamic viscosity and kinematic viscosity of protic and aprotic polar solvents (pure and mixed) systems: An experimental and theoretical insight of thermophysical properties

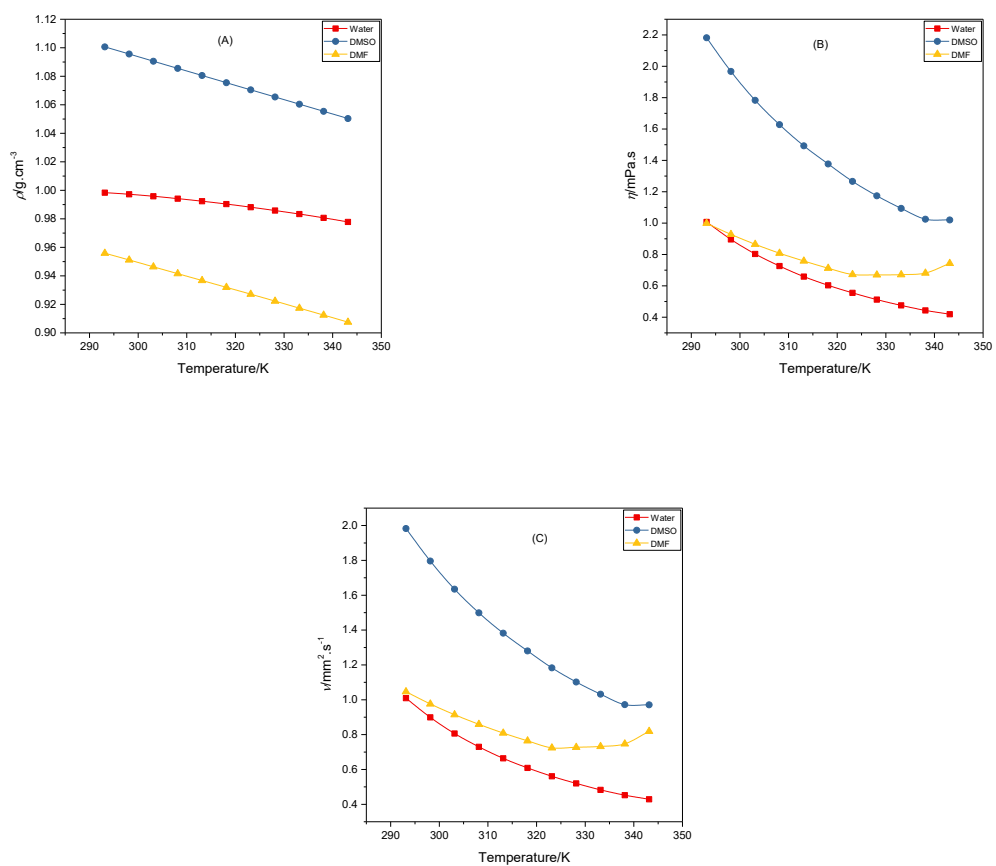
Md. Sayem Alam<sup>a,b,\*</sup>, Baskar Ashokkumar<sup>a</sup>, A. Mohammed Siddiq<sup>a</sup>

<sup>a</sup>*Council of Scientific and Industrial Research (CSIR) — Central Leather Research Institute (CLRI), Polymer Science and Technology Laboratory, Adyar, Chennai 600 020, India.*

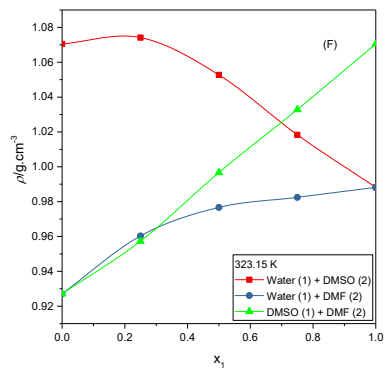
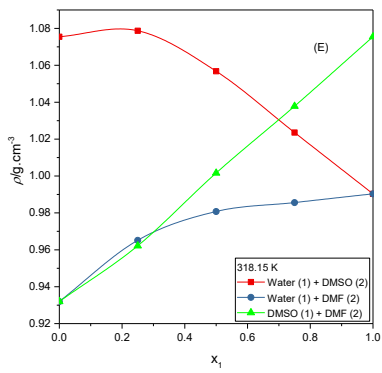
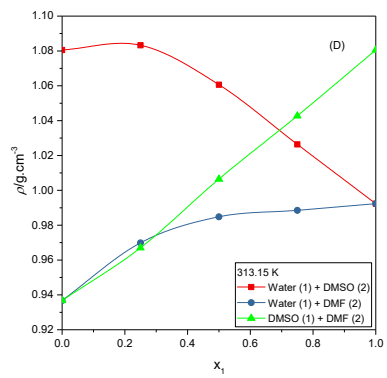
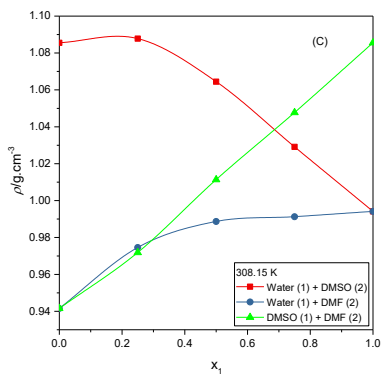
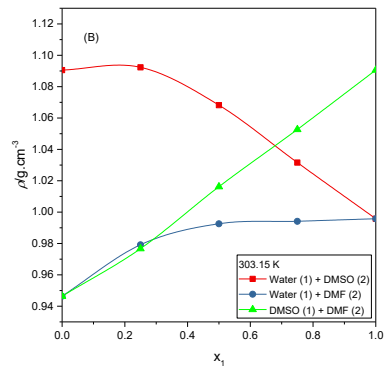
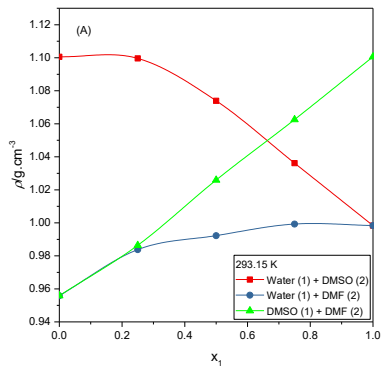
<sup>b</sup>*Chemical Sciences, Academy of Scientific and Innovative Research (AcSIR), New Delhi-110001, India.*

