

Intermolecular force range and noncritical shear viscosity of the critical binary mixture of benzene - coconut oil

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ABSTRACT

Shear viscosity coefficients of benzene and coconut oil binary mixture have been measured using digital viscometer. The measured values were taken over the entire concentration range above the critical temperature. The results above the critical temperature were analyzed by the mode coupling theory. The anomaly of shear viscosity was detected as a function of temperature and concentration. The value of noncritical part of shear viscosity near the critical temperature was measured to be $\eta_0 = 1.30\text{cP}$. The Debye momentum cutoff is found to be 0.358 , the constant $A = 0.052 \pm 0.003$, the correlation length $\zeta' = 1.02\text{\AA}$, and the intermolecular force range $L = 9.90\text{\AA}$ by using the mode coupling approach. The forces are weak because of the small value of the viscosity.

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INTRODUCTION

Binary mixture is a mixture that contains two constituents; it has definition at a critical temperature and critical concentration, where the mixture mixes homogeneously. The two components do not mix homogeneously in all proportions below this temperature, and can define two different phases^[1,2].

The viscosity of a fluid is a property which measure of its resistance to gradual deformation by shear stress or tensile stress. For liquids, it corresponds to the informal notion of "thickness"^[3]. Viscosity is due to friction between neighboring particles of the fluid that are moving at different velocities. When fluid is forced through a tube, the fluid generally moves faster near the axis and very slowly near the walls^[4]. Experimental data of shear viscosity are important for the scientists whom working on the improvement and development of molecular theories. Viscosity is affected by different fac-

tors such as temperature, shear rate, catalyst, pressure, and molecular weight concentration^[4,5].

The viscosity in binary mixtures was studied and the theory is given of the sharp viscosity rise in mixtures in the critical mixing region. Marshall Fixman found good agreement with the experimental dependence of viscosity on composition and temperature^[6-8]. Katti and Chaudhri measured viscosities of binary mixtures of Benzyl Acetate - Dioxane, Aniline - m-cresol^[9]. Teja and Rice measured Viscosities of binary liquid mixtures of *n*-hexane - benzene, toluene - ethylbenzene at 298.15, 308.15 and 323.15K using Cannon Fenske viscometers^[10].

The existence of shear viscosity anomaly has been experimentally observed in some binary liquid systems near the critical temperature and concentration^[11-21]. D'Arrigo and his group measured the shear viscosity as a function of temperature and molar composition near the consolute point of aniline-cyclohexane binary mix-

ture^[22]. Nieuwoudt and Sengers made a reevaluation for the critical exponent of viscosity for binary mixtures near consolute point^[23].

Abdelraziq measured the shear viscosity coefficient of nitrothane - 3-methelpentane binary mixture^[20]. Domanska and Krolikowska determined the densities and viscosity correlations for five different binary mixtures of the ionic liquids^[24]. The tracer diffusion coefficients of some binary mixtures like tritiated water and acetonitrile in water - acetonitrile have been determined at several temperatures^[25].

The mode coupling theories have been applied in pure fluids and binary mixtures to explain the concentration and temperature dependence of shear viscosity of the critical mixture by Swinney and Donald^[26]. Gotze reached to essential results of the mode-coupling theory for the evolution of structural relaxation in simple liquids such as the Debye-Waller-factor anomaly, and their importance to the understanding of experiments in glass-forming systems^[27]. Kob and Anderson tested the mode coupling theory for super cooled binary mixture (Lennard-Jones) and they calculated Van Hove correlation function^[28].

The main goals of this study are to measure the critical temperature and critical concentration of benzene - coconut oil binary mixture, and determine the constants of benzene - coconut oil binary mixture using the mode coupling theory.

THEORY

The viscosity of a fluid is a measure of its resistance to gradual deformation by shear stress or tensile stress. For liquids, it corresponds to the informal notion of “thickness”. Viscosity is due to friction between neighboring particles of the fluid that are moving at different velocities^[3].

The viscosity of pure liquids affects on temperature, pressure, density, and surface tension. Dynamic viscosity of liquids decreases with increasing temperature, and it increases with increasing pressure^[29].

The relationship that explains the relation between the dynamic viscosity and the temperature is in the form of an Arrhenius-type equation^[30].

$$\eta = \eta_0 e^{\frac{E_\eta}{RT}} \quad (1)$$

Where η is the measured dynamic shear viscosity, η' is the dynamic shear viscosity at some reference temperature, is the temperature coefficient for viscosity, R is the universal gas constant, and T is the absolute temperature (K).

