An-Najah National University
Faculty of Graduate Studies

Concentration and Temperature Dependence of Viscosity in Mode-Coupling Theory of Binary Mixture of Water and Phenol

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Dedication

To my parents, my husband and my daughter with respect and love

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الإقرار

أنا الموقعة أدناه مقدمة الرسالة التي تحمل العنوان:

Concentration and Temperature Dependence of Viscosity in Mode-Coupling Theory of Binary Mixture of Water and Phenol

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The work provided in this thesis, unless otherwise referenced, is the researcher's own work, and has not been submitted elsewhere for any other degree or qualification.

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List of Abbreviations

T_c	Critical temperature
x_c	Critical concentration
q_D	Debye momentum cutoff
η_0	Noncritical part of viscosity
ξ_0	Correlation length
L	Intermolecular force
g	Adiabatic coupling constant
α_p	Isobaric thermal expansion coefficient
$\alpha_\lambda(crit)$	Attenuation per wavelength at critical concentration
D	Diffusion coefficient
β	Adiabatic compressibility
V	Free volume
L_f	Free length
ρ	Mass density
V^E	Excess molar volume
ΔB_s	Deviation in isentropic compressibility
α	Critical exponent
X_A	Mole fraction
VFT	Vogel-Fulcher-Tammann
η	Dynamic viscosity
F	External force
H	The distance separating between the plates
$\frac{v}{h}$	Shear of rate deformation (shear velocity)
$\frac{dv}{dh}$	Velocity gradient or deformation rate
$\tau = \frac{F}{A}$	Shear stress
ν	Kinematic viscosity
E_a	Activation energy
T	Absolute temperature
R	Gas constant
MCT	Mode coupling theory
A	Constant equals to 0.054

$X_\eta \nu$	Critical exponent equals to 0.04
T	Reduce temperature
γ	Critical exponent equals to be 1.25
RGT	Renormalization group theory
α_λ	The sound attenuation coefficient per wavelength
$u(\omega)$	Velocity of sound wave
H	Critical amplitude
$f(\omega^*)$	Scaling function
ω^*	Reduced frequency
DST	Dynamic scaling theory
$\frac{\alpha(T_c)}{f^2}$	Critical absorption at critical temperature and critical concentration
c_{pc}	Critical part of specific heat at constant pressure
c_{pb}	Background part of specific heat at constant pressure
$\frac{\alpha_b}{f^2}$	Classical absorption
$\mathrm{C_6H_5OH}$	Chemical formula of phenol
$\mathrm{H_2O}$	Chemical formula of water
R^2	Coefficient of determination
cP	Centipoise
R_ξ^+	Universal combination of critical amplitude
$R_c^+ = R_\xi^-$	Universal combination of critical amplitude
η_w	Dynamic shear viscosity of water
K_B	Boltzmann's constant
$z\nu$	Critical exponent equals to 1.9
$\propto_p$	Isobaric thermal expansion coefficient
$\propto_{pc}$	Critical term of isobaric thermal expansion coefficient
$\propto_{pb}$	Background term of isobaric thermal expansion coefficient
η_{ph}	Dynamic shear viscosity of phenol

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Abstract

The dynamic shear viscosity of a binary liquid mixture of water and phenol has been measured at different temperatures ($32.0\ ℃ \leq T \leq 75.0\ ℃$) and different concentrations (0.00% up to 100.00% by weight of phenol) by using glass capillary viscometer and Brookfield viscometer model DV-I+. The critical temperature and critical concentration have been determined to be 67.0 ℃ and 33.90% by weight of phenol respectively. The mode coupling theory (MCT) has been used to calculate the value of background viscosity (noncritical part of shear viscosity) $\eta_0 = 0.684$ cP, the Debye momentum cutoff $q_D = 0.786\ \text{Å}^{-1}$ and the MCT constant A = 0.050. The intermolecular force range L of water and phenol molecules in a binary mixture has been calculated to be 11.17 Å, this large value indicates that the mutual force between binary mixture molecules considered as a week attractive force. The critical amplitude of specific heat at constant pressure at critical concentration and above critical temperature C_{pc} has been found to be $259.16 \frac{J}{kg.K}$ by using the two scale factor university.

Chapter One

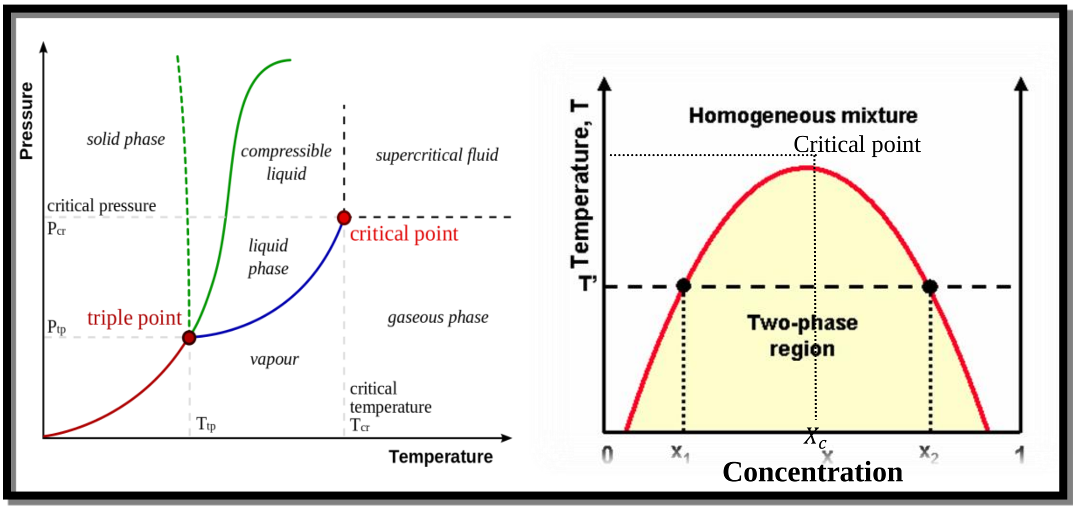
Introduction

A mixture is combination of two or more different materials that are mixed but are not combined chemically. Most substances in nature are mixtures of pure elements such as air, natural gas, gasoline, seawater, etc. The reason for this widespread occurrence is that there is a natural tendency for entropy to increase in the mixing process which leads to minimize the energy.

1.1 Binary Liquid Mixtures

A binary liquid mixture is a system consisting of two liquid substances. Pair of liquid are classified as completely immiscible such as benzene and water, completely miscible in all proportions like ethanol and water, or as partially miscible as diethyl ether and water (Narayanan, 2004: Connors, 2003: Verma *et al*, 2005).

The phase behavior of a binary system is described by a phase diagram as shown in Fig.(1.1(a,b)). The most difference between the phase behavior of a pure material and a binary mixture is in the characteristics of the phase diagram itself. For a pure material, there is a single vapor pressure curve represents the vapor – liquid equilibrium phase, whereas for a binary mixture, there is a broad region (phase envelope, saturation envelope or two – phase region) in which the two phases coexist in equilibrium (Dandekar, 2013; Putnis,1992) .



(a) (b)

Fig.(1.1): The phase diagram of (a) pure liquid (b) binary liquid mixture

The components of mixture become completely miscible above a certain temperature and concentration which is called critical temperature $\mathrm{T_c}$ and critical concentration x_c (Acree, 1984; Kittany, 2014). Any infinitesimal change in some thermodynamic parameter such as temperature, concentration and specific heat near the critical point will lead to separate the mixture to two distinct liquid phases.

There are two types of liquid-liquid critical points as shown in Fig. (1.2), these points are (Habdas, 1999; Stenland, 1995):

- Upper critical point: it is a maximum value of temperature at which the components of mixture become miscible. In the most cases, the mixture will be homogenous above the critical points (x_c and $\mathrm{T_c}$).
- Lower critical point: at which the components of mixture are miscible at lower temperature and separable at higher temperature.

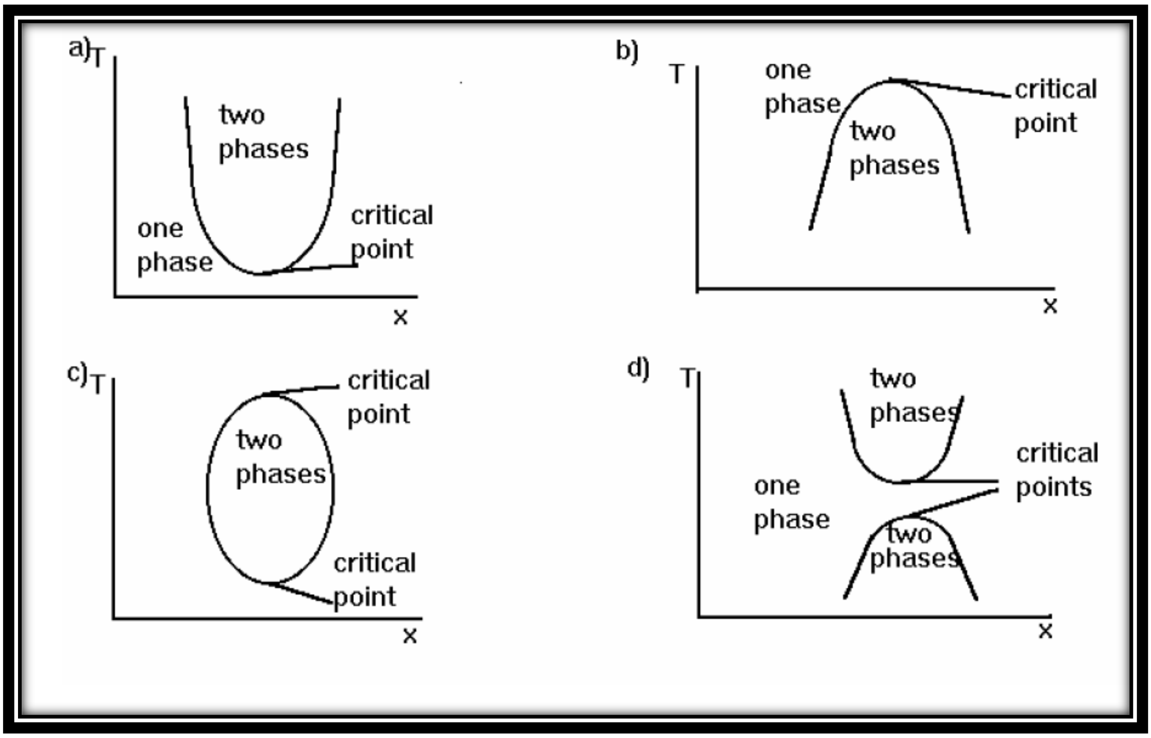


Fig.(1.2): Phase diagrams of different types of binary mixtures of limited miscibility with: (a) lower critical point (b) upper critical point (c) closed coexistence curve (d) two-phase region separated by a one-phase region (Habdas, 1999)

The importance of liquid mixture viscosity in chemical process design makes it one of the most measured transport properties. Viscosity of binary liquid mixture has direct effect on heat transfer coefficient, which is important for heat exchangers and various other heat transfer considerations.