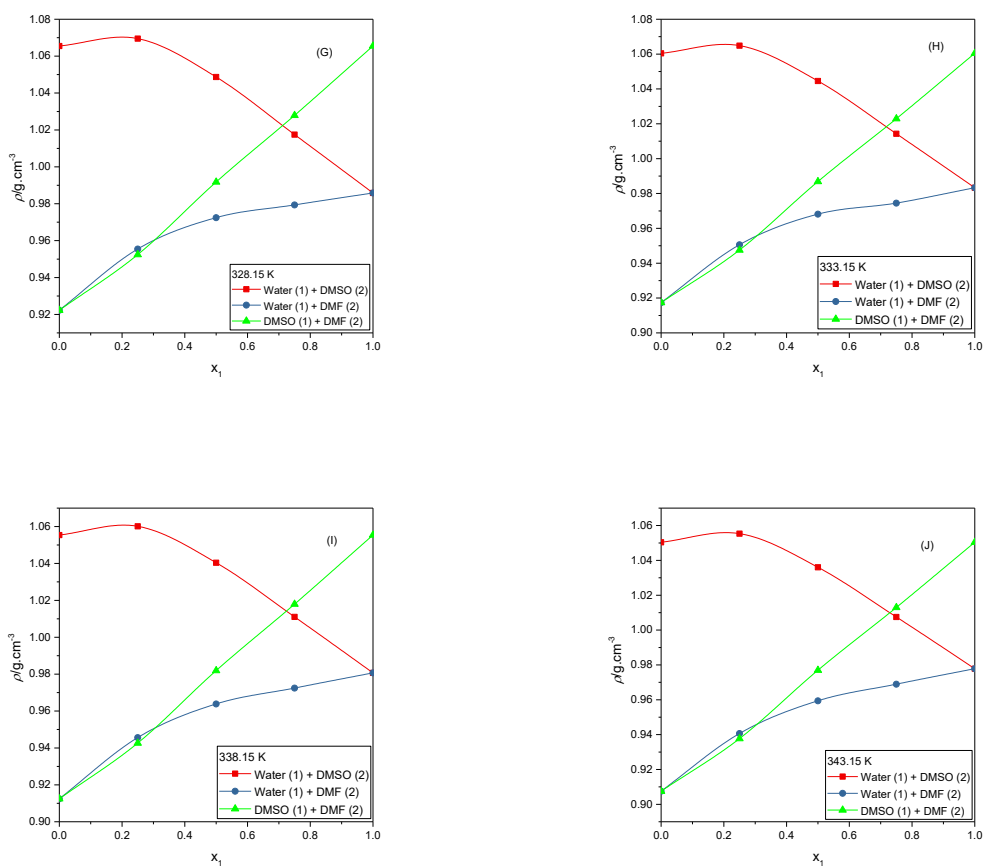
*\*To whom correspondence may be addressed: Tel.: + 91-44-24437106. E-mail: [sayemalam@gmail.com](mailto:sayemalam@gmail.com), [sayem@clri.res.in](mailto:sayem@clri.res.in).*

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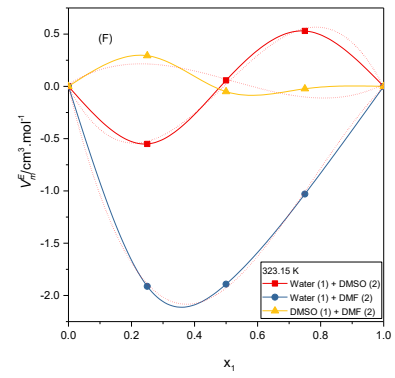
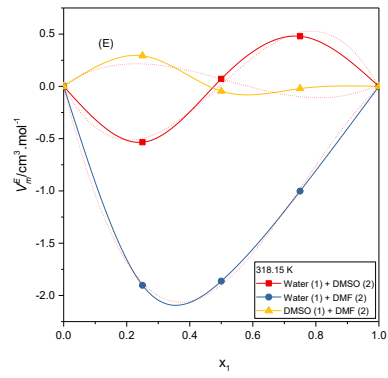
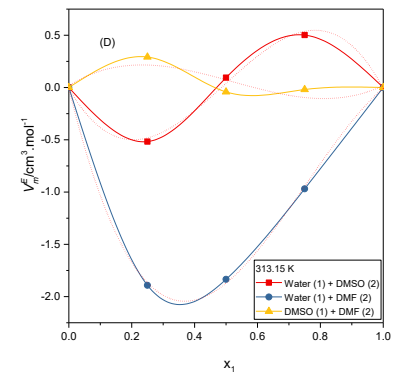
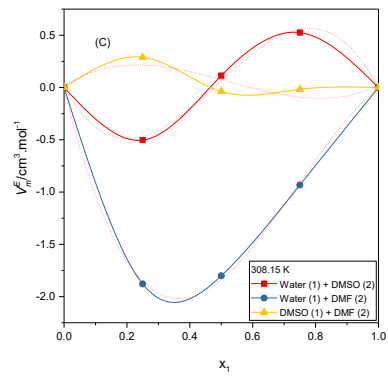
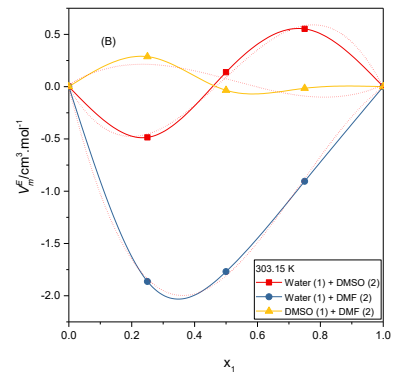
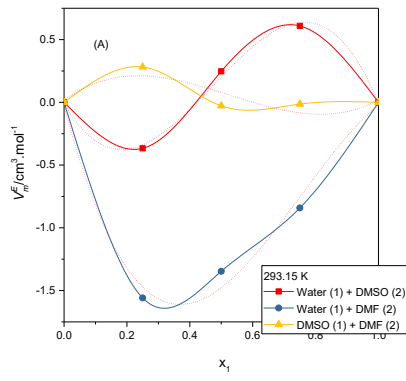


