



A New Mathematical Model for the Characterization of Viscosity Variations in the Aqueous Solutions of Monoalcohols and Polyalcohols

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Abstract

This study aims to investigate the viscosity of binary mixtures of water with different monoalcohols and polyalcohols. The viscosity values of the mixtures of methanol, ethanol, 1-propanol, and 2-propanol were taken from a study published by Herraez and Belda (2004). The values for ethylene glycol, glycerin, and sorbitol were measured using a similar protocol. The viscosity of the mixture is expressed as a function of the molar fraction of the alcohols that ranges from 0 (pure solvent, water in this case) to 1 (pure solute). To represent the viscosity of these mixtures, a new variable, Φ , was introduced. This representation provides a function, $\eta = f(\Phi)$, that has an exponential nature, with which excellent correlation coefficients are achieved with only one coefficient for monoalcohols or two coefficients for polyalcohols. Additionally, the variable Φ highlights the possible critical points, where physicochemical interactions occur between the components of the mixture.

Keywords Monoalcohols · Polyalcohols · Concentration · Viscosity · Mole ratio (Φ)

1 Introduction

Short-chain alcohols and polyols, such as ethylene glycol and glycerin, can be used in various mixtures to create formulations for topical, oral mucosal, and oral administration. These formulations can take the form of liquids, pastes, emulsions, and gels. These substances have many applications in the pharmaceutical [1], food [2], and cosmetic [3, 4] industries [5]. They are frequently used in the pharmaceutical and food industries to control intermediate and final product manufacturing processes.

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Alcoholic solvents have the unique property of –OH functional groups, which form hydrogen bonds with water [6]. This has a significant impact on the physicochemical properties of the mixture, particularly on viscosity. This can be beneficial in certain applications, such as in the creation of thicker emulsions or gels. However, it can also be a disadvantage in cases where a lower viscosity is desired. In addition to impacting viscosity, the hydrogen bonding between alcohols and water also affects other physicochemical properties of the mixture, such as its surface tension and solubility. Understanding and controlling these properties is important in a variety of industries, including pharmaceuticals, food, and cosmetics [7].

Viscosity is a crucial thermodynamic property of all types of solutions [8–14] as it helps to characterize the purity of a product. In the cosmetic industry, viscosity can be modified to create appropriate emulsions in formulations [15]. In academic and research settings, understanding the viscosity of a substance can provide insight into the interactions between solute and solvent molecules [16–18]. This is particularly important in industrial processes and pharmaceutical formulations, in which binary mixtures are commonly used in the form of solutions for active principles and excipients.

The viscosity of the mixture is expressed as a function of the mole fraction of the alcohols (methanol, ethanol, 1-propanol, and 2-propanol) that ranges from 0 (pure solvent, water in this case) to 1 (pure solute). The values are shown in Table 1 and are represented in Fig. 1.

Figure 1 illustrates the relationship between viscosity and concentration for the four monoalcohols studied. It shows that a maximum viscosity occurs at different weight fractions for each alcohol, but when represented as a function of the mole fraction (Fig. 1, right), it can be seen that the maximum viscosity corresponds to a mole fraction of 0.25 for all four alcohols, which is equivalent to three water molecules for each alcohol molecule. Therefore, solutions were prepared at this concentration for each monoalcohol. From this point, the viscosity decreases as the proportion of alcohol increases until reaching the viscosity of pure alcohol at a mole fraction of 1.

This is because, when water is added to alcohols, hydrogen bonds form between the water molecules and the –OH groups of the alcohols up to a maximum of three water molecules for each –OH group. This association causes an increase in the size of the molecular package, resulting in increased viscosity. If water is added beyond this point, the new molecules no longer have free –OH groups to associate with, and the viscosity of the mixture tends towards that of water as the proportion of water increases.

Therefore, a new mathematical model has been developed to predict the variations of viscosity of a mixture based on the viscosity of its pure components and the proportion in which it is made.