Viscosity data is essential for calculating the critical point at which the binary mixture becomes one phase (homogenous). Also, it is used to measure and calculate some parameters like background shear viscosity, Debye momentum cutoff, and adiabatic coupling constant, isobaric thermal expansion coefficient. These parameters play an important role in chemical and pharmaceutical industry.

1.2 Literature Review

Studying the thermodynamic and transport properties of binary liquid mixtures near the critical points where homogenous mixture could be obtained become an important issue in these days. As for example, Krishnan who measured the critical points of some binary liquid mixtures, reported that the critical temperature and concentration of water-isobutryic acid are 50% by weight of water and 25.5℃, for phenol-water are 34% by weight of phenol and 69 ℃ and for water-triethyl amine are 70% by weight of water and 19℃ (Krishana, 1935).

Katti and Chaudhri measured the viscosity as a function of composition of binary mixtures of benzyl acetate with dioxane, aniline and meta-cresol. The value of interaction energy has also been calculated for each of the mixtures (Katti and Chaudhri, 1964).

In addition, Pusey and Goldburg measured the intensity and spectral width of light scattered by a critical mixture of phenol and water as a function of temperature above and below the critical temperature T_c. The diffusion constant D and correlation length ξ_T and Fixman term $K^2\xi_T^2$ have been calculated above critical temperature (Pusey and Goldburg, 1969).

Ansimore and his group measured the specific heat $C_{p,x}$ (where p is the pressure and x is the concentration) of a binary mixture of methanol and cyclohexane near the critical point. It is found that for $3 \times 10^{-5} < t < 5 \times 10^{-3}$ the specific heat given by the formula $C_{p,x} = At^{-\propto} + B$. In addition the mean range of the intermolecular force has been calculated to be $\sim 5A^0$ this value is close to that obtained from light scattering experiment (Ansimore *et al*, 1972).

Felix and Hyskens measured the dynamic viscosity of phenol and amine mixture. They found that the mixing of amines with phenol brings about a spectacular increase of viscosity which is in some case becomes more than a hundred times greater than that of pure liquid compounds, the high value of viscosity of this mixture is thus related to the presence of ions (Felix and Hyskens, 1975).

Sound wave absorption and dispersion near critical point in critical binary mixture have been studied numerically by using framework of the Kawaski mode coupling theory. They showed that the existing mode coupling approach breaks down over a wide range of temperatures and frequencies (Harada *et al*, 1980).

Ohta and Kawaski studied the dynamic critical exponents of classical liquids by using mode coupling theory. They conclude that shear viscosity for the spatial dimensionality between two and four exhibits the weak power law divergence at critical point (Ohta and Kawaski, 1976: Ohta, 1980).

The critical behavior of ultrasonic attenuation for triethylamine and water binary mixture has been studied by using the dynamic scaling theory. The experimental values of $\frac{\propto}{f^2}$Vs. $f^{-1.06}$ has been compared to the calculated value based on the dynamic scaling theory at critical temperature. The value of $\frac{\propto}{f^2}$Vs. $f^{-1.06}$ yield a straight line as predicted by theory. In addition the adiabatic coupling constant has been calculated and compared to the experimental value (Fast and Yun, 1985).

P'pein and his group studied the refractive index in a critical binary mixture of triethylamine and water. They found that the critical anomaly in the refractive index includes an intrinsic effect opposite in sign to the density contribution (P'pein *et al,* 1988).

Ultrasonic velocity, absorption and shear viscosity have been measured as a function of concentration and temperature for a binary solution of polyvinylpyrrolidone and water. The result shows that the velocity increase non linearly with a temperature and linearly with concentration. Also, the viscosity values increase monotonically with concentration and decrease with temperature (Spikler *et al*, 1989).

Kinematic viscosities and densities of 68 linear, branched, cyclic and aromatic hydrocarbons binary liquid systems have been measured at 298.15 K over the entire composition rang. The McAllister model has been used to fit experimental values of kinematic viscosities (Chevaller *et al*, 1990).

The refractive index, density, turbidity and specific heat for a mixture of triethylamine and water at critical concentration have been measured. The values of the correlation length and osmotic compressibility amplitudes have been calculated. The universal amplitude combinations $R_{\xi}^{+} = 0.29 \pm 0.02$ and $R_{c}^{+} = R_{\xi}^{-} = 0.056 \pm 0.01$ have been determined, this value agree with the two scale factor universality (Zalczer and Beysens, 1990).

Abdelraziq and his group measured the ultrasonic velocity and absorptions at the critical concentration as a function of frequency and temperature for a carbon tetrachloride and coconut oil mixture. The result indicates that the absorption coefficient at critical concentration increases as the critical temperature is approached from high temperature region for all frequencies, and it decreases at critical temperature and concentration with increasing frequency (Abdelraziq *et al*, 1992).

Esquivel-Sirvent and his group studied the absorption and velocity of ultrasound in binary mixture of poly (ethylene glycol) and water, they found that the ultrasonic absorption decreases with increasing temperature at given concentration and increases with increasing concentration at given temperature. Velocity increases with increasing temperature and increasing concentration, the shear viscosity is decreasing with temperature but increasing with concentration (Esquivel-Sirvent, 1993).

Kob and Andersen tested the mode coupling theory for a supercooled binary Lennard-Jones mixture, and calculated the Van Hove, correlation function (Kob and Andersen, 1995), intermediate scattering function and dynamic susceptility (Kob and Andersen, 1995).

Abdelraziq studied the ultrasonic absorption as a function of temperature and frequency for a binary mixture of cyclohexane and aniline. The results of the absorption coefficient for the critical concentration increase as the critical temperature is approached from high temperature region for all frequencies (Abderaziq, 1996). The heat capacity of trithylamine and water binary mixture has been measured near its lower critical point by using a scanning, adiabatic calorimeter. The critical exponent $\propto$ was determined to be 0.107 ± 0.006, consistent with theoretical predictions (Flewelling *et al*, 1996).

The dynamic shear viscosity as a function of molar composition and temperature of a binary mixture of aniline-cyclohexane has been reported. The results are analyzed in terms of the mode coupling theory. The needed dependence of the correlation length on x and t is established using a scaling equation of state. The agreement between experiments and theory is quite good (D'Arrigo *et al*, 1997).

The shear viscosity as a function of temperature near the critical temperature and concentration of a binary mixture of nitrobenzene and n-heptane have been measured and analyzed by using mode coupling theory. The Debye momentum cutoff q_D was determined to be $0.5A^{-1}$ (Abdelraziq *et al*, 1997). The critical amplitudes of the thermal expansion, the specific heat and the adiabatic coupling constant have been calculated by using two scale factor universality. The experimental values of $\frac{\alpha}{\alpha_c}$ (where α is the absorption at critical concentration above the critical temperature) for this mixture are compared to the scaling function $\mathrm{F}(\omega^*)$ and show a good agreement with theory. In addition the velocity of this system at critical concentration and above critical temperature has been studied; the result indicates that the velocity decrease linearly with increasing temperature (Abdelraziq *et al*, 1990).

The ultrasonic absorption at 5, 7, 10, 15, 21 and 25 MHz, above critical temperature for a binary mixture of perfluomethyl - cyclohexane and carbon tetrachloride have been measured and analyzed by using the dynamic scaling theory. The values of $\frac{\alpha}{f^2}$ Vs. $f^{-1.06}$ show good agreement with a theory (Abdelraziq, 2000).

The experimental values $\frac{\propto_\lambda(crit)}{\pi u(\omega)^2 A(T)}$ (where, $\propto_\lambda$ $(crit)$ is the attenuation per wavelength at critical concentration, $u(\omega)$ is the sound velocity and A(T) is the critical amplitude) from mode coupling theory of aniline and cyclohexane binary mixture have been compared with the theoretical expressions given by Fixman, Kawasaki, Mistura and Chaban. The experimental data at low reduced frequency has been found to agree well with the Hornowski's model of A(T). However, for the large values of reduced frequency $(\omega^* > 10)$ the mode coupling theory of Kawaski and Shiwa still show poor agreement with the measured data. The correlation length $\xi_0 = 2.25 \pm 0.1\ A^0$, the adiabatic coupling constant g = $-$ 0.15 and diffusion coefficient $D_0 = 9.24 \times 10^{-6}\frac{cm^2}{s}$ of a binary mixture of aniline and cyclohexane have been calculated by Abdelraziq (Abdelraziq, 2001).

The mass density ρ and speed of sound have been measured for six binary mixtures containing n-alkane at temperature 298.15K. The values of excess molar volumes (V^E) and deviation in isentropic compressibilities (ΔB_S) have been calculated by Vyas and Nauliyal. They found that the devation in isentropic compressibility is negative for all six binary mixtures, while the excess molar volumes exhibit inversion in sign in one binary mixture (n-heptane + n-hexane) and remaining in the other mixtures (n-heptane + toluene, cyclohexane + n-heptane, cyclohexan + n-hexane, toluene + n-hexane and n-decane + n-hexane) (Vyas and Noutiyal, 2002). Wahab and his group measured the excess mole volumes of a binary mixtures of

$(\mathrm{x})\mathrm{C_6H_5CH_3} + (1-\mathrm{x_1})\mathrm{CH_3CN}$ or $(1-\mathrm{x_1})\mathrm{C_6H_5No_2} + (1-\mathrm{x_1})\mathrm{C_2H_5No_2}$ as a function of mole fraction of $\mathrm{C_6H_5CH_3}(\mathrm{x})$ at temperature 303.15K (Wahab *et al*, 2002).

The shear viscosity, mutual diffusion coefficient and ultrasonic attenuation spectra from 100KHz to 500KHz have been measured at different temperatures for the ethylamic and water mixture. The adiabatic coupling constant for this mixture has been derived from the amplitude of the Bhattacharge-Ferrell term in the ultrasonicspectra (g = 0.19) and a thermodynamic relation(g = 0.98). This difference in the g values is taken as an indication of the limitations of the Bhattacharjee–Ferrell model (Brhrends *et al*, 2002).