**Fig. S1.** (A) Density ( $\rho$ ), (B) dynamic viscosity ( $\eta$ ) and (C) kinematic viscosity ( $\nu$ ) of pure systems: water, DMSO and DMF as function of temperature from 293.15 K to 343.15 K.

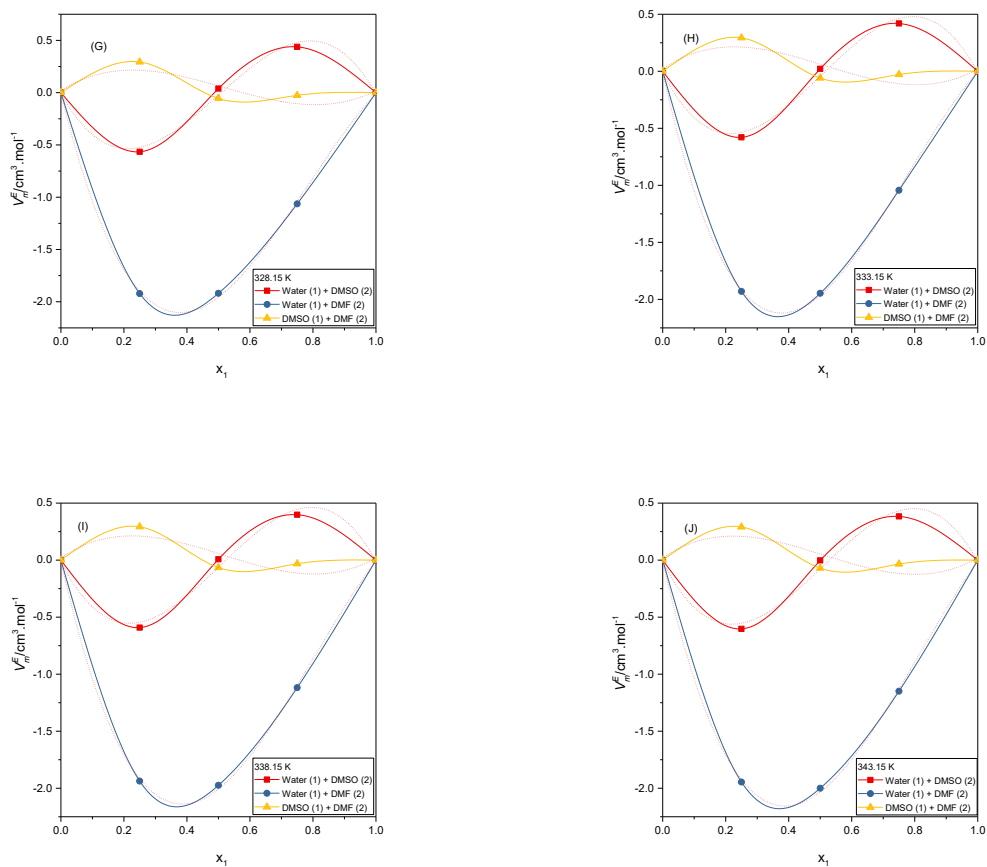




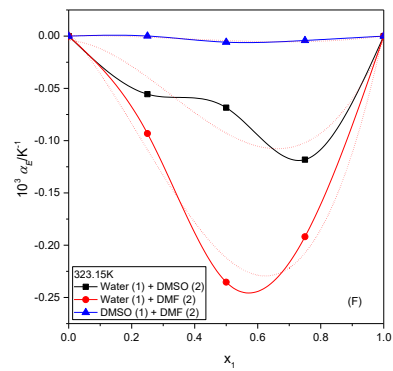
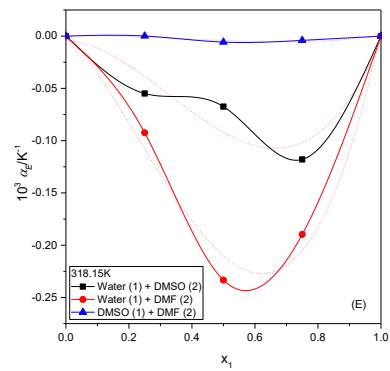
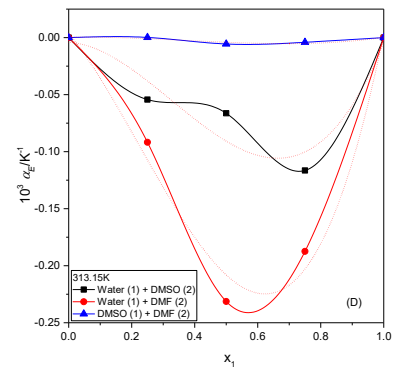
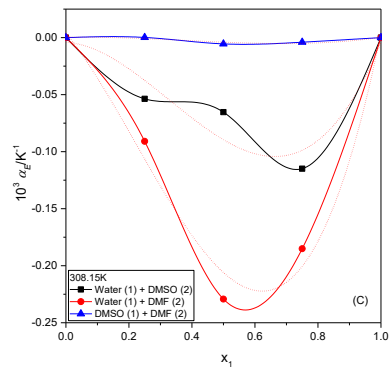
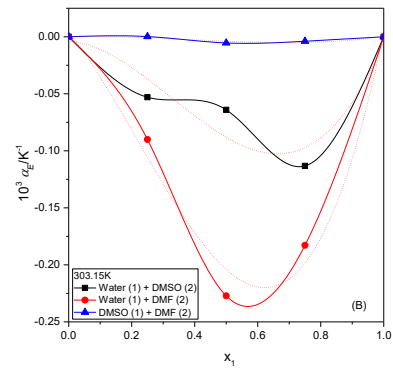
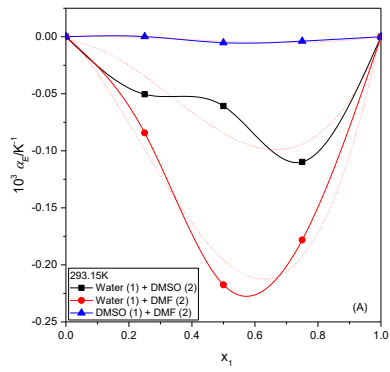
**Fig. S2.** Density ( $\rho$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).