2 Experimental

2.1 Materials

In this work, the viscosity of mixtures containing short-chain alcohols (methanol, ethanol, 1-propanol, and 2-propanol) and polyols (ethylene glycol, glycerin, and sorbitol) were studied. The viscosity values for the mixtures of methanol, ethanol, 1-propanol, and 2-propanol, with a purity of over 99%, were taken from a study by Herraes and Belda [19]. The values for ethylene glycol, glycerin, and sorbitol were measured using a similar protocol as

Table 1 Values of the viscosities in mPa.s for the indicated monoalcohols, at the different concentrations together with the corresponding mole fractions

Mixture Methanol + Water			Mixture Ethanol + Water		
C wt%	Mole fraction	Viscosity	C wt%	Mole fraction	Viscosity
0.00	0.0000	0.891	0.00	0.0000	0.891
10.00	0.0580	1.170	10.00	0.0417	1.328
20.00	0.1233	1.410	20.00	0.0891	1.820
30.00	0.1945	1.571	30.00	0.1436	2.201
37.21	0.2500	1.672	40.00	0.2069	2.344
40.00	0.2727	1.658	46.00	0.2500	2.372
50.00	0.3600	1.576	50.00	0.2813	2.355
60.00	0.4576	1.417	60.00	0.3698	2.257
70.00	0.5676	1.226	70.00	0.4773	2.028
80.00	0.6923	1.018	80.00	0.6102	1.768
90.00	0.8351	0.783	90.00	0.7789	1.403
100.00	1.0000	0.553	100.00	1.0000	1.076
Mixture 1-Propanol + Water			Mixture 2-Propanol + Water		
C wt%	Mole fraction	Viscosity	C wt%	Mole fraction	Viscosity
0.00	0.0000	0.891	0.00	0.0000	0.891
10.00	0.0323	1.258	10.00	0.0323	1.286
20.00	0.0698	1.848	20.00	0.0698	1.964
30.00	0.1139	2.208	30.00	0.1139	2.389
40.00	0.1667	2.436	40.00	0.1667	2.660
50.00	0.2308	2.591	50.00	0.2308	2.847
52.63	0.2500	2.624	52.63	0.2500	2.886
60.00	0.3103	2.597	60.00	0.3103	2.837
70.00	0.4118	2.538	70.00	0.4118	2.750
80.00	0.5455	2.421	80.00	0.5455	2.594
90.00	0.7297	2.227	90.00	0.7297	2.358
100.00	1.0000	1.946	100.00	1.0000	2.031

The dispersion of the measurements was less than 1%, thus the average of the three measurements was taken as the final value [Data taken from Herraéz and Belda [19]]

the one described in the cited study. The polyalcohols as the ethylene glycol and glycerin with a purity of over 99.0% by volume in each case and sorbitol 70 wt% in water (Merk®) were used.

2.2 Viscosity Measurement

The viscosities of mixtures containing methanol, ethanol, 1-propanol, and 2-propanol were measured using an Ostwald viscometer at a temperature of 25.0 ± 0.1 °C. To ensure accuracy, the liquid was kept in a thermostatic water bath (25.0 ± 0.1 °C) for a minimum of 10 min before measurement, and three successive measurements were taken. The

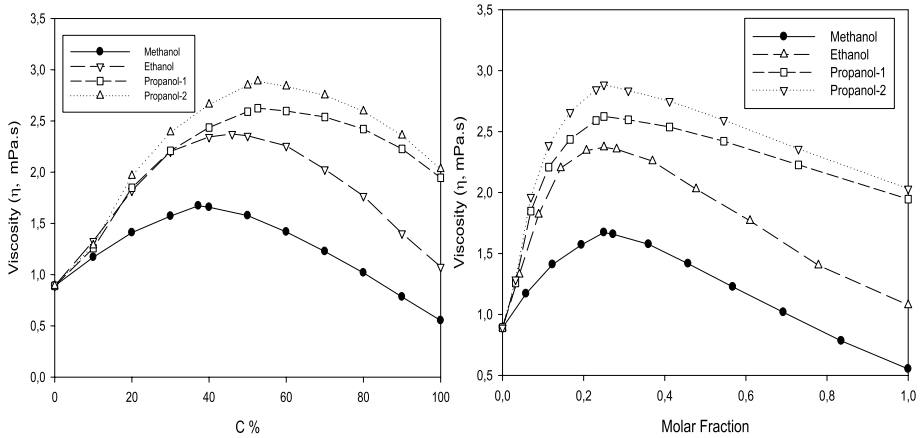


Fig. 1 Viscosity values vs. concentration (wt%) (left) and viscosity values as a function of mole fraction (right) for the monoalcohols. Data taken from Herraez and Belda [19]

dispersion of the measurements was less than 1%, thus the average of the three measurements was taken as the final value.