Shear viscosity coefficients of a binary mixture of nitroethan and 3-metheylpentane have been measured. The noncritical part of viscosity $\eta_0 = 0.358$ cP, Debye momentum cutoff $q_D = 0.275\text{Å}^{-1}$, the intermolecular force range L=10.65 Å and the constant A = 0.054 in the mode coupling theory have been determined by. Our values of η_0 and A are in good agreement with the literature values (Abdelraziq, 2002).

The universal quantity R_{ξ^+} of some binary liquid mixtures (Nitrobenzene + n-heptane, Nitroethane + isooctane, Methanol + n-heptane …) has been calculated by Abdelraziq. The results indicates that the value of $R_{\xi^+} = 0.2691 \pm 0.0028$ is agrees with the theoretical value of $R_{\xi^+} = (\frac{n}{4\pi})^{1/d}$ for n = 1 and d = 3 (Abdelraziq, 2003).

Iwanowski's group measured the ultrasonic attenuation spectra, shear viscosity and mutual diffusion coefficient for a binary mixture of n -

pentanol and nitromethane at different temperature near the critical point. The relaxation rate of order parameter fluctuation shows power law behavior as theoretically predicted. The adiabatic constant g = 0.03 has been calculated (Iwanowski *et al*, 2004).

Ultrasonic velocity has been studied at frequency 2MHz by using ultrasonic pulse echo system. The values of internal pressure, molar free volume and dielectric constant have been calculated. It is found that there is a linear relation between ultrasonic velocity and acidity constant (Nayakula *et al*, 2005). Chen and his team studied the UV-V spectra of probe phenol blue in Co_2 + ethanol and Co_2 + n-pentane binary mixtures at temperature 308.1K and different pressures (Chen *et al*, 2005).

The ultrasonic absorption and velocity of a binary mixtures of benzene-coconut oil and hexan - β,β'- dichloroethyl ether have been measured as a function of temperature. The ultrasonic absorption at 5, 7, 10, 15, 21 and 25 MHz and above critical temperature T_c is analyzed by the dynamic scaling theory of Ferrell and Bhattacharjee. The results indicates that ultrasonic absorption of these binary mixtures exhibit strong temperature and frequency dependence near T_c. Also, ultrasonic velocity behaves as a linearly decreasing with increasing temperature above critical temperature (Abdelraziq, 2005).

The density and viscosity of a binary mixtures of water and three ionic liquids (1-ethyl-3-methylimidazolium ethylsulfate, 1-ethyl-3-methylimidazolium trifluoracetate and 1-ethyl-3-methylimidazolium trifluoromethane sulfonate) have been measured at atmospheric pressure and temperature from 278.15K to 348.15K. The temperature dependence of density and viscosity for these mixtures described by an empirical second order polynomial and by the Vogel-Fulcher-Tammann equation, respectively (Rodriguez and Brennecke, 2006).

Iwanowski and his group studied the acoustical attenuation spectrometry, dynamic light scattering, shear viscosity, density and heat capacity for methanol and n-hexane mixture (Iwanowski *et al*, 2006). The fluctuation correlation length, relaxation rate of fluctuation and universal critical exponents have been calculated for critical system of 3-methyl pentane, nitroethane and cyclohexane (Iwanowski *et al*, 2008).

Toumi and his group measured the shear viscosity and the electrical conductivity in the vicinity and far from the critical temperature for the binary mixture of triethylamine and water with an ionic impurity ($\mathrm{K^+, Cl^-}$ions) at various concentrations. They found that the electrical conductivity exhibits a monotonous deviation from the Vogel-Fulcher-Tammann (VFT) behavior at temperature range $\Delta\mathrm{T} = \mathrm{T_c} - \mathrm{T} < 2℃$ (Toumi *et al*, 2011).

The densities and viscosity correlations for four pure ionic liquids (1-butyl-3-methylimidazolium thiocyanate, 1-butyl-4-methylpridinium thiocyanate, 1-butyl-1-methylprrolidinium thiocyanate and 1-butyl-1-methylpiperidinium thiocyanate) and their mixtures with water have been measured at temperatures from 298.15K up to 348.15K and ambient pressure. The result of the correlations with the second order polynomials, Redlich-Kister equation, and Vogel-Fulcher-Tammann (VFT) equation of density, viscosity, excess molar volume and viscosity deviation are with very low standard deviations (Domanska and Krolikowaska, 2012).

Srilakshmi and his group measured the density, speed of sound and viscosity for a binary mixture containing o-anisidine and amyl acetate at temperatures 303.15K, 308.15K, 313.15K and 318.15K at atmospheric pressure. The adiabatic compressibility (β), free volume (V), free length (L_f), internal pressure and their excess values have been calculated. They found that there exist strong molecular interactions between o-anisidine and amyl acetate (Srilakshmi *et al*, 2014).

Qasem modified the analytic function of Fixman's theory to get a good agreement with the experimental behavior of the binary liquid mixtures at critical concentration and above critical temperature. This modification of Fixman's scaling function depends on the ratio of heat capacities at constant pressure and volume (Qasem, 2014; Qasem and Abdelraziq, 2014).

The dynamic shear viscosity at different temperature and concentration of a binary mixture of methanol and cyclohexane has been measured by Omar. The adiabatic coupling constant g = -0.29, the isobaric thermal expansion coefficient $\alpha_p = 7 \times 10^{-3}{}^{\circ}\mathrm{C}^{-1}$ and diffusion coefficient D =$1.89 \times 10^{-5} cm^2/s$ have been calculated by using the dynamic scaling theory (Omar, 2014; Omar and Abdelraziq, 2014).

The Debye momentum cutoff of the critical binary mixture of carbon tetrachloride and coconut oil has been studied by Kittany, the result was $q_D = 0.126\text{Å}^{-1}$. The noncritical part of the dynamic shear viscosity

$\eta_0 = 2.59\ cP$, the mode coupling theory universal constant A = 0.054 have been measured (Kittany, 2014).

The critical temperature and critical concentration of benzene and coconut oil binary mixture have been measured by Abdo. The Debye momentum cutoff $q_D = 0.358\,\text{Å}^{-1}$, the noncritical part of shear viscosity $\eta_0 = 1.30\,\text{cP}$, the constant A = 0.052 ± 0.003, the correlation length $\xi_0 = 1.02\,\text{Å}$and the intermolecular force rang L = 9.90 have been measured by using the mode coupling theory. The intermolecular forces between molecules are weak because the value of viscosity is very small (Abdo, 2014; Abdo and Abdelraziq, 2014).

1.3 Objectives and Motivations

The knowledge of thermodynamic parameters and transport properties of binary liquid mixtures are interest as they control the field of technological application.

Namely, the binary liquid mixture of phenol and water play vital role in chemical industry and chemical research. There are rarely published research articles about its physical and thermodynamics properties of this mixture. Thus, we are motivated to study the dynamic shear viscosity, critical behavior and mode coupling parameters for the mixture of water and phenol.

The main aims of this thesis are:

- To study the concentration and temperature dependence viscosity of binary mixture of water and phenol.

- To determine the critical temperature (T_c) and critical concentration (x_c).
- To calculate the noncritical part of the measured shear viscosity η_0, Debye momentum cutoff q_D (Debye parameter), the intermolecular force rang L and the constant A by using mode coupling approach.
- To calculate the critical amplitude of specific heat at constant pressure C_{pc} .

1.4 Organization of the Thesis

This thesis is divided into five main chapters:

- **Chapter One** contains brief introduction to binary liquid mixtures, previous studies which are focusing on the properties of binary mixture, motivation and objectives of this research.
- **Chapter Two** discusses the theory of viscosity for pure liquids and binary liquid mixtures, temperature and concentration dependence viscosity, and mode coupling theory.
- **Chapter Three** has a brief description of the methodology of the experiment, the characteristics of sample, experiment apparatus, procedure and statistical analysis.
- **Chapter Four** reports the result and the relative discussion of the measured data, determining the critical points and calculating the mode coupling theory (MCT) parameters.
- **Chapter Five** lists the concluding remarks about the water and phenol binary mixture.

Chapter Two

Theory

2.1 Introduction

The viscosity is an important internal property for all fluids; it can be used to determine the quality and stability of food system. It is defined as the resistance of fluid to flow, this resistance is due to intermolecular friction when layers fluids slide by one another (Latini *et al*, 2006; Pal Arya, 2001). In order to clarify the concept of dynamic viscosity, we consider a layer of fluid flow between two infinite, horizontal, parallel plates separated by a distance h (Fig. (2.1)). The lower layer is fixed while the upper one pulled by an external force to move with constant velocity ν. This force distorted the fluids motion from its original shape (a b c d) to another shape (a $b^* c^* d$). The force needed to move the upper plate is proportional to the area of plate A and the velocity ν, and inversly proportional to the distance that seperates the plates h, it can be described in the following mathematical form (Serway and Vuille, 2001; Totten, 2006; Grot and Antonsson, 2009):

$$\mathrm{F} = \eta \, \mathrm{A} \, \frac{\nu}{\mathrm{h}} \tag{2.1}$$

Where η is the dynamic viscosity, $\frac{\nu}{\mathrm{h}}$ is the rate of shear deformation (shear velocity).

The differential form of equation (2.1) is called the Newton's law of viscosity

$$\frac{F}{A} = \tau = \eta \frac{d\nu}{dh} \tag{2.2}$$

Where $\frac{F}{A} = \tau$ is the shearing stress (N/m^2) and $\frac{dV}{dh}$ is the velocity gradient or deformation rate (1/s).