Liquids have complex nature, so there is more than one theory on the viscosity, like the theoretical methods of calculating liquid viscosities, the molecular dynamic approaches, semi-empirical and empirical methods.

At temperatures below the normal boiling point, the logarithm of liquid viscosity varies linearly with the reciprocal of the absolute temperature T is described by the empirical model;

$$\ln(\eta) = \dot{A} + \frac{B}{T} \quad (2)$$

Where A and B are constants determined empirically.

At temperatures above the normal boiling point, the $\ln(\eta)$ versus relationship becomes nonlinear and is described by a number of semi-empirical methods.

Viscosities of solutions (homogeneous products of dissolution of solids or gases in liquids) and fluid mixtures (homogeneous mixtures resulting out of mixing of two or more fluids) studied by several methods.

Some of the methods developed using excess Gibbs free energy models include Wilson, non-random two-liquid (NRTL), and universal quasi-chemical (UNIQUAC)^[29].

The mode coupling theory is used to study the critical anomaly of the shear viscosity and the coefficients.

In the term mode coupling, the word “mode” refers to eigenmodes of an idealized, “unperturbed”, linear system. The superposition principle says that eigenmodes of linear systems are independent of each other. In most real systems, there is at least some perturbation that causes energy transfer between different modes. This perturbation is interpreted as an interaction between the modes. It is called “mode coupling” theory^[31].

The mode coupling theory is used in important applications as fiber optics, lasers, and condensed-matter physics, to describe the critical behavior slowing

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down.

The mode coupling theories were studied by Kawasaki in 1970, by Mistura in 1971 and by Perl and Ferrell in 1972^{[32],[33],[34]}. Reichman and Charbonneau reviewed the mode-coupling theory of the glass transition, and derived mode coupling equations for the description of density fluctuations from microscopic considerations. They derived schematic mode-coupling equations from a field-theoretic perspective. They reviewed the mode coupling theory successes and failures^[35].

The critical anomaly of the shear viscosity coefficient by Kawasaki and Perl and Ferrell is given according to the law^[34]:

$$\frac{\eta - \eta_0}{\eta} = \frac{\Delta\eta}{\eta} = A \ln \zeta + A \ln q_D \quad (3)$$

Where η_0 is the noncritical part of the measured shear viscosity, A is constant = $\frac{8}{15\pi^2} = 0.054$ which was calculated theoretically by D'Arrigo^[22], ζ is the correlation length, and q_D is Debye momentum cutoff.

The dynamic shear viscosity is also temperature dependent at the critical concentration which is given by the power law

$$\eta = \eta_0 \tau^{-x_\eta v} \quad (4)$$

Where τ is the reduced temperature, which is given by $\frac{T - T_c}{T_c}$, v is critical exponent = 0.64^{[22],[36]}, $x_\eta v$ is critical exponent = 0.037.

The correlation length is defined as estimate of the scale at which the power law gives to a flat spectrum at law wave number^[37]. The correlation length is the average distance between two molecules at temperatures near the critical temperature.

The correlation length expression is given by

$$\zeta = \zeta_0 \tau^{-v} \quad (5)$$

This is related to the reduced temperature τ near the critical temperature T_c and critical concentration x_c .

Intermolecular forces are the forces that act between stable molecules or between functional groups of macromolecules. Sometimes they are referred to as Van der Waals forces. The intermolecular force range is the reciprocal of the correlation length ζ at $\eta = \eta_0$ ^[38].

The intermolecular force range L (Debye parameter) can be calculated from light-scattering experiment by the relation^[36].

$$L = 2.45 \zeta_0 T_c^{\frac{\gamma-1}{2}} \quad (6)$$

Where d is the ratio between specific heat at constant pressure and specific heat at constant volume, $\gamma = 1.25$ ^[36].

The two – scale – factor universality has been used in modern theories to explain the critical phenomena of binary liquid mixtures by predict R_ζ ^[39]. This quantity is defined as

$$R_{\zeta=\zeta_0} = \left[\alpha \frac{\rho_c C_{pc}}{k_B} \right]^{\frac{1}{d}} = \zeta_0 \left[\alpha \frac{T_c \alpha_{pc}}{k_B T_c} \right]^{\frac{1}{d}} \quad (7)$$

Where R_ζ is the universal quantity and it is equal 0.270^[39], α is the critical exponent and it is equal 0.11, ρ_c is the mass density at the critical temperature, C_{pc} is the critical amplitude of the specific heat, k_B is the Boltzmann's constant, T_c is the critical temperature, α_{pc} is the critical amplitude of the thermal expansion, and T_c is the pressure derivation of the critical temperature along the critical line^[40].

The specific heat at constant pressure is c_p is given by

$$c_p = c_{pc} \tau^{-\alpha} + c_{pb} \quad (8)$$

Where c_{pc} is the critical amplitude of specific heat at constant pressure, and c_{pb} is the critical background of specific heat at constant pressure.

METHODOLOGY

High-purified samples of benzene and coconut oil were used, in this work. The shear viscosities from 18 to 42 °C temperature range were measured for benzene - coconut oil binary mixture, and over the entire composition range. The concentration and temperature dependence of shear viscosity were measured. Each measured datum of the viscosity of benzene - coconut oil samples represents the average of two measurements.

Experimental apparatus

The instrument that was used to measure the viscosity of the critical mixture of benzene and coconut oil is Brookfield Viscometer Model DV-I+, Digital Prima Long Thermometer is used to measure the temperature; its accuracy is $\pm 1\%$ with measures temperature range is from -20°C to $+100^{\circ}\text{C}$, and Fried Electric model WB-23 was used to measured and control the temperature of the samples to a specific temperature.

Statistical analysis

The obtained results were tabulated and statistically analyzed. The statistical analysis of the data was done by using the statistical program (Microsoft Office Excel). Coefficient of determination R^2 was calculated as a measure of the strength of the correlation between viscosities of the binary mixture with the temperature.

RESULTS AND DISCUSSION

The critical temperature and critical concentration

The dynamic viscosity values of benzene - coconut oil samples of different concentrations (0%-100%) by weight of benzene were measured at temperature range (18-42°C). The dynamic viscosity of benzene - coconut oil of different concentrations (0%-80%) benzene as a function of temperature are plotted in Figure (1) – (2).

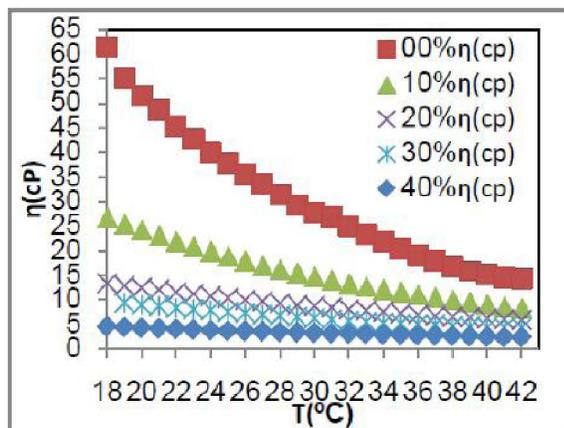


Figure 1: The dynamic viscosity of benzene – coconut oil as a function of temperature at concentrations (00%-40%) by weight of benzene.

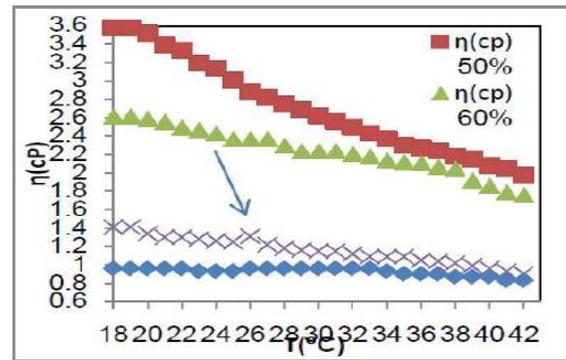


Figure 2: The dynamic viscosity of benzene – coconut oil as a function of temperature at concentrations (50%-80%) by weight of benzene.

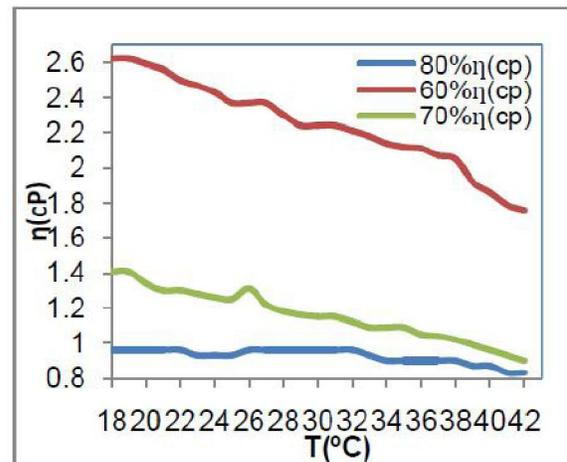


Figure 3: Dynamic viscosity at different temperatures for 60%, 70%, and 80% by weight of benzene.

The anomaly can be noticed by plotting 60%, 70%, and 80% by weight of benzene as shown in Figure (3).

Figure (2) and (3) show a cusp at $x_c = 0.700$ by weight of benzene and $T_c = 26.00^{\circ}\text{C}$. That point is considered to be a critical point where the two liquids become one phase.

The critical point of the binary mixture benzene - coconut oil was determined by Battacharya and Deo where, their critical temperature is 25.40°C and the critical concentration x_c is 0.6037 by weight of benzene^[41]. This will affect the value of the non-critical part of the shear viscosity η_0 .

The noncritical part shear viscosity η_0

The dynamic shear viscosity is temperature dependent at the critical concentration which is given by the power law. The data of dynamic shear viscosity are fitted using the power law $\eta = \eta_0 \tau^{-x_\eta} v$, where x_η and v are critical exponents. $\ln(\eta)$ is plotted against $\ln(\tau)$ to

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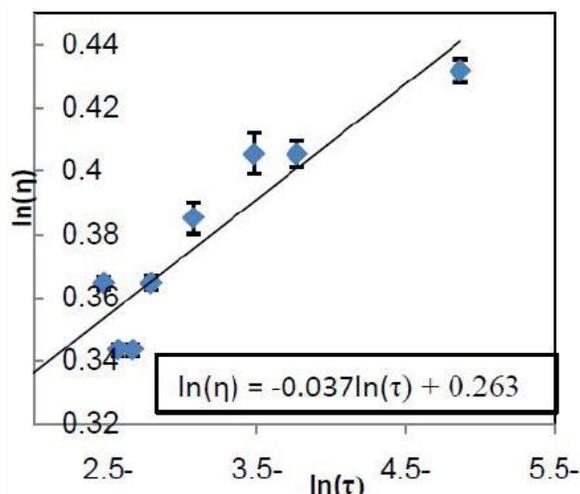


Figure 4 : Ln(η) vs. Ln(τ), using power law $\eta = \eta'$

find the value of the critical exponent as shown in Figure (4).

Figure (4) shows that the values of the shear viscosity fit the power law

$$\eta = \eta_0 \tau^{-x_{\eta}^v}$$

$$\ln(\eta) = \ln(\eta_0) - x_{\eta}^v \ln(\tau)$$

The constant x_{η}^v is the slope which equals to 0.037 ± 0.003 it is in good agreement with the critical exponent x_{η}^v of D'Arrigo and Klein and Woermann which equal 0.037 ± 0.003 [22],[36].

The intercept equals $\ln(\eta_0) = 0.263$. The non-critical part of the measured shear viscosity $\eta_0 = 1.30\text{cP}$; η_0 was found by Abdelraziq to be 2.76cP [1]. The noncritical part of the measured shear viscosity η_0 is small compared to Abdelraziq's result because of the different in values of critical temperature and critical concentration.

Calculation of the correlation length ζ'

Two – scale – factor universality is used to find the correlation length ζ' of the benzene-coconut oil binary liquid mixture Eq.(7).

$$R_{\zeta} = \zeta \left[\frac{\rho_c C_{p,c}}{k_B} \right]^{\frac{1}{d}} = \zeta \left[\frac{T_c \alpha_{p,c}}{k_B T_c} \right]^{\frac{1}{d}} = 0.270$$

Where critical exponent $\alpha = 0.11$, dimension $d = 3$, mass density at the critical temperature $\rho_c = 0.878\text{cm}^3$ which found in this work, where Boltzmann's constant

is given by $= 1.38 \times 10^{-23} \frac{\text{J}}{\text{K}}$.

The correlation length ζ_0 for benzene-coconut oil binary liquid mixture is calculated and it is equal 1.98\AA . The correlation length ζ' for benzene-coconut oil binary liquid mixture was calculated at 25.4°C by Abdelraziq and it is equal 1.02\AA [1].

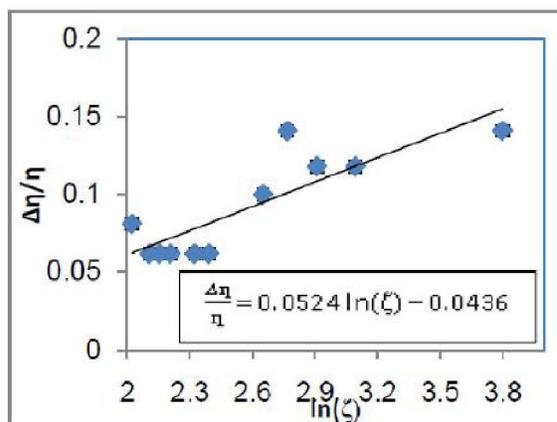
The value of correlation length ζ_0 is deferent than Abdelraziq's value may be because of the deferent in the critical temperature and the critical concentration. In this work T_c and x_c are given by $(26.00^\circ\text{C}, 0.7000)$ by weight of benzene) respectively. While in Abdelraziq's work were $(25.40^\circ\text{C}, 0.6037)$ by weight of benzene) respectively.

The intermolecular force range l (debye parameter)

The intermolecular force range L (Debye parameter) is calculated by using Eq. (2.9), Where ζ' equals to 1.98\AA , d is the ratio between specific heat at constant pressure and specific heat at constant volume, $\gamma = 1.25$ [36], and the critical temperature T_c is 26.00°C .

The intermolecular force range L (Debye parameter) equals 9.90\AA . Determining the intermolecular force range L using benzene – coconut oil system for the first time supports the mode-coupling theory of the anomalous viscosity. The value L relatively large because ζ_0 and T_c values are large. The value $L = 9.90\text{\AA}$ gives an indication that the forces between the components of the benzene – coconut oil binary mixture is

Figure 5 : Versus the logarithm of the average correlation length ln ζ.



weak attractive force.

Calculation of the constant A

The constant A of the mode coupling theory has a theoretical value 0.054^[22]. The value of A can be determined by using Eq. (2.5), Where η_0 is the noncritical part shear visc $\frac{\Delta\eta}{\eta}$ which is calculated in section (4.2) to be 1.30 cP, η is the relative anomalous dynamic shear viscosity, ζ is the average correlation length of the concentration fluctuations which is give $\frac{\Delta\eta}{\eta}$ Eq. (2.8). The relative anomalous shear viscosity η versus the logarithm of the average correlation length $\ln \zeta$ is plotted in Figure (5) at the critical concentration and above the critical temperature.

Figure (5) shows that the relation between η and $\ln \zeta$ is a linear relation with the slope equals to A value, which is 0.052 ± 0.003 . agreement with the literature values^{[26],[22],[20],[17]}.

The value of the constant A for different binary mixtures was measured or calculated for example; Swinney and Henry, D'Arrigo and his group, and Abdelraziq and his group obtained the constant A experimentally to be 0.054, 0.056, 0.054 ± 0.002 and 0.058, respectively. Our value of A is in good agreement with the literature values^{[26],[22],[20],[17]}.

Determination of the debye momentum cutoff

The Debye Momentum Cutoff can be found from the intercept of Figure (4.7) and the value of the constant A. Using Eq.(2.6), the value of is 0.358 \AA^{-1} .

Determining q_D of the binary liquid mixture benzene - coconut oil is important for the scientists working on the improvement of molecular theories, and supports the mode coupling theory of the critical viscosity.

CONCLOUION AND FUTURE WORK

The mode-coupling theory of the anomalous dynamic shear viscosity near the critical point has been studied using benzene – coconut oil binary liquid mixture.

The critical temperature and critical concentration have been measured of benzene - coconut oil binary mixture, the noncritical part of shear viscosity has been obtained, the correlation length ζ_0 has been determined to be 1.98 \AA , the intermolecular force range L (Debye parameter) has been obtained to be 9.90 \AA .

the constant A has been found and its value is consistent with the literature values. Finally, the Debye momentum cutoff q_D is determined to be 0.358 \AA^{-1} .

The experimental values of L, q_D and A constants are determined to the first time, which support the data obtained by theoretical calculations using the mode coupling approach.

Intermolecular forces are defined as the attractive forces between molecules, if the viscosity is high; the Intermolecular attractive force is high. The value of the intermolecular force range L (Debye parameter) has been obtained to be 9.90 \AA ; it is large distance and gives an indication that the force between the components of the benzene – coconut oil binary mixture is weak attractive force compared to CCl_4 -coconut oil and others. The dynamic viscosity η_0 of the binary mixture is also smaller than the dynamic viscosity η_0 of CCl_4 - coconut oil binary mixture.

As a future work, there are a lot of binary mixtures that can be studied using the mode coupling approach to find the unknown parameters, and other kinetic coefficients can be studied near the critical point like; heat capacity, refractive index, susceptibility and thermal expansion coefficient, for the binary mixture benzene - coconut oil binary mixture and other binary mixtures.

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