In this study, for the measurements of ethylene glycol, glycerin, and sorbitol, a BOHLIN VISCO 88BV rotary viscometer was used following the same protocol as the one used for the monoalcohols. In the case of the rotational viscometer, a silicone standard solution (at 25.0 ± 0.1 °C) has been used for the viscometer calibration. The viscosity of this solution was obtained with an accuracy of $\pm 1\%$ of the nominal value.

3 Results and Discussion

Based on the results presented, it is possible to represent the viscosity of the mixture as a function of the relationship between the number of solvent molecules (water) and the number of solute molecules (alcohol), which allows the definition of a new variable, Φ , as the ratio of the number of moles of water to the number of moles of alcohol.

The value of Φ can be easily obtained from the mole fraction of alcohol as follows:

$$\begin{aligned}\phi &= \frac{\text{Moles water}}{\text{Moles alcohol}} = \frac{N_w}{N_{ol}} \\ F_m &= \frac{\text{Moles alcohol}}{\text{Moles totals}} = \frac{N_{ol}}{(N_w + N_{ol})} \\ \phi &= \frac{1}{F_m} - 1\end{aligned}\quad (1)$$

Equation 1 allows for an easy transfer of the values of the mole fraction (F_m) to the corresponding values of Φ . The results for methanol and ethanol are shown in Table 2, and for propanol 1 and propanol 2 in Table 3. At a concentration of 0% alcohol, a value of Φ equal to 100 is attributed so that the adjustment functions $\eta=f(\Phi)$ tend asymptotically to the viscosity of water.

Table 2 Values of the viscosity increase (inc) and decrease (dec) of the mixtures of monoalcohols (methanol and ethanol) and water for the indicated values of Φ ($n=3$)

Methanol			Ethanol		
Φ	Viscosity (η , mPa·s) (inc)	Viscosity (η , mPa·s) (dec)	Φ	Viscosity (η , mPa·s) (inc)	Viscosity (η , mPa·s) (dec)
0.1976	0.783	–	0.2850	1.403	–
0.4445	1.018	–	0.6390	1.768	–
0.7600	1.226	–	1.0951	2.028	–
1.1830	1.417	–	1.7030	2.257	–
1.7800	1.576	–	2.587	2.355	–
2.6600	1.658	–	3.0000	2.372	2.372
3.0000	1.672	1.672	3.8300	–	2.344
4.1500	–	1.571	5.9400	–	2.201
7.1300	–	1.410	10.241	–	1.820
16.240	–	1.170	22.810	–	1.320
100	–	0.891	100	–	0.891

Table 3 Values of the viscosity of the mixtures of monoalcohols (1-propanol and 2-propanol) and water (increase and decrease) for the indicated values of Φ ($n=3$)

1-propanol			2-propanol		
Φ	Viscosity (η , mPa·s) (Inc)	Viscosity (η , mPa·s) (Dec)	Φ	Viscosity (η , mPa·s) (Inc)	Viscosity (η , mPa·s) (Dec)
0.0000	1,946	–	0.0000	2.031	–
0.3704	2.227	–	0.3704	2.358	–
0.8332	2.421	–	0.8332	2.594	–
1.4284	2.531	–	1.4284	2.750	–
2.2227	2.597	–	2.2227	2.837	–
3.0000	2.62	2.624	3.000	2.886	2.886
3.3328	–	2.591	3.3328	–	2.847
4.9990	–	2.436	4.999	–	2.660
7.780	–	2.208	7.780	–	2.389
13.36	–	1.848	13.36	–	1.964
29.99	–	1.258	29.99	–	1.286
100	–	0.891	100	–	0.891

In Tables 2 and 3, it can be observed that the viscosity increases up to a value of $\Phi=3$, at which point the maximum value of viscosity is reached. After this point, there is a rapid decrease in viscosity for increasing values of Φ , with an asymptotic tendency towards the value of the viscosity of water. The discontinuity that appears at $\Phi=3$, which is the point at which the water molecules stop forming hydrogen bonds with the alcohols, leads to the proposal of a function for the growing phase (for values of Φ less than 3) and a different function for the decreasing phase (for values of Φ greater than 3).

The proposed functions for the viscosity of the mixture are as follows:

(a) For the increase phase:

$$\eta = (\eta_{\max} - \eta_{\min}) \left(1 - e^{(-m_1 \cdot \phi)} \right) + \eta_{\min}, \quad (2)$$

where η_{\max} is the maximum value of the viscosity reached by the mixture at the point $\Phi=3$ and η_{\min} is the viscosity of the solute, i.e. the corresponding alcohol. This function is only applicable for the interval of Φ between 0 and 3, and at $\Phi=0$ the viscosity corresponds to that of the solute.

(b) For the decrease phase

$$\eta = (\eta_{\max} - \eta_{\min}) \cdot e^{(-m_2 \cdot (\phi-3))} + \eta_{\min}. \quad (3)$$

In this case, the value of η_{\min} is still the maximum value of the mixture reached for $\Phi=3$, but η_{\min} is now the viscosity of the solvent, which is water in this case.

The results, along with their corresponding graphs, are presented in Fig. 2 for both the ascending and descending phases. It can be observed that there is an excellent correlation with the use of a single parameter in both cases (Table 3).

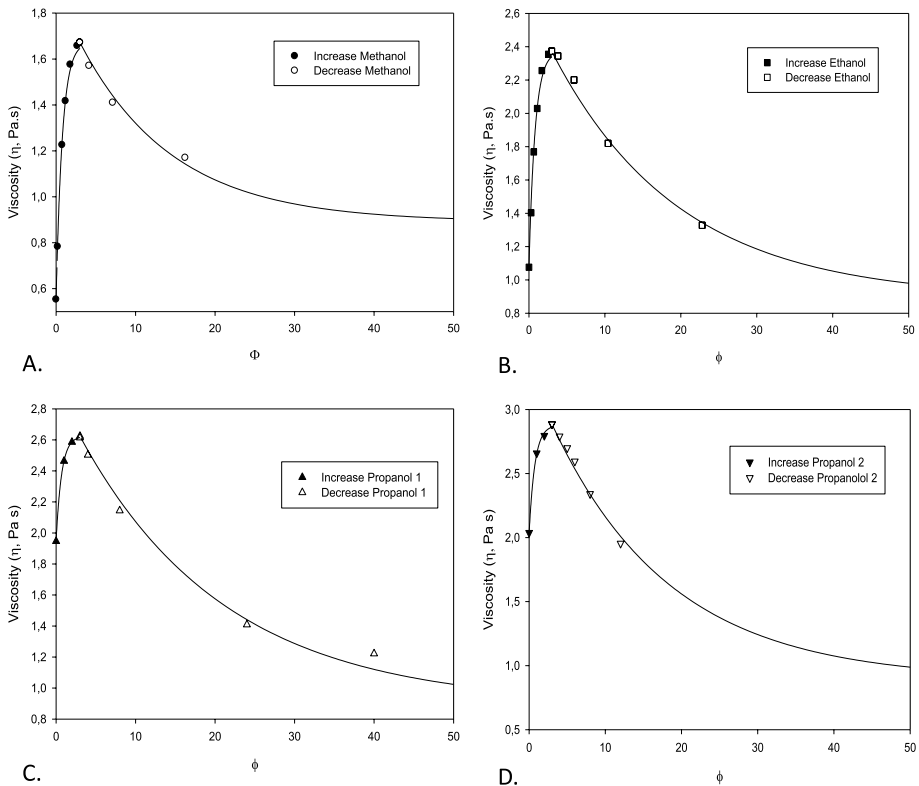


Fig. 2 Graphical representation of the viscosities (increase and decrease) against the values of Φ for methanol (A), ethanol (B), 1-propanol (C), and 2-propanol (D)

Table 4 Values for the parameters obtained: m_1 (increasing phase) and m_2 (decreasing phase) and the correlation coefficients (R)

Monoalcohols	Increase Phase		Decrease Phase	
	$m_1 (\pm \text{SD})$	R >	$m_2 (\pm \text{SD})$	R >
Methanol	1.26 ± 0.07	0.999	0.085 ± 0.005	0.997
Ethanol	1.23 ± 0.10	0.997	0.059 ± 0.003	0.998
1-Propanol	1.45 ± 0.06	0.999	0.055 ± 0.004	0.996
2-Propanol	1.32 ± 0.16	0.998	0.064 ± 0.003	0.998

Table 5 Viscosity values of mixtures of polyalcohols and water with different proportions for the indicated values of Φ ($n=3$)

Ethylene glycol		Glycerin		Sorbitol	
Φ	Viscosity (η , mPa·s)	Φ	Viscosity (η , mPa·s)	Φ	Viscosity (η , mPa·s)
0.0000	15.271	0.0000	938.000	–	–
0.4840	9.920	0.5679	284.350	–	–
0.9677	6.610	1.2780	46.521	–	–
1.9350	4.600	2.1900	21.754	–	–
2.9030	3.480	3.4100	9.630	–	–
4.1520	2.770	5.1110	5.891	4.33	209.000
4.8390	2.350	7.6670	3.457	4.51	150.000
5.8070	2.120	11.93	2.472	4.79	100.000
7.2420	1.870	20.44	1.638	5.71	53.400
7.7400	1.780	46	1.213	6.66	41.300
9.6810	1.600	80	0.892	8.34	28.100
11.61	1.500	100	0.891	14.54	13.700
23.21	1.170			18.02	7.300
48.00	0.993			24.54	4.018
100	0.891			100	0.891

In all cases the dispersion of the viscosity measurements was less than 2%. The average of the three measurements was taken as the final value

The values of the parameters resulting from the fitting of the viscosity as a function of Φ for the four monoalcohols are indicated in Table 4, with an excellent correlation coefficient ($R > 0.995$).

In a similar manner to the method used for the monoalcohols, Table 5 presents the experimental values of the viscosities (mPa·s) of polyalcohols: ethylene glycol, glycerin, and sorbitol 70. The measurements were taken using a similar protocol as the one used for the monoalcohols, and the dispersion of the measurements was less than 2%. The average of the three measurements was taken as the final value. The table shows the viscosity values for different proportions of polyalcohols and water, with the corresponding values of the mole ratio (Φ).

In the case of mixtures of ethylene glycol, glycerin, and sorbitol with water, there is a physical–chemical interaction that leads to a rapid decrease in the viscosity of the mixture as the amount of water increases. This necessitated the introduction of a correction

factor, and after testing various equations, the best results were obtained with the following equation:

$$\eta = \eta_{\min} + (\eta_{\max} - \eta_{\min}) \cdot e^{(-m_1 \cdot \phi^{m_2})}. \quad (4)$$

Due to the rapid decrease in the viscosity (Table 4) for ethylene glycol, glycerine, and sorbitol mixture when adding water, it has been necessary to introduce a new coefficient (m_2) as an exponent of Φ to achieve good correlations (Eq. 4). In this case, the value of maximum viscosity (η_{\max}) is the value of the viscosity of the pure solute (i.e. ethylene glycol, glycerine, and sorbitol 70), and the minimum viscosity of the pure water (η_{\min}) is 0.891 mPa.s.

As can be seen in Fig. 3, the viscosity values decrease rapidly as a function of the mole ratio (Φ) for the different polyalcohols tested. This decrease in viscosity is more pronounced in the case of glycerin compared to ethylene glycol.

In the case of sorbitol, the commercially available preparation has a concentration of 70%. It is evident that the viscosity of pure sorbitol (100%) cannot be measured as this compound is solid. As shown in Fig. 4, the continuous line represents the theoretical values obtained by fitting Eq. 4, while the experimental viscosity values are shown with black dots between 0 and sorbitol 70% (Table 5).

In order to compare the viscosity profiles between the different polyalcohols, the sorbitol was fitted to Eq. 4, by assigning different hypothetical viscosity values (η_{\max}) for the mole ratio $\Phi=0$. The fit to this equation shows that the best result was obtained with the values of $\eta_{\max} = 1400$ mPa.s for the hypothetical sorbitol (100%), with an excellent correlation coefficient value ($R=0.9998$), as can be seen in Table 6.

4 Conclusions

To represent the viscosity of binary mixtures of water with different alcohols, a new variable, Φ , was introduced. It is defined as the ratio between the moles of water and the moles of the corresponding alcohol. This representation provides a function, $\eta=f(\Phi)$, that has

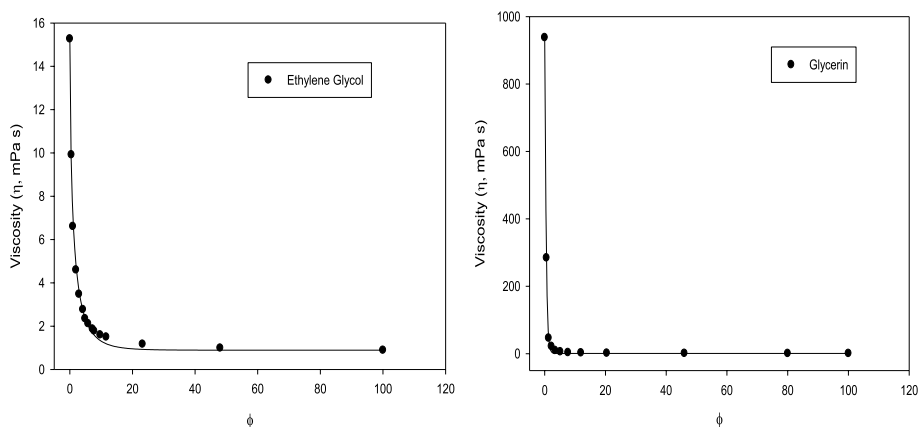


Fig. 3 Graphical representation of the experimental viscosities (η , mPa.s) vs. the values of Φ for ethylene glycol and glycerin (Table 5)

Fig. 4 Graphical representation of the experimental values of viscosities (η , mPa·s) against the values of Φ for the sorbitol 70 (- • -)

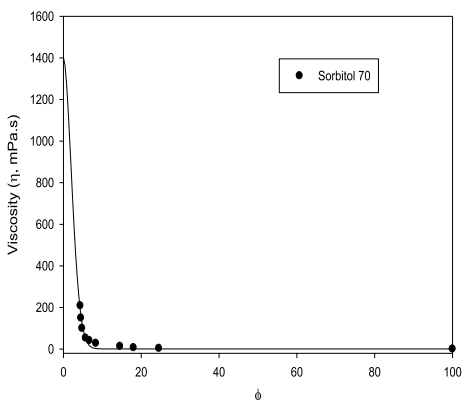


Table 6 Values of m_1 , m_2 and their respective correlation coefficients ($R >$) corresponding to Eq. 4, for the aqueous mixtures of the indicated polyalcohols; ethylene glycol, glycerin, sorbitol 70, and sorbitol

Polyalcohols	$m_1 (\pm \text{SD})$	$m_2 (\pm \text{SD})$	$R >$
Ethylene Glycol	0.85 ± 0.04	0.62 ± 0.03	0.997
Glycerin	2.23 ± 0.07	1.10 ± 0.06	0.999
Sorbitol 70	0.04 ± 0.03	2.4 ± 0.5	0.994
Sorbitol*	0.137 ± 0.006	1.8 ± 0.3	0.999

* Hypothetic fitted values of the sorbitol between 0 and 100%

an exponential nature, with which excellent correlation coefficients are achieved with only one coefficient for monoalcohols for the increase (Eq. 2) and decrease phase (Eq. 3). In the case of polyalcohols, the viscosity has a strictly monotonous variation, and Eq. 4 has two parameters which allows the adjustment of the viscosity values in the entire range of concentrations. Additionally, the variable Φ highlights the possible critical points where physicochemical interactions occur between the components of the mixture.

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Declarations

Competing interests The authors declare no competing interests.

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