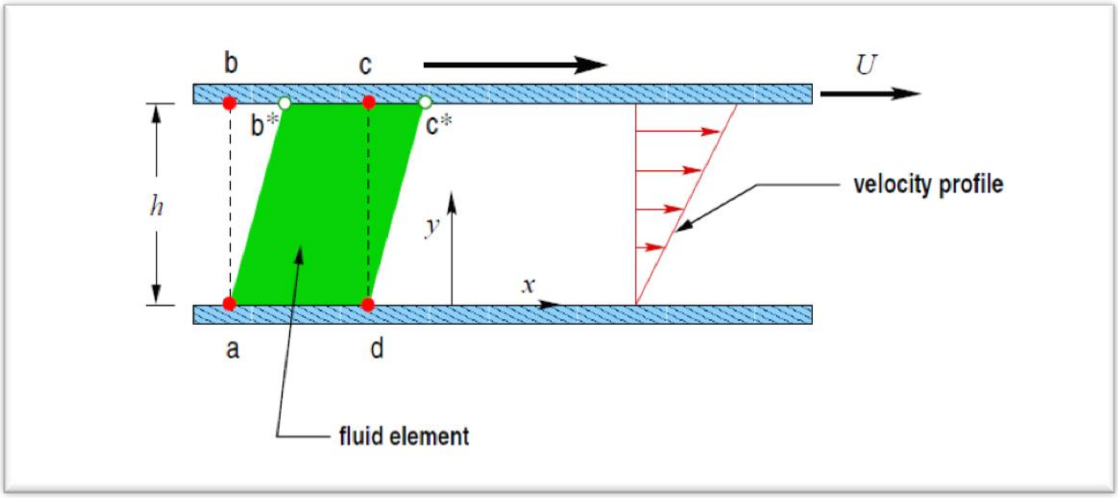


Fig. (2.1): Fluid flow between two horizontal, parallel plates separated by a distance h

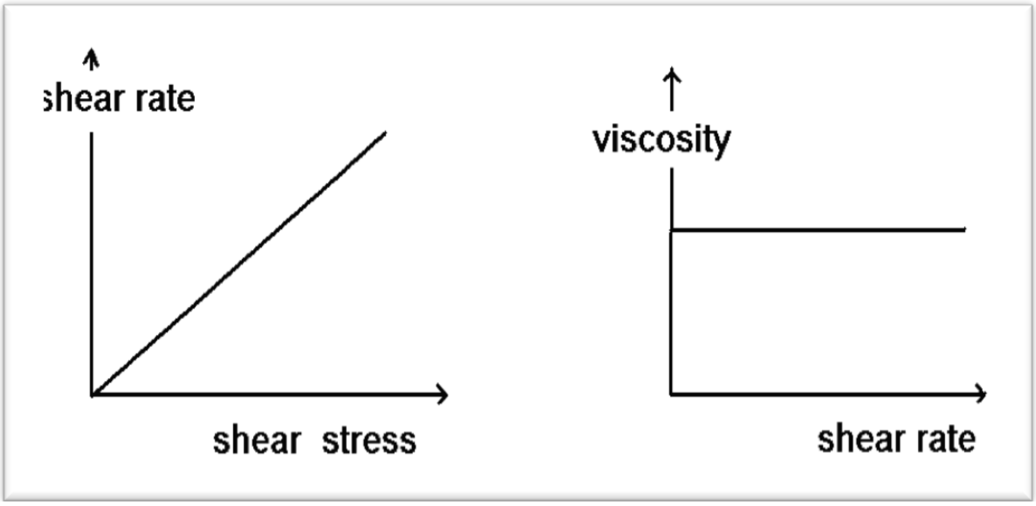
The ratio of dynamic shear viscosity (absolute viscosity) to density of fluid at same temperature and pressure is called kinematic viscosity (Liptak, 1994; Wang *et al*, 2009; Shashi Meron, 2005).

$$\nu = \frac{\eta}{\rho} \tag{2.3}$$

Where ν is the kinematic viscosity in Stoke, η is the absolute viscosity in Poise and ρ is the density in $\frac{g}{cm^3}$.

According to equation (2.2) the fluid can be divided into two categories:

- **Newtonian Fluid:** the viscosity of these fluids remains constant at given temperature. It is independent of shear stress or rate, as shown in Fig. (2.2) which represent the linear relationship between shear stress and shear rate (Mcketta, 1990; Hartnett *et al*, 1996).



(a) (b)

Fig. (2.2): (a) The linear relationship between shear stress and shear rate

(b) The relationship between viscosity and shear rate

- **Non-Newtonian Fluid:** in this type of fluids the viscosity at given temperature depends on the shear rate, the viscosity may increase or decrease depending on the type of fluid (Mcketta, 1990; Berk, 2013; Hartnett *et al*, 1996). The non-Newtonian fluids are divided into two group:
  - (I) **Time Independent**: the viscosity of fluid does not change as a function of time, like psuedoplastic and dilatants fluid.
  - (II) **Time Dependent**: the viscosity of fluid changes with time when measure at a specific shear rate, like thixotropic whose viscosity decreases with time at constant shear rate and rheopectic material in which the viscosity increases with time as shown in Fig. (2.3) (Mcketta, 1992).

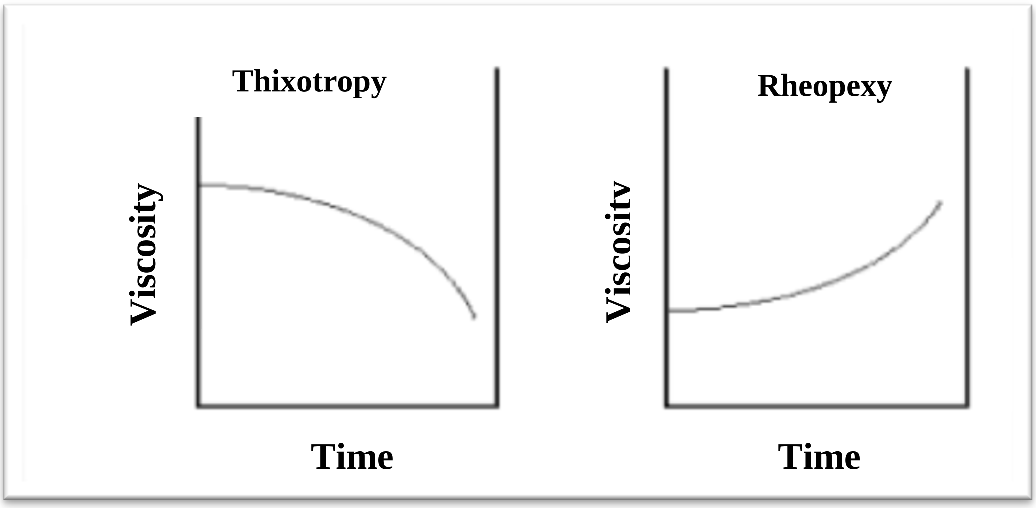


Fig. (2.3): Viscosity as a function of time for thixotropic and rheopectic materials

2.2 Pure Liquids – Viscosity Theories

The viscosity of pure fluid is affected by many factors such as temperature, pressure, concentration, particle size, attractive force, catalyst, molecular weight, shear rate and storage age (Lide, 2005).

The relationship between dynamic shear viscosity and temperature is fitted with Arrhenius-type equation.

$$\eta = Ae^{\frac{E_a}{RT}} \qquad (2.4)$$

Where η is the dynamic shear viscosity (Pa.s), A is a constant (Pa.s), E_a is the activation energy (J/mol), T is the absolute temperature (K) and R is the gas constant (J/mol.K) (Fasina and Colley, 2008; Giap, 2010).

Due to the complex nature of liquids, there is no comprehensive theory to describe the viscosity of liquids. Theoretical methods of calculating liquid viscosity and molecular dynamic approaches give the result in large deviations from the measured data. While, the empirical and semi-empirical theories provide good results (Viswanath, 2007).

Andrade suggested the existence of a linear relation (two-constants equation) between the logarithm of dynamic viscosity and the reciprocal of absolute temperature.

$$\ln \eta = A + \frac{B}{T} \tag{2.5}$$

where η is the dynamic viscosity, A and B are constants determined empirically at temperature above the boiling point and T is the absolute temperature (Cheremisinoff, 1996).

2.3 Dynamic Viscosity of Binary Mixtures

The behavior of the viscosity for binary liquid mixtures is like the pure liquids except at critical point. Many properties of systems close to critical point follow the power laws, so their thermodynamic properties diverge or vanish at the critical point.

There are some models that predict the behavior of viscosity of binary liquid mixtures around the critical point such as the mode coupling theory, a renormalization group theory and dynamic scaling theory.

2.3.1 Renormalization Group Theory (RGT)

The renormalization group theory developed by Wilson (Wilson, 1975). It is designed to deal with the fluctuations of all wavelength scales, and to predict non-classical critical exponents and universal features near the critical point (Ramana and Menon, 2012: Fisher, 1975).

The basis of this theory is to describe the behavior of ultrasonic attenuation in order to calculate the bulk shear viscosity. The expression of sound attenuation coefficient per wavelength at critical concentration ($\alpha_\lambda(crit)$) can be written as the following equation shows (Kroll and Ruhland, 1981)

$$\frac{\alpha_\lambda(crit)}{u^2(\omega)} = \pi H \lim f(\omega^*) \tag{2.6}$$

Where $\alpha_\lambda(crit)$ is the sound attenuation coefficient per wavelength at critical concentration, u(ω) is the velocity of sound wave, H is the critical amplitude, $f(\omega^*)$ is the scaling function and $\omega^* = \frac{\omega}{\omega_0}$is the reduced frequency, such that $\omega_0 = \frac{\omega_D}{2}$ is the characteristic frequency of order parameter fluctuations.

The characteristic frequency ω_D can be written as:

$$\omega_D = \frac{K_B T}{3\pi\eta\xi^3} = 2D_0\xi_0^{-2}t^{zv} \tag{2.7}$$

Where ξ_0 is the correlation length at critical point, K_B is the Boltzmann's constant, z and v are critical exponent and *t* is the reduced temperature.

The critical amplitude H in equation (2.6) is given by:

$$H = \frac{c_1' c_3'}{|g(t,\omega^*)|^2}\, t^{\frac{\alpha^*}{v}} \tag{2.8}$$

Where $g(t,\omega^*) = c_2' + c_3' t^{\frac{\alpha^*}{v}} f(\omega^*)$, c_1', c_2' and c_3' are non-universal constant and α^* is critical exponent equals to 0.11.

2.3.2 Dynamic Scaling Theory (DST)

The dynamic scaling theory was suggested by Ferrell and Bhattacharjee (Ferrell and Bhattarjee, 1981). Then, it was generalized for magnetic systems by Halperin and Hohenberg (Hohenberg and Halperin, 1969;

Hohenberg and Halperin, 1977). It deals with a scaling function that determined directly from acoustical spectrometry.

Dynamic scaling theory is a theory of critical attenuation in binary liquid mixtures based on the frequency-dependent specific heat concept. It predicts a linear relationship between $\frac{\propto}{f^2} \propto f^{-1.06}$ above the critical point, as shown in the following equation (Iwanowski, 2007; Abdelraziq, 2000):

$$\frac{\propto(crit,T_c)}{f^2} = \left[\frac{\pi^2\alpha^* g^2 u(\omega) C_{pc}}{2z\nu T_c C_p^2(t_f)}\left(\frac{a\omega_0}{2\pi}\right)^{\frac{\alpha^*}{z\nu}}\right] f^{-\left(1+\frac{\alpha^*}{z\nu}\right)} + \frac{\propto_b}{f^2} \tag{2.9}$$

Where $\frac{\propto(crit,T_c)}{f^2}$ is the critical absorption at critical temperature and critical concentration, $u(\omega)$ is the sound velocity, c_{pc} is the critical amplitude of specific heat at constant pressure, $C_p(t_f)$ is the specific heat at a characteristic reduced temperature, α^* and $z\nu$ are critical exponents equals to 0.11 and 1.9 respectively, $a = \frac{\omega}{\omega_0} t_f^{-1.9}$ is dimensionless scaling factor of order unity and $\frac{\propto_b}{f^2}$ is the classical absorption.

The specific heat at constant pressure of binary mixture of critical composition is given by:

$$C_p = C_{pc} t^{-\alpha^*} + C_{pb} \tag{2.10}$$

Where C_{pb} is the background specific heat at constant pressure.

The adiabatic coupling constant g is a dimensionless parameter depends on the pressure, temperature and thermal expansion. It was introduced by Ferrell and Bhattachariee and is given by:

$$g = \rho_c C_p \left(\frac{\partial T_c}{\partial p} - \frac{T\alpha_p}{\rho C_p}\right) \tag{2.11}$$

$$g \cong \frac{C_{pb} \propto_{pc} T_c}{C_{pc}} - \propto_{pb} \tag{2.12}$$

Where ρ_c is the mass density at critical point, α_p is the isobaric thermal expansion coefficient given by:

$$\propto_p = \propto_{pc} t^{\alpha^*} + \propto_{pb} \tag{2.13}$$

Where $\propto_{pc}$ and $\propto_{pb}$ are the critical and background terms of isobaric thermal expansion coefficient.

The critical term of absorption coefficient $\propto (crit, \omega, T)$ can be written as a function of reduced frequency

$$\frac{\propto (crit,\omega,T)}{\propto (crit,\omega,T_c)} = \frac{\propto}{\propto_c} = F(\omega^*) = (1+\omega^{*0.5})^{-2} \tag{2.14}$$

Where $\propto (crit, \omega, T)$ is the critical term of absorption coefficient at critical concentration and temperature T, $\propto (crit, \omega, T_c)$ is the critical term of absorption coefficient at critical concentration and critical temperature, $F(\omega^*)$ is the theoretical scaling function and ω^* is the dimensionless reduce frequency which given as:

$$\omega^* = \frac{\omega}{\omega_D} = \frac{2\pi f}{\omega_0 t^{z\nu}} \tag{2.15}$$

Where $\omega = 2\pi f$ is the angular frequency, ω_D is a characteristic temperature dependent relaxation rate given by

$$\omega_D = \frac{K_B T}{3\pi\, \eta \xi^3} = \frac{K_B T_c}{3\pi\, \eta_0 \xi_0^3} t^{z\gamma} = \omega_0 t^{z\gamma} \tag{2.16}$$

2.3.3 Mode-Coupling Theory (MCT)

The mode coupling theory has been developed by Kawasaki (Kawasaki, 1970), Perl and Ferrell (Perl and Ferrell, 1972). It is used to explain the experimental behavior of binary mixtures at critical concentration and above critical temperature.

This theory applied when the modes of one fluid perturbed by the modes of the other, these perturbations lead to coupling and exchange of energy. The basic idea is that nonlinear couplings between the slow dynamical modes can lead to divergences in kinetic coefficients like viscosity (Kittany, 2014).

The mode-coupling theory predicts a critical anomaly of shear viscosity coefficient (Abdelraziq, 2002).

$$\frac{\eta-\eta_0}{\eta} = \frac{\Delta\eta}{\eta} = A \ln\xi + A \ln q_D \tag{2.17}$$

Where η_0 is the noncritical part of measured shear viscosity (The background viscosity), A is constant, A = $\frac{8}{15\pi^2} \approx 0.054$ (D’Arrigo *et al*, 1977), q_D is the Debye momentum cutoff and ξ is the correlation length.

The background viscosity (η_0) is used to dissipate the long wavelength modes of motion, this damping can be attributed to a dispersion medium (Robert, 2008). At critical concentration, the dynamic shear viscosity η as a function of reduce temperature can be written as a power law (Kawasaki, 1976):

$$\eta = \eta_0 t^{-x_\eta v} \tag{2.18}$$

Where x_η, υ are critical exponents, $x_\eta \upsilon$ = 0.04, t is the reduced temperature $t = \frac{T - T_c}{T}$ (Klein and Woermann, 1978; Abdelraziq, 2002).

The correlation length (ξ) is a measure of the range through which fluctuations in one region of space are correlated with those in the other region (Sessoms *et al*, 2009). It is related to critical temperature as the following equation shows (Chang *et al*, 1971; Abdelraziq, 2002):

$$\xi = \xi_0 t^{-\nu} \tag{2.19}$$

Where the ξ_0 is the critical amplitude of the correlation length of the concentration fluctuations, t is the reduced temperature and ν is critical exponent and it is equal to 0.64.

The intermolecular force range is responsible for the mechanism of momentum and energy transfers between molecules in dense fluid and in particular in liquids (Byung, 2006). It is represented as a function of correlation length around the critical point (Klein and Woermann, 1978):

$$L = 2.45\, \xi_0 {T_c}^{(\gamma-1)/2} \tag{2.20}$$

Where L is the intermolecular force, T_c is the critical temperature for binary system (K) and γ is critical exponent ($\gamma = 1.25$).

2.4 Molecular Vibration Modes

Molecules are made of atoms connected by chemical binds (distance between atoms) is not fixed. These atoms are in periodic motion while the molecule as a whole has constant translational and rotational motion this periodic motion known as molecular vibration which can be considered like motion of particles connected by spring (harmonic oscillator) (Guozhen, 2008; Dresselhaus *et al*, 2008).

A nonlinear molecule with N atoms has 3N-6 degree of freedom for vibrations of 3N-6 modes, 6 correspond to degrees of freedom for translation and rotation of the center of mass, which have no or very small restoring force. Any atomic motion of the molecule can be expressed by a linear combination of these 3N-6 independent, orthogonal vibrations called normal modes. A linear molecule has 3N-5 normal modes, since we need to subtract three translational and two rotational degrees of freedom, as rotation about the molecular axis doesn't change in motion. For example, water (H_2O) has $3(3) - 6 = 3$ normal modes, and HCN has $3(3) - 5 = 4$ normal modes (Jorio *et al*, 2011; Lewars, 2010).

Vibrations divided into two basic categories (Serdyuk *et al*, 2007):

- Stretching vibration: which involves a continuous change in interatomic distance along the axis of the bound between atoms (change in the length of bond).
- Bending vibration: which represented the change in the angle between two bonds. It classifies into four types: scissoring, rocking, wagging and twisting vibrations. The stretching and bending vibrations modes are illustrated in Fig.(2.4).

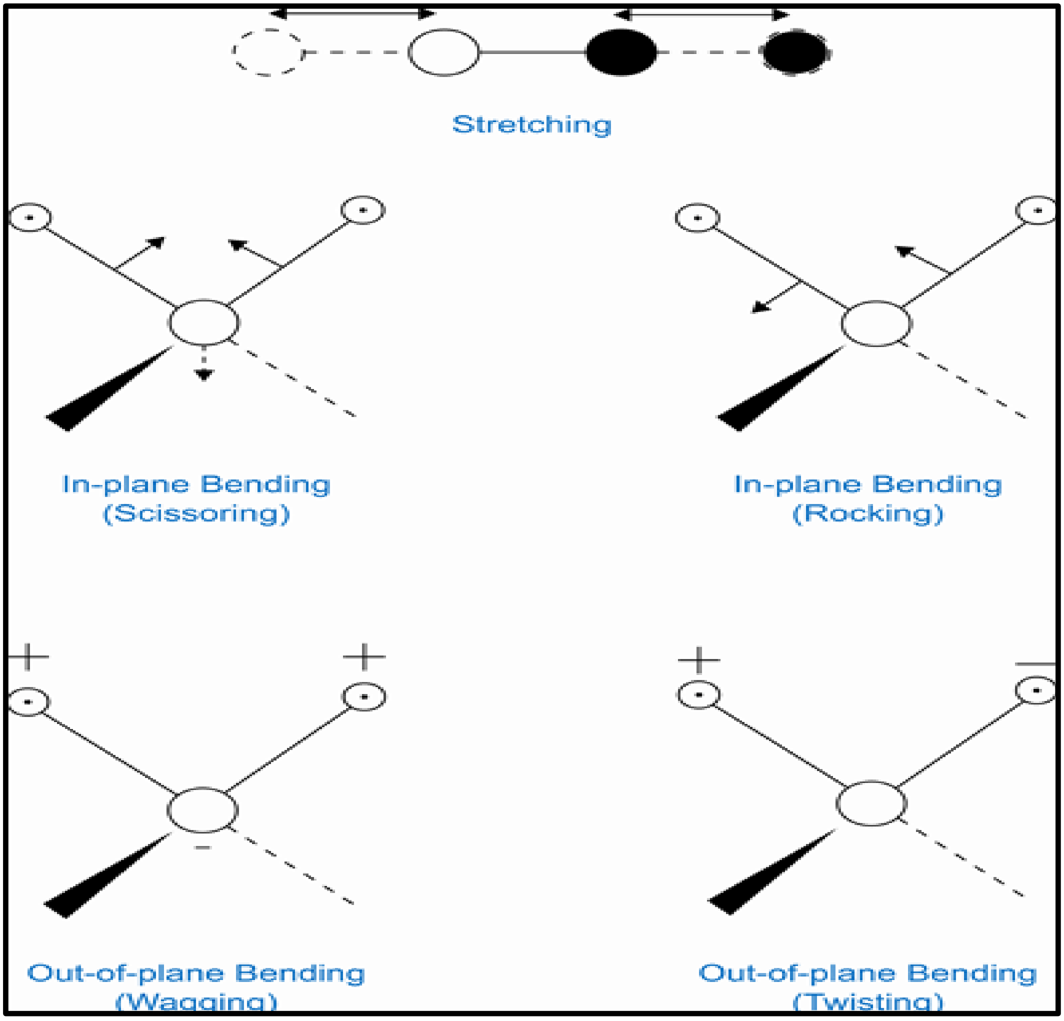


Fig.(2.4): Types of molecular vibration. Note that (+) the motion towered the reader and (-) motion away from the reader

Chapter Three

Methodology

The samples of a binary liquid mixture have been prepared from phenol and pure water with different concentration (0.00%, 10.17%, 20.80%, 30.20%, 31.80%, 32.90%, 33.90%, 35.00%, 37.10%, 40.20%, 49.80%, 59.79%, 70.40%, 79.77%, 89.90%and 100.00% by weight of phenol). The dynamic shear viscosity for each sample has been measured at temperature range from 32.0 ℃ up to 75.0 ℃ by using glass capillary viscometer and Brookfield viscometer model DV-I+.

3.1 The Samples

3.1.1 The Characteristics of Phenol and Water Materials

- Phenol: it is an organic compound with molecular formula C_6H_5OH. It consists of phenyl group (C_6H_5) and hydroxyl group (OH) as shown in Fig. (3.1). Phenol is soluble in water with rate 84.2 g phenol in 1000 ml water (Ahluwalia and Raghav, 1997; Mishra *et al*, 2013). Phenol (carbolic acid) is the simplest aromatic alcohol and it can act in intermolecular interactions as both proton donor and proton acceptor (Gor *et al*, 2011).

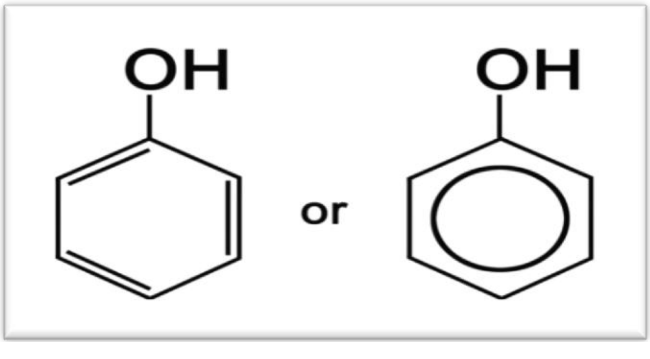


Fig. (3.1): Chemical structure of phenol.

- Distilled water: is a chemical compound with chemical formula H_2O. Water is a colorless liquid material at standard temperature and pressure (Manaha, 2002). Some physical and chemical properties of water and phenol are given in table (3.1).

Table (3.1): Physical and chemical properties of water and phenol.

Property	Phenol	Water
Chemical Formula	C_6H_5OH	H_2O
Physical State	Solid Crystalline	Liquid
Color	White	Colorless
Solubility in water	Soluble in water	____
Melting point	40.5℃	0℃
Boiling point	181.7℃	100℃
Density	1.07 g/cm^3	1 g/cm^3
Dynamic viscosity (at 27℃)	8.56 cP	0.89 Cp
Dynamic viscosity (at 20℃)	12.72 cP	1.0028 cP
Purity	99.5-100%	100%

3.1.2 The Characteristics of Phenol and Water Binary Mixture

In an aqueous solution, phenol is easily hydrated and forms the phenol-water complex. The hydrogen bonded of phenol with water is very complex; there are three configurations of the phenol-water mixture (Dai, 1995; Gor *et al*, 2011; Kryachko and Nakatsuji, 2002):

(I) With the hydrogen bonding between the phenol hydrogen from hydroxyl group and the water oxygen (PW1).

(II) With the hydrogen bonding between the phenol oxygen and the water hydrogen (PW2).

(III) With water molecule above and in the center of the benzene ring (PW3). These three complexes are presented in Fig.(3.2).

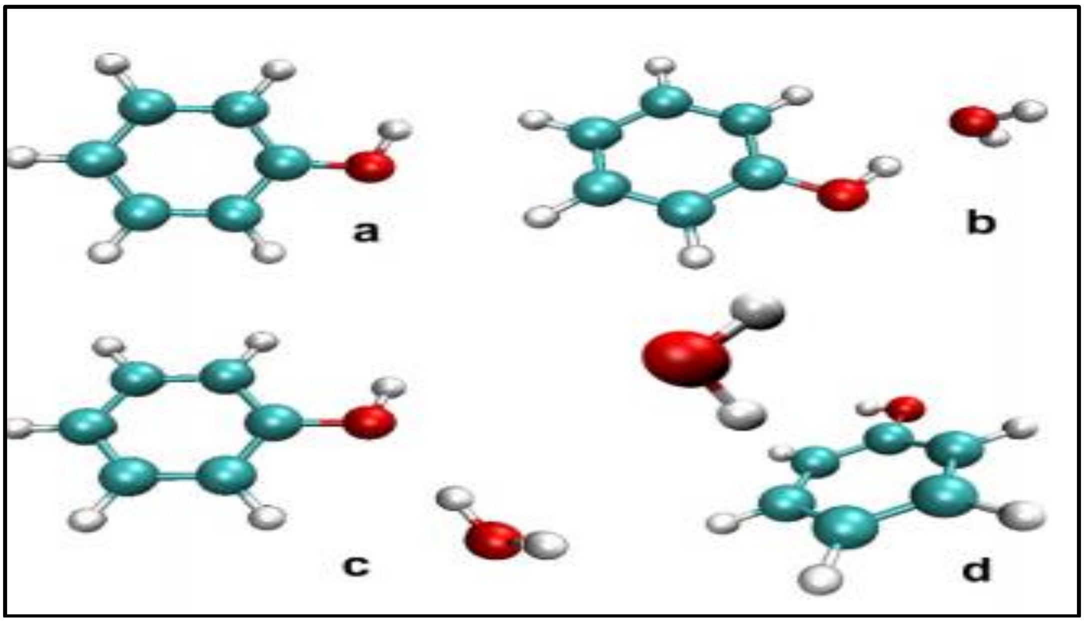


Fig.(3.2): (a) Phenol molecules (b) PW1 complex (c) PW2 complex (d) PW3 complex

There is a strong anharmonic coupling between the hydrogen bonded OH stretching and bending modes in water and phenol complex (Yamamoto *et at*, 2003).

3.2 Experimental Apparatus

3.2.1 Viscosity Apparatus

- A Brookfield Viscometer Model DV-I+:

  It was used to measure the dynamic viscosity from 100 up to 1.33×10^7cP with accuracy $\mp 1\%$ (Fig. (3.3(a)). It consists of a set of seven spindles (RVSPINDLE SET) and UL-ADAPTER. The rotational speeds of the spindles are divided into two set. The first is:

0.0, 0.3, 0.6, 1.5, 3, 6, 12, 30, 60RPM and the second set is: 0.0, 0.5, 1, 2, 2.5, 4, 5, 10, 20, 50, 100 RPM (Brookfield manual, 1999).

- Glass Capillary Viscometer:

  It is used to calculate the viscosity of liquid sample by measuring the time required for the level of liquid to drop from one mark to the other (Fig. (3.3 b)), according to the following equation (Viswanath, 2007; Troy and Beringer, 2006):

$$\eta = K\rho t \tag{3.1}$$

Where η is the dynamic viscosity. K is constant of the viscometer (mm^2/s^2), ρ is the density of liquid ($\mathrm{mg/mm}^3$) and t is the flow time (s).

(a) (b)

Fig. (3.3): (a) A Brookfield Viscometer Model DV-I+

(b) Glass Capillary Viscometer

3.2.2 Density Apparatus

The densities of the pure liquids of water and phenol have been measured by using the following instruments:

1. The 10 ml pycnometer (density bottle), it is shown in Fig.(3.4.a).
2. HX-Z analytical balance: it used to measure the mass of liquids with accuracy±1%. It is shown in Fig.(3.4.b).

(a) (b)

Fig. (3.4): (a) The 10ml pycnometer (b) The HX-Z analytical balance

The density of a binary mixture of water and phenol has been measured at different temperature for each concentration.

3.2.3 Temperature Apparatus

- Digital Prima long Thermometer:

  It is used to measure the temperature of the samples with accuracy ±1%, the range of the temperature from -50°C up to 200°C (Fig. (3.5.a)).

- Julabo F25-MV Refrigerated and Heating Circulator**:**

  It is used to control the temperature of the samples with accuracy ±1% (Fig.(3.5.b)).

(a) (b)

Fig. (3.5): (a) The Digital Prima Long Thermometer

(b) The Julabo F25-MV Refrigerated and Heating Circulator

3.3 Procedure

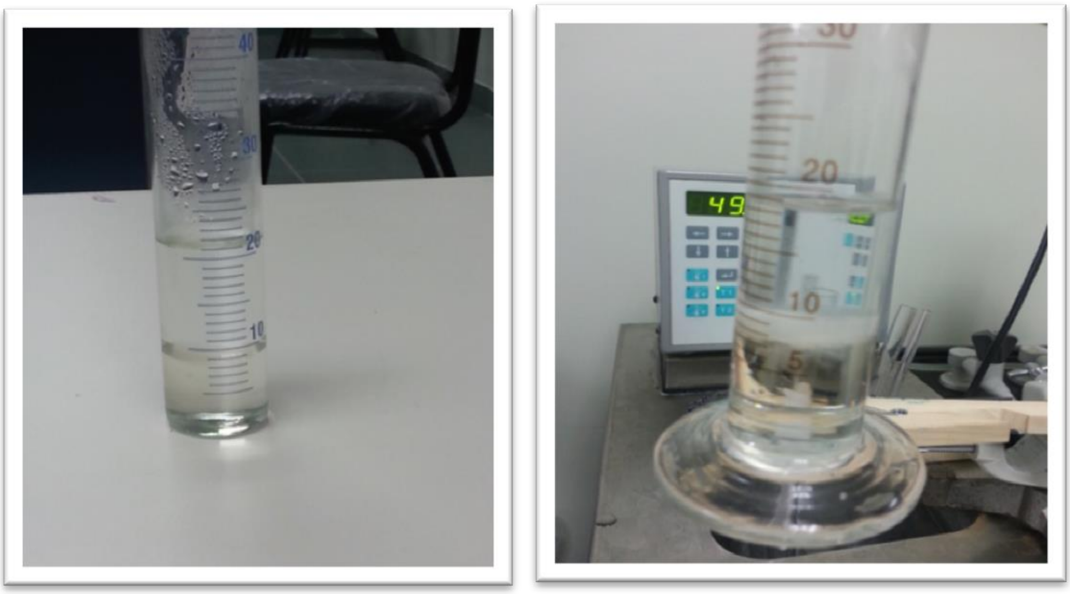
3.3.1 Samples Preparation

The samples of a binary mixture of water and phenol have been prepared according to the following equation:

$$x_{ph} = \frac{m_{ph}}{m_t} \times 100\% = \frac{\rho_{ph}V_{ph}}{\rho_{ph}V_{ph}+\rho_w V_w} \times 100\% \tag{3.3}$$

Where x_{ph} is the concentration by weight of phenol, m_{ph} is the mass of phenol, m_t is the total mass, ρ_{ph} the density of phenol, V_{ph} is the volume of phenol, ρ_w is the density of water and V_w is the volume of water.

Fig.(3.6) shows the water and phenol samples at different concentrations (30.20% and 33.90% by weight of phenol) and different temperatures (75.0 °C and 50.0 °C) respectively.



(a) (b)

Fig.(3.6): Water and phenol mixture (a) at x = 30.20% by weight of phenol and T= 75.0 ℃ (b) at x = 33.90% by weight of phenol and T = 50.0 ℃

3.3.2 Density Measurements

The density of pure liquids water and phenol at room temperature was measured by using the pycnometer. The mass of pycnometer when it is empty and filled with liquids is measured by using HX-Z analytical balance. The density was calculated from the next equation:

$$\rho = \frac{m_1 - m_0}{V} \qquad (3.2)$$

where m_0 is the mass of empty pycnometer in gm, m_1 the mass of filled pycnometer in gm and V is the volume of liquid in cm^3.

The measured densities for distilled water is 0.9987 gm/cm^3 and phenol is 1.1437 gm/cm^3.

3.3.3 Viscosity Measurements[1]

The Brookfield viscometer model DV-I+ was used to measure the viscosity of pure phenol. The sample was put in the jacket of the viscometer; the spindle/speed (a spindle at specific speed) was set at 100 rpm.

The viscosities of the other samples have been measured by using glass capillary viscometer. The time required for the level of liquid to drop between two mark points has been measured by using stopwatch. The viscosity at each temperature was calculated by using this formula (Generalic, 2014):

$$\eta_s = \frac{\eta_w t_s \rho_s}{t_w \rho_w} \tag{3.4}$$

Where the η_s is the viscosity of the sample, ρ_s is the density of the sample, t_s the average flow time of sample, η_w is the viscosity of water, ρ_w is the density of the water and t_w is the average flow time of water.

3.4 Statistical Analysis

The measured data were tabulated and statistically analyzed by using computer software program (Microsoft excel). The relationship between viscosity and temperature of a binary mixture of water and phenol for each concentration was discussed.

The coefficient of determination R^2 was calculated for each curve, which measures the strength of correlation between the viscosity of a binary mixture of water and phenol and the temperature.

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[1] Each measured datum of the shear viscosity of water and phenol samples represents the average of three measurements

Chapter Four

Result

The dynamic shear viscosities for a binary mixture of water and phenol are measured at different temperatures $32.0\ ℃ < T < 75.0℃$ and different concentrations 0.00%, 10.17%, 20.80%, 30.20%, 31.80%, 32.90%, 33.90%, 35.00%, 37.10%, 40.20%, 49.80%, 59.79%, 70.40%, 79.77%, 89.90% and 100.00% by weight of phenol.

4.1 Viscosity – Temperature Dependence

4.1.1 Viscosity of Pure Liquids

The values of dynamic shear viscosity and the natural logarithm of the dynamic shear viscosity of pure water and phenol are calculated in Table (4.1).

Table (4.1): The reciprocal of the absolute temperature and the natural logarithm of the dynamic shear viscosity of the pure water and phenol

T(℃)	$\frac{1}{T}(K^{-1}) \times 10^3$	$\eta_w(cP)^*$	$\ln(\eta_w)$	$\eta_{ph}(cP)^*$	$\ln(\eta_{ph})$
32.0	3.277	0.765	- 0.267	Solid	-
40.0	3.193	0.653	- 0.426	4.280	1.454
45.0	3.143	0.596	- 0.517	3.678	1.302
50.0	3.094	0.547	- 0.603	3.291	1.191
55.0	3.047	0.504	- 0.685	2.934	1.076
57.0	3.028	0.489	- 0.715	2.681	0.986
58.0	3.019	0.481	- 0.731	2.705	0.995
59.0	3.010	0.474	- 0.746	2.735	1.006
60.0	3.001	0.467	- 0.761	2.537	0.931
61.0	2.992	0.460	- 0.776	2.459	0.900
62.0	2.983	0.453	- 0.791	2.421	0.884
63.0	2.974	0.447	- 0.805	2.288	0.827
64.0	2.966	0.440	- 0.820	2.170	0.774
65.0	2.957	0.434	- 0.834	1.849	0.614
66.0	2.948	0.428	- 0.848	1.822	0.599
67.0	2.939	0.422	- 0.862	1.790	0.582
68.0	2.931	0.416	- 0.877	1.719	0.542
69.0	2.922	0.410	- 0.891	1.693	0.526
70.0	2.914	0.404	- 0.906	1.779	0.576
75.0	2.872	0.378	- 0.972	1.604	0.473

*** η_w the dynamic shear viscosity of distilled water.**

*** η_{ph} the dynamic shear viscosity of pure phenol.**

The dynamic shear viscosity of pure materials of water and phenol as a function of temperature are plotted in Fig. (4.1).

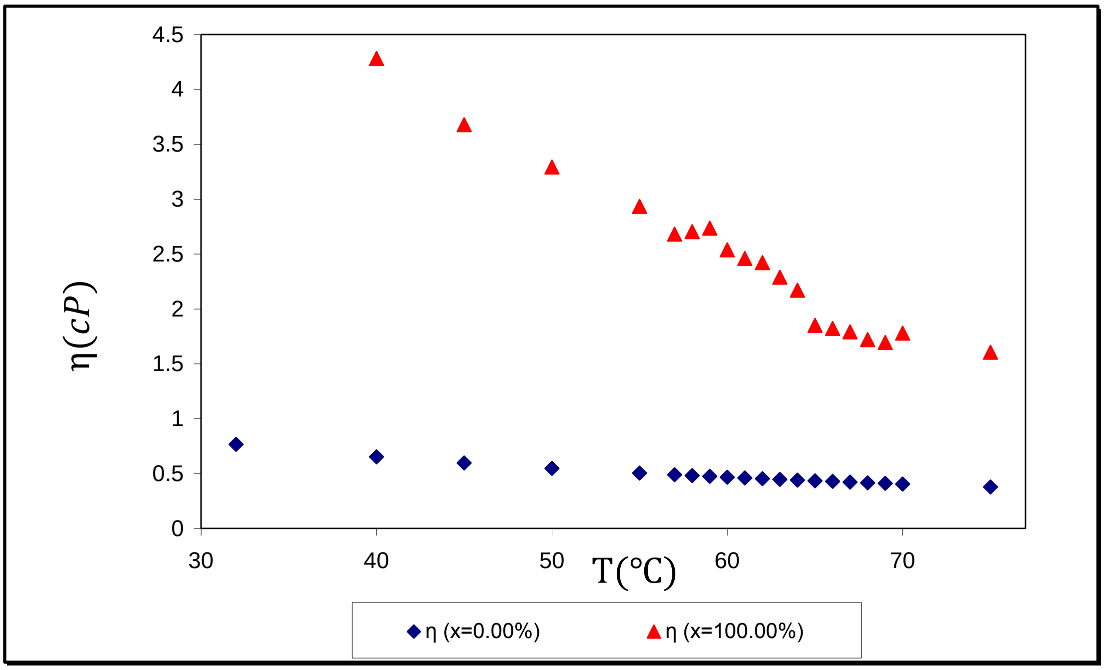

Fig. (4.1): The dynamic shear viscosity of water and phenol as a function of temperature

The data which is presented in table (4.1) and related result in Fig.(4.1) indicate that there is inversely relationship between dynamic shear viscosity and temperature for both of water and phenol liquids. The values of the dynamic shear viscosity and the mass density for phenol liquid are higher than the values of pure water, the measured values of the mass density for pure water, pure phenol and all different concentration samples is calculated and reported in table (A.2) in appendix A.

Fig.(4.2) shows dependency of the natural logarithm of dynamic shear viscosity on the reciprocal of the absolute temperature for water and phenol liquids. The numerical value of the constants A and B in equation (2.5) $[\ln \eta = A + \frac{B}{T}]$ are determined by fitting a linear relation between $\ln(\eta)$ and $\frac{1}{T}$.

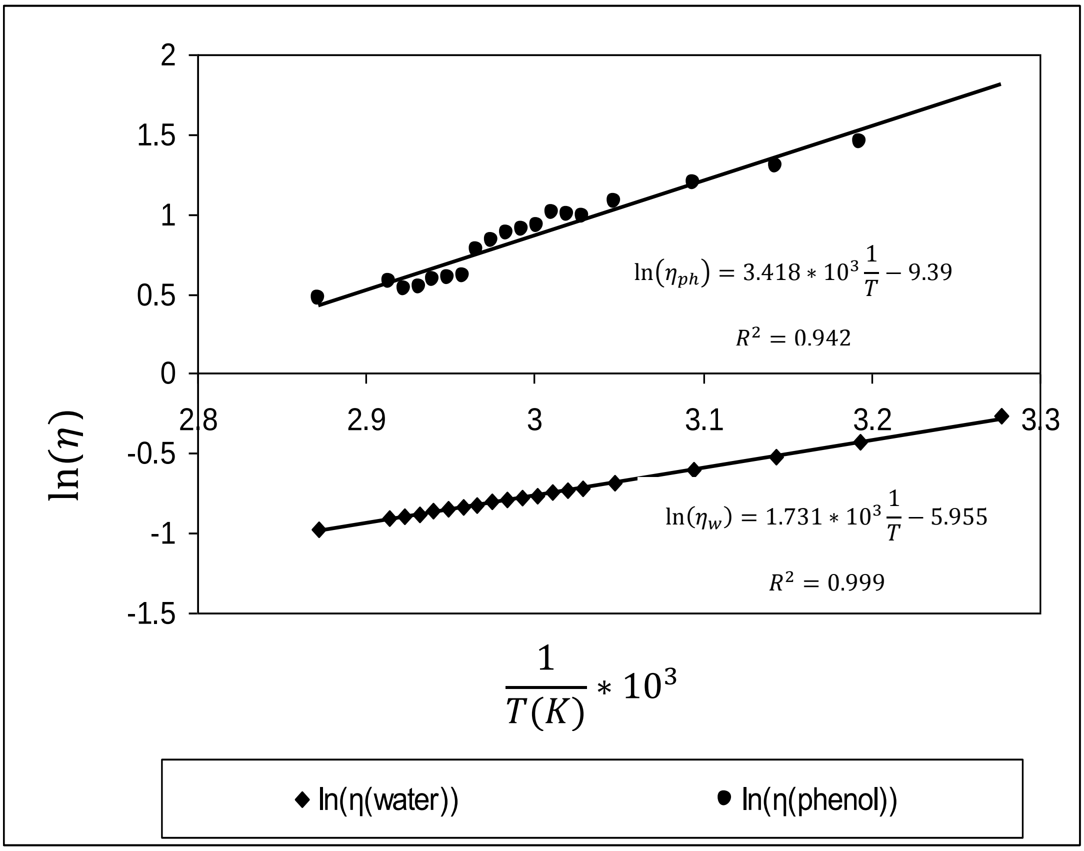

Fig. (4.2): The logarithm of the dynamic shear viscosity as a function of the reciprocal of absolute temperature for water and phenol

The plots show that $\ln(\eta)$ - $\frac{1}{T(K)}$ linearly depend for pure phenol liquid at $32℃ \leq T \leq 75℃$. The mathematical relationships can be presented by,

$$\ln(\eta_{ph}) = 3.418 \times 10^3 \frac{1}{T} - 9.39 \qquad (4.1)$$

Where A = - 9.939 and B = 3.418×10^3 °C. The correlation coefficient $R^2 = 0.942$.

The linear relation between $\ln(\eta)$ and $\frac{1}{T(K)}$ for distilled water is described as:

$$\ln(\eta_w) = 1.731 \times 10^3 \frac{1}{T} - 5.955 \qquad (4.2)$$

Where A = - 5.955 and B = 1.731×10^3 ℃. The correlation coefficient $R^2 = 0.999$.

In equations (4.1) and (4.2), η_w and η_{ph} are the dynamic shear viscosity for water and phenol respectively and T(K) is the absolute temperature.

These analysis shows that the dynamic shear viscosity of water and phenol exponentially increase with increasing temperature.

4.1.2 Viscosity of Binary Liquid Mixture Samples

The results of the dynamic shear viscosities of a binary mixture of water and phenol at different temperatures and concentrations are given in Table (A.1).

The behavior of the dynamic shear viscosities of water – phenol samples are presented in Fig.(4.3), Fig.(4.4) and Fig.(4.5). The dynamic shear viscosity for all samples decreases as the temperature increase. There are some fluctuations of viscosity values at certain temperatures due to contamination of viscometer and air bubbles that leads to increase the measured flow time and the viscosity value.

The one phase solution (homogenous solution) of water and phenol mixture is observed experimentally at concentration equals to 33.90% by weight of phenol.

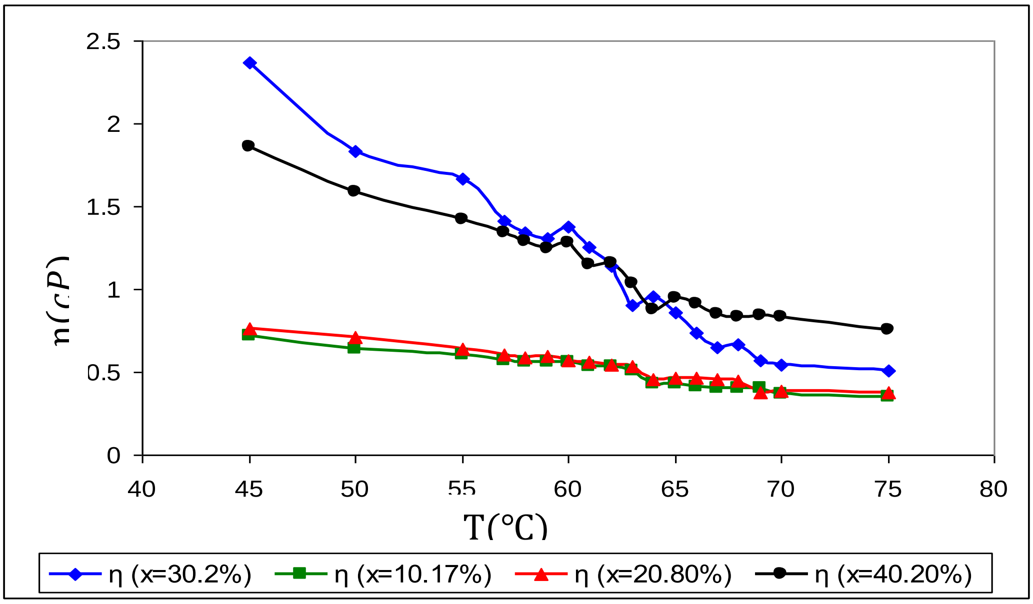

Fig. (4.3): The dynamic shear viscosity of water-phenol mixture as a function of temperature at concentrations 10.17%, 20.80%, 30.20% and 40.20% by weight of phenol

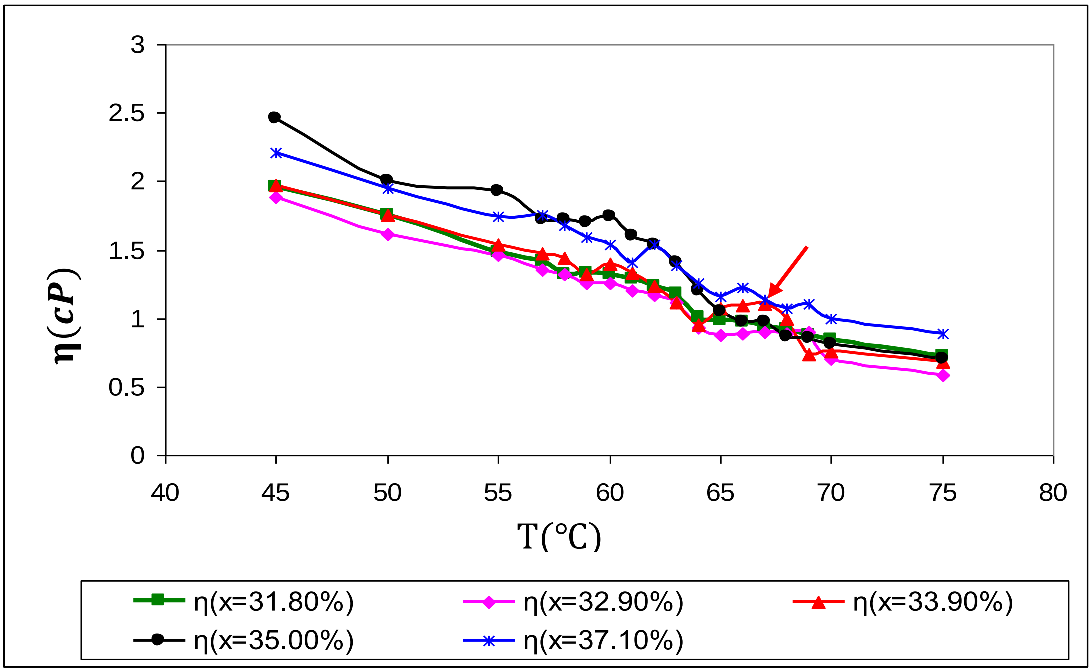

Fig. (4.4): The dynamic shear viscosity of water-phenol mixture as a function of temperature at concentrations 31.80%, 32.90%, 33.90%, 35.00% and 37.10% by weight of phenol

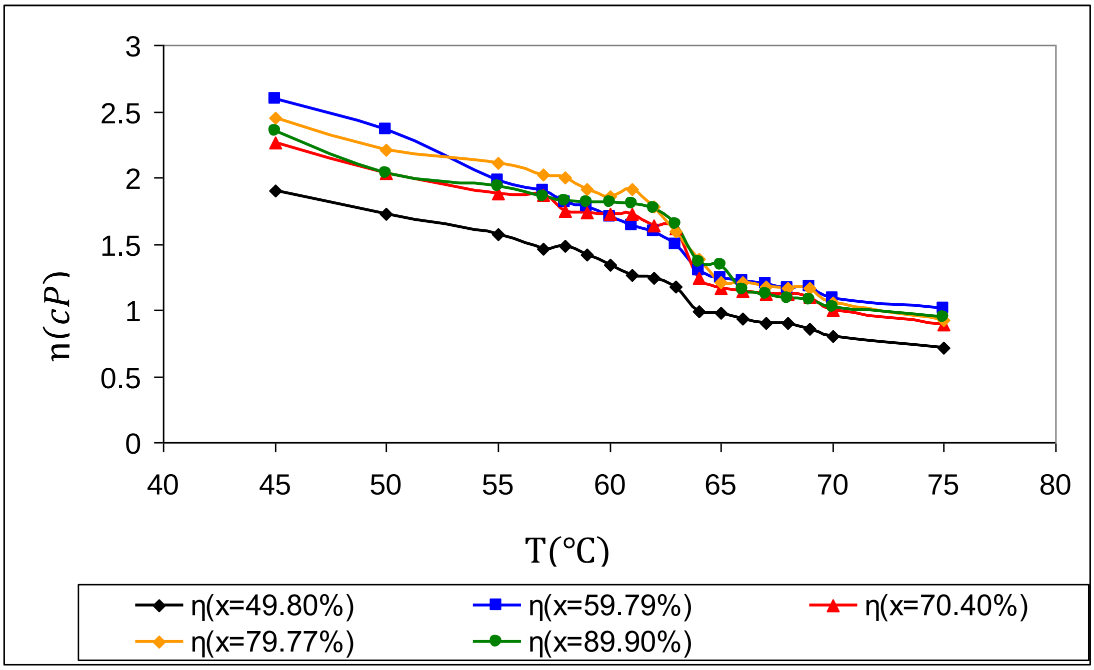

Fig. (4.5): The dynamic shear viscosity of water-phenol mixture as a function of temperature at concentrations 49.80%, 59.79% 70.40%, 79.77% and 89.90% by weight of phenol .

The anomaly behