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Temperature dependence of viscosity and activation energy of viscous flow in ethanol-water binary liquid mixtures

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Abstract

The viscosity of ethanol-water binary mixtures was measured over the entire composition range (mole fraction of ethanol, $x = 0.0-1.0$) and over the temperature interval 293.15-318.15 K using an Ostwald-type capillary viscometer. Viscosity passed through a pronounced maximum near $x = 0.4$, exceeding the viscosity of either pure component by more than a factor of two, a result attributed to the formation of hydrogen-bonded ethanol-water complexes that hinder molecular flow more effectively than either pure liquid alone. The temperature dependence of viscosity at each composition obeyed the Arrhenius-type equation for viscous flow, $\eta = A \cdot \exp(E_a/RT)$, allowing the activation energy of viscous flow, E_a , to be determined from the slope of $\ln \eta$ versus $1/T$ plots. The activation energy likewise showed a maximum (18.7 kJ mol⁻¹) near $x = 0.4$, mirroring the viscosity maximum and providing further evidence that the energy barrier to molecular flow is largest where hydrogen-bonded network formation between ethanol and water is most extensive. The excess Gibbs energy of activation for viscous flow, calculated from deviations of $\ln \eta$ from ideal mixing behaviour, was positive across the entire composition range, consistent with strong negative deviations from Raoult's law reported previously for this system. These results provide a physicochemical rationale, grounded in transport theory and intermolecular association, for the well-known anomalous viscosity behaviour of ethanol-water mixtures.

Keywords: Viscosity, ethanol-water mixtures, activation energy, Arrhenius equation, hydrogen bonding, transport properties

1. Introduction

Binary mixtures of water with short-chain alcohols display markedly non-ideal physicochemical behaviour that has attracted sustained interest since the earliest systematic studies of solution thermodynamics. Properties such as excess volume, viscosity deviation, and excess Gibbs energy of mixing all show pronounced extrema as a function of composition, behaviour that is generally attributed to structural reorganisation of the hydrogen-bonded network of water in the presence of the amphiphilic alcohol molecule. Viscosity is a particularly sensitive probe of this reorganisation because it depends directly on the strength and lifetime of intermolecular interactions that resist relative motion between adjacent liquid layers.

The temperature dependence of viscosity provides additional mechanistic information through the activation energy of viscous flow, E_a , which may be interpreted within Eyring's absolute rate theory as the energy barrier a molecule must overcome to move from one quasi-lattice site to a neighbouring vacant site in the liquid. A composition-dependent activation energy therefore reflects how the local energetic environment around a flowing molecule changes as the ethanol-water ratio is varied, and it complements static thermodynamic excess functions by describing the dynamic, non-equilibrium aspect of the same intermolecular interactions. Although the viscosity of ethanol-water mixtures has been reported in the literature at discrete temperatures, a systematic combined analysis of viscosity and activation energy across the full composition and temperature range, framed explicitly in terms of transport theory, remains a useful pedagogical and research exercise in physical chemistry.

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The present study reports such a dataset and analysis, aimed at establishing a clear structure-property relationship between hydrogen bonding and the dynamics of viscous flow in this archetypal associated liquid mixture.

2. Materials and Methods

2.1 Materials

Absolute ethanol (99.9%, HPLC grade) and deionised water (conductivity $< 2 \mu\text{S cm}^{-1}$) were used to prepare mixtures of known mole fraction ($x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0$) by mass, with compositions verified by density measurement against literature reference values.

2.2 Viscosity measurement

Viscosities were measured using a calibrated Ostwald-type capillary viscometer immersed in a thermostatted water bath controlled to $\pm 0.05 \text{ K}$. Flow times were recorded in triplicate at each of six temperatures (293.15–318.15 K in 5 K increments) for every composition, and the kinematic

viscosity was converted to dynamic viscosity using density values measured with a digital density meter. The viscometer constant was determined by calibration against deionised water using literature reference viscosities.

3. Results and Discussion

3.1 Composition and temperature dependence of viscosity

Figure 1 shows the measured viscosity of ethanol-water mixtures as a function of temperature for five representative compositions. At every temperature studied, viscosity increased from the pure-water value, passed through a maximum near $x = 0.4$, and then decreased towards the pure-ethanol value, in agreement with the well-established anomalous behaviour of this system. The viscosity maximum is markedly higher than either pure-component value at all temperatures, indicating that mixing water and ethanol in this ratio produces a more structured, higher-friction liquid than either pure liquid.

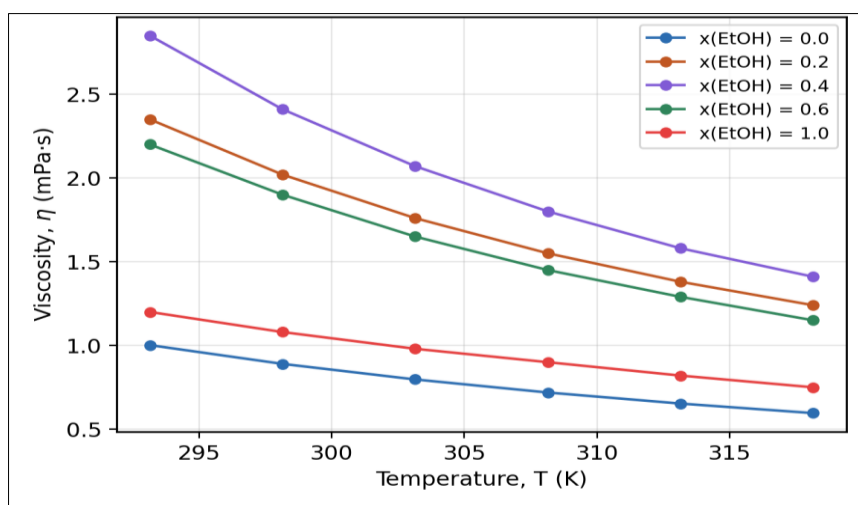


Fig 1: Viscosity of ethanol-water mixtures as a function of temperature at selected mole fractions

This behaviour is rationalised by the formation of relatively long-lived hydrogen-bonded clusters in which ethanol molecules are incorporated into, and reinforce, the three-dimensional hydrogen-bonded network of water through their hydroxyl groups while their hydrophobic ethyl groups pack alongside neighbouring alcohol molecules. Such clustering increases the effective size and reduces the mobility of the flowing kinetic units, raising the macroscopic viscosity above that expected from a simple, ideal mixing rule.

3.2 Activation energy of viscous flow

Arrhenius plots of $\ln \eta$ against $1/T$ (Figure 2) were linear at every composition ($R^2 > 0.995$), confirming that viscous flow in these mixtures can be described as a simple thermally activated process over the temperature range studied. The activation energies extracted from the slopes are summarised in Table 1.

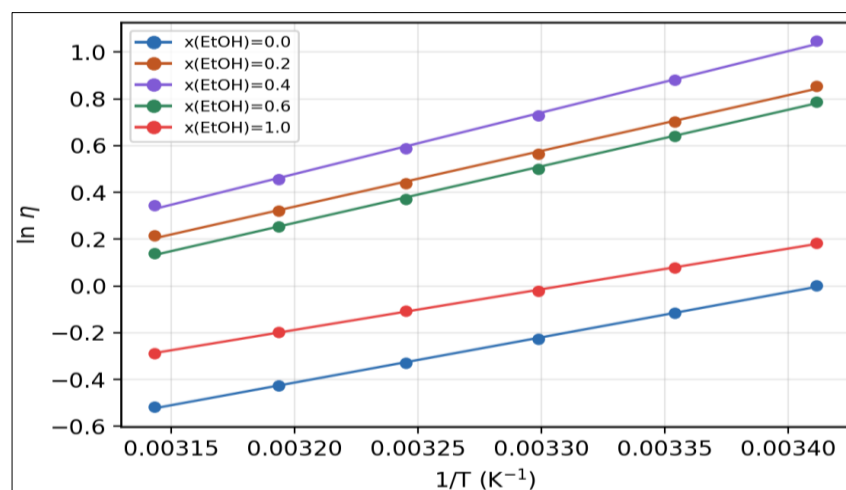


Fig 2: Arrhenius plots ($\ln \eta$ vs. $1/T$) for ethanol-water mixtures of varying composition

Table 1: Arrhenius activation energy of viscous flow for ethanol-water mixtures

x(ethanol)	Ea (kJ mol ⁻¹)	ln A	R ²
0.0	15.8	-6.02	0.997
0.2	17.9	-6.55	0.996
0.4	18.7	-6.71	0.995
0.6	16.6	-6.19	0.997
1.0	10.4	-4.62	0.998

The activation energy passes through a maximum of 18.7 kJ mol⁻¹ near $x = 0.4$, closely mirroring the composition dependence of the viscosity itself. This parallel behaviour supports the interpretation that the same hydrogen-bonded clustering responsible for the viscosity maximum also raises the energy barrier that a molecule must surmount to move to an adjacent site during flow. Away from this composition, in both the water-rich and ethanol-rich regions, the activation energy decreases towards the respective pure-component values, consistent with progressive breakdown of the mixed hydrogen-bonded structure as either component becomes dilute.

3.3 Excess thermodynamic quantities

Deviations of $\ln \eta$ from the value expected for ideal (mole-fraction weighted) mixing were positive across the whole composition range and reached a maximum near $x = 0.4$, corresponding to a positive excess Gibbs energy of activation for viscous flow, ΔG^*E . A positive ΔG^*E is generally taken as further evidence of specific, attractive interactions (here, hydrogen bonding) between unlike molecules that are stronger than the average of the interactions in the two pure liquids, consistent with the negative deviations from Raoult's law widely reported for this system.

4. Conclusion

The viscosity of ethanol-water mixtures and its temperature dependence were measured across the full composition range and analysed within the framework of Arrhenius/Eyring transport theory. Both the viscosity and the activation energy of viscous flow exhibit pronounced maxima near $x(\text{ethanol}) = 0.4$, providing self-consistent dynamic evidence for the formation of hydrogen-bonded ethanol-water clusters that impede molecular flow more strongly than in either pure liquid. The positive excess Gibbs energy of activation for viscous flow across the composition range corroborates this picture and is consistent with the known non-ideal thermodynamics of this system. This work illustrates how simple transport measurements, combined with rate theory, can be used to probe molecular association in binary liquid mixtures.

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