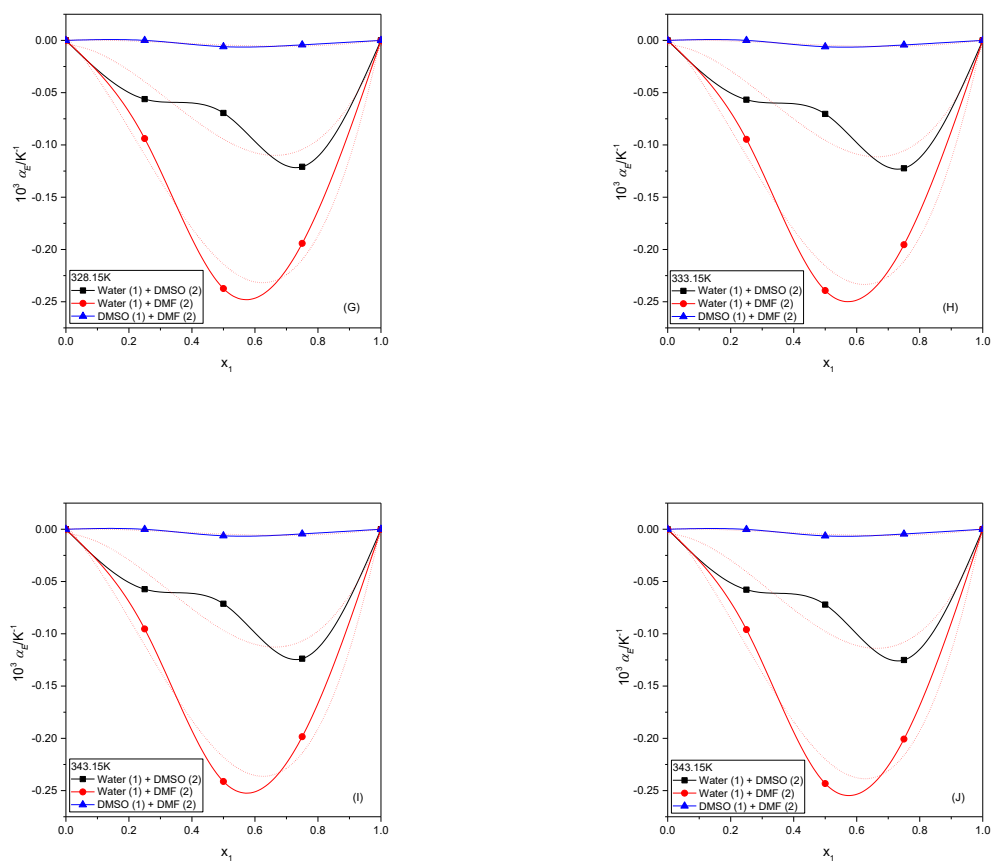




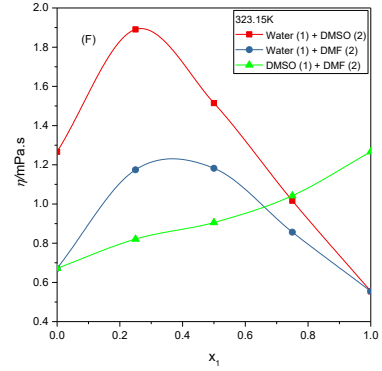
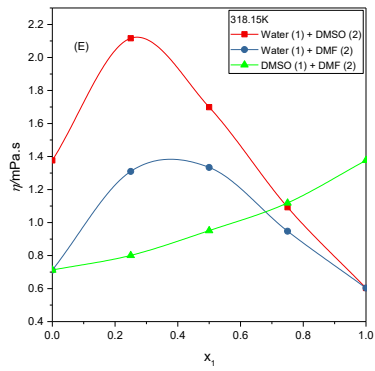
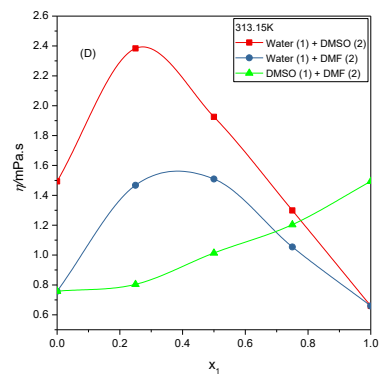
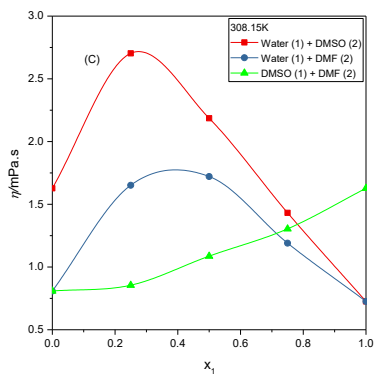
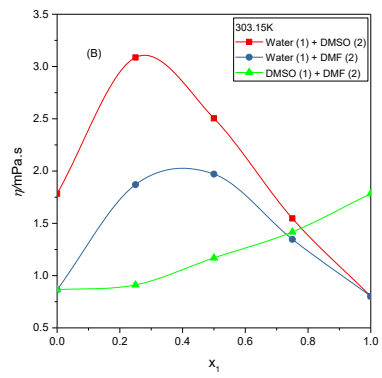
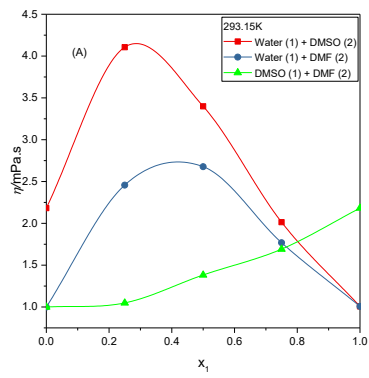


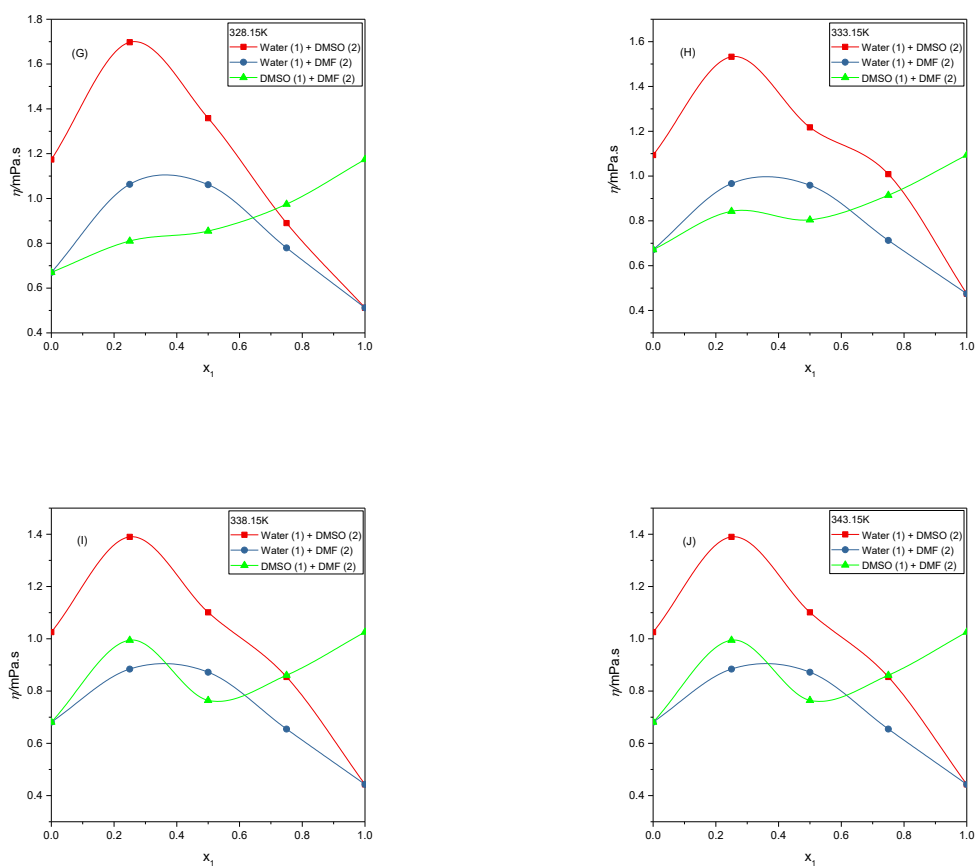
**Fig. S3.** Excess molar volume ( $V^E$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).



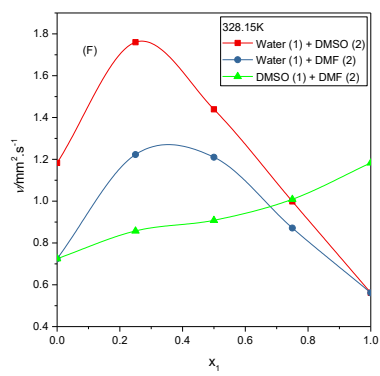
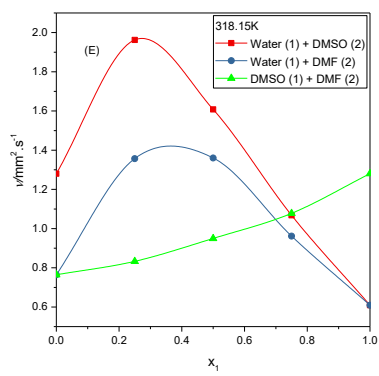
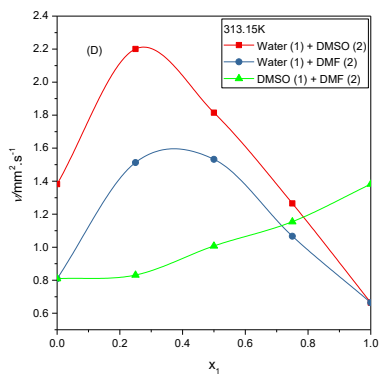
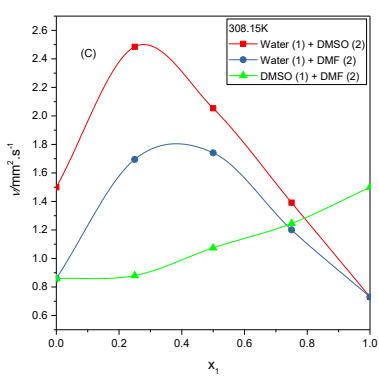
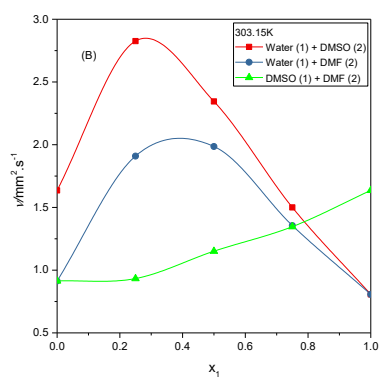
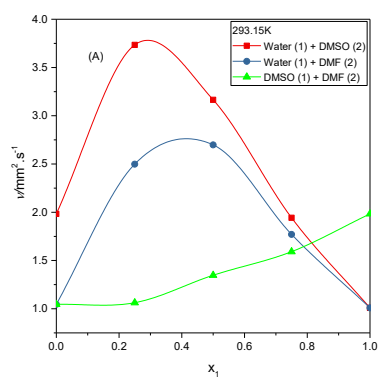


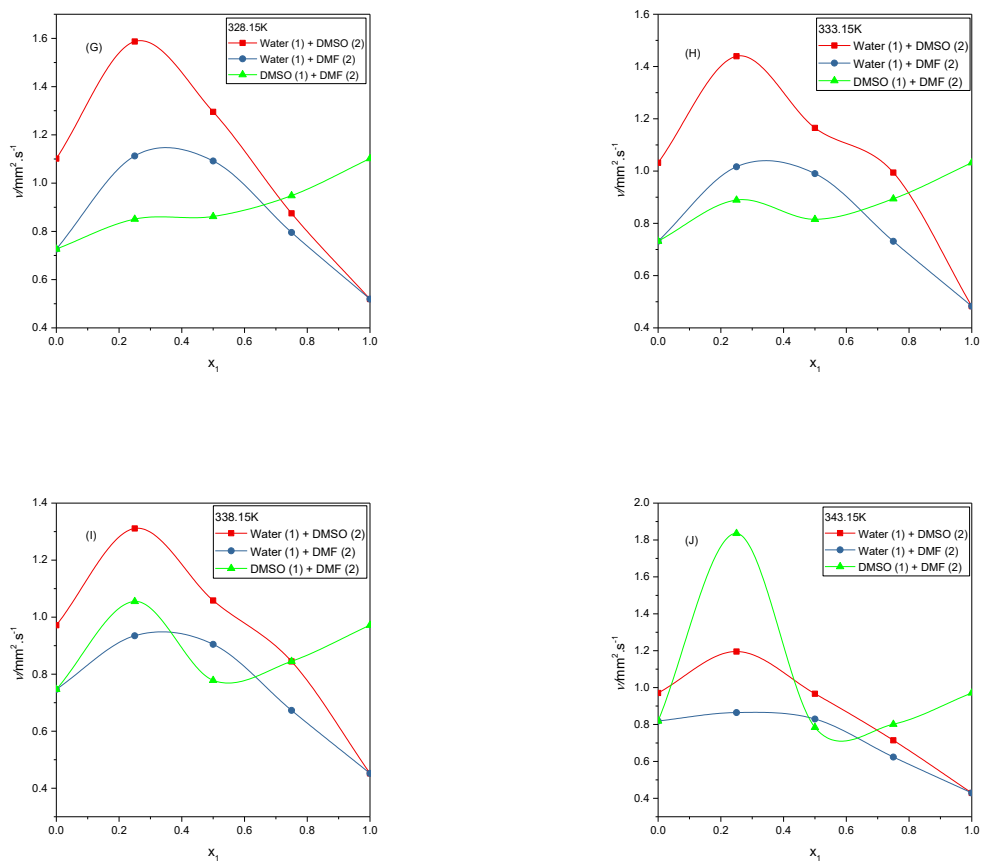
**Fig. S4.** Excess thermal expansion coefficient ( $\alpha_E$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).



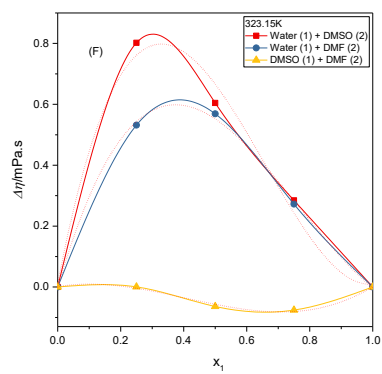
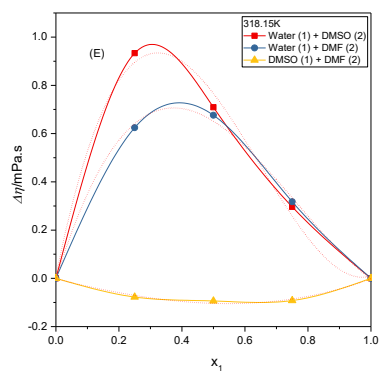
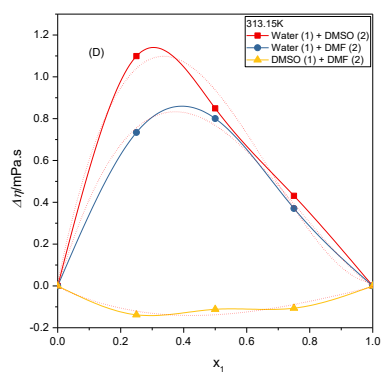
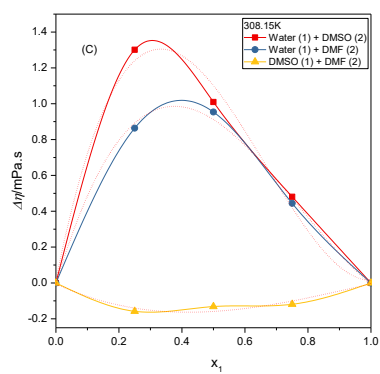
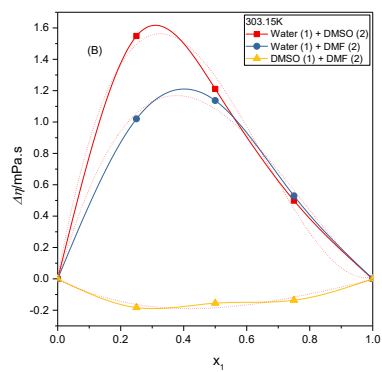
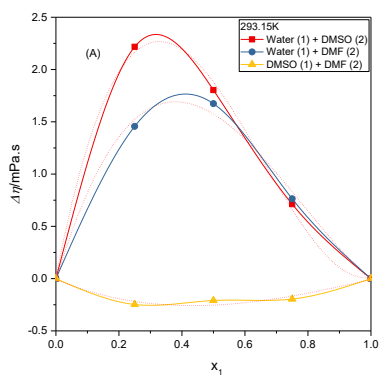


**Fig. S5.** Dynamic viscosity ( $\eta$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).

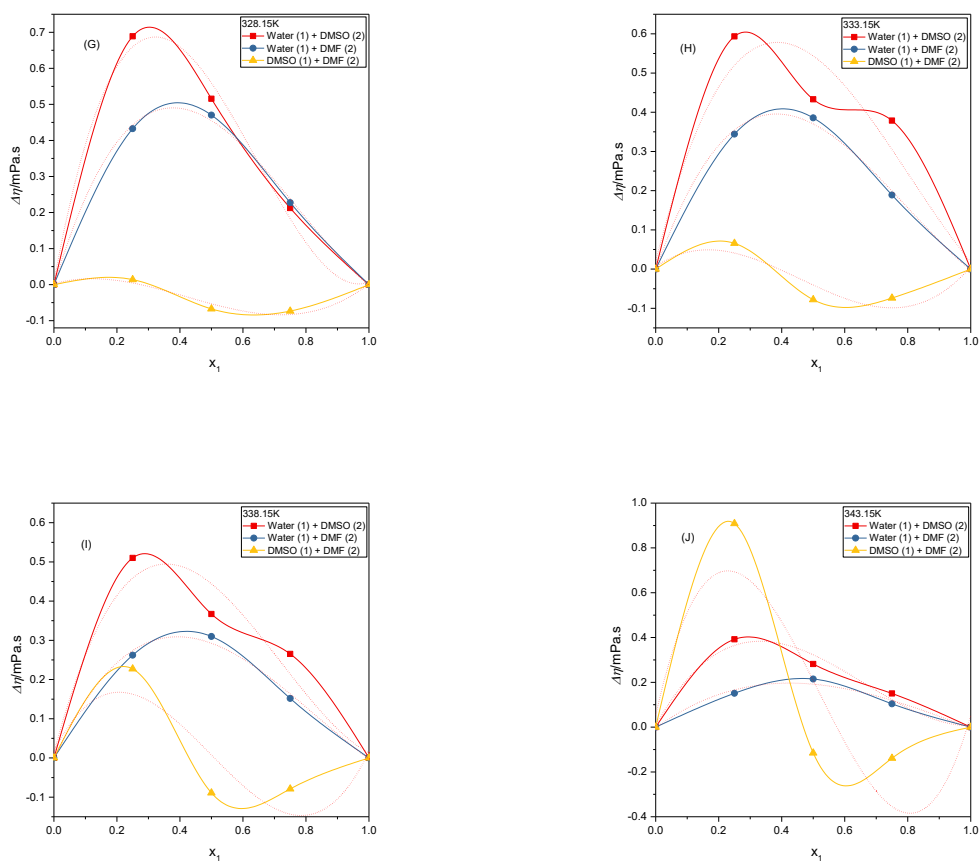




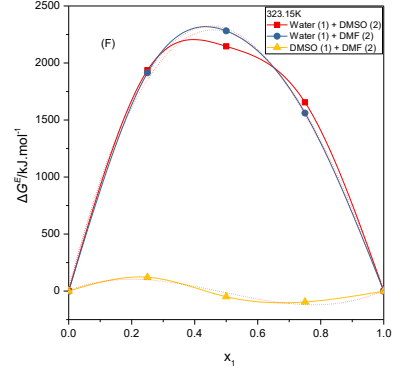
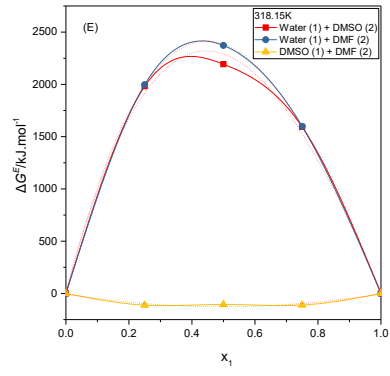
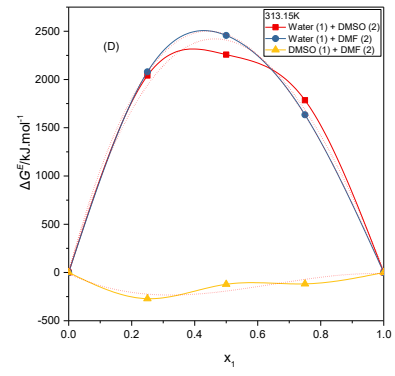
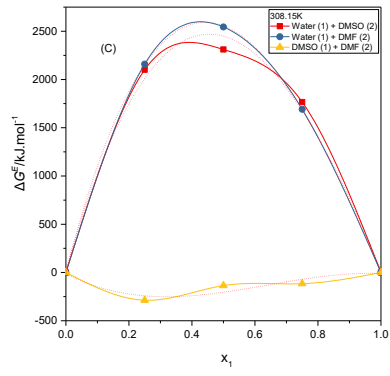
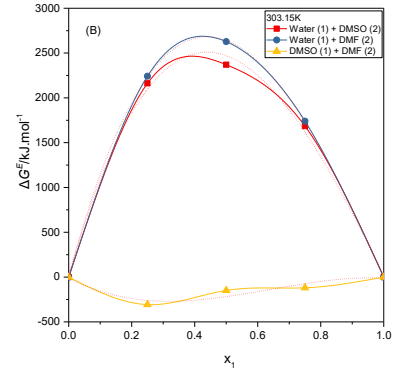
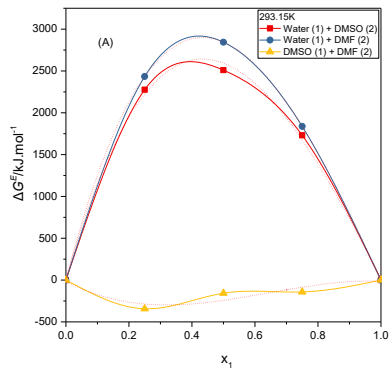
**Fig. S6.** Kinematic viscosity ( $\nu$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).

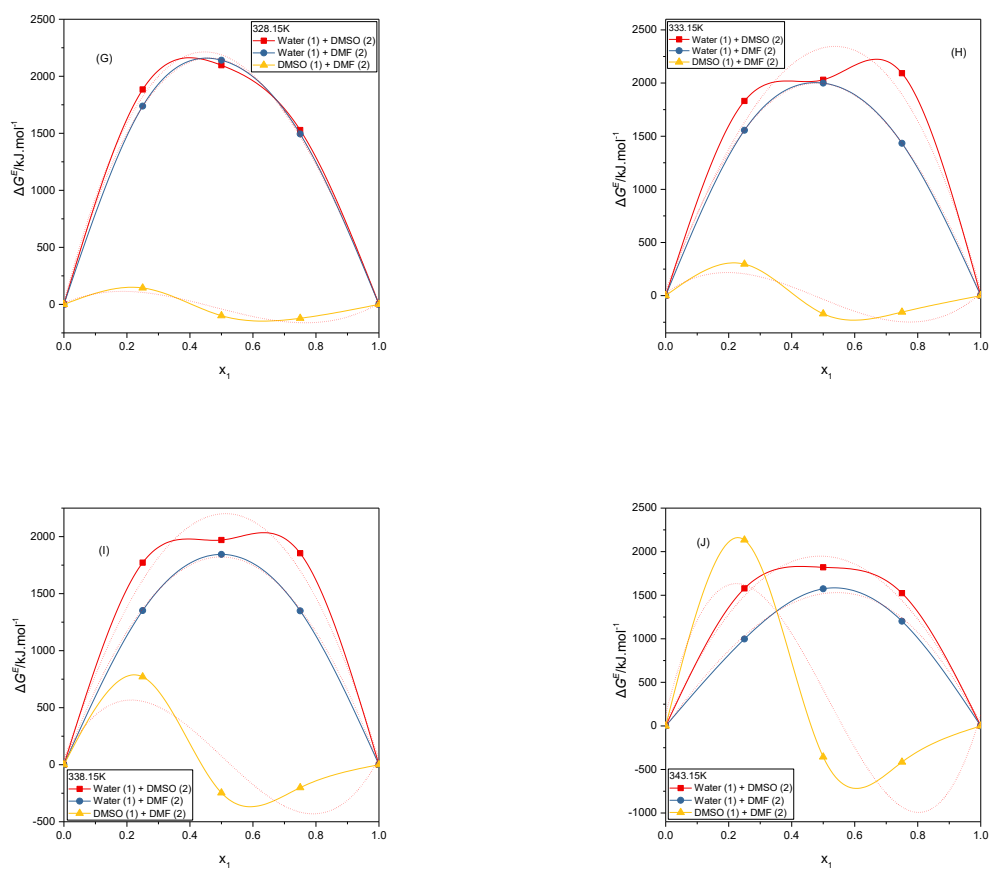






**Fig. S7.** Viscosity deviation ( $\Delta\eta$ ) of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).





**Fig. S8.** Excess Gibb's free energy ( $\Delta G^E$ ) for activation of viscous flow of binary mixtures: water (1) + DMSO (2), water (1) + DMF (2) and DMSO (1) + DMF (2) as function of mole fraction from 293.15 K to 343.15 K (A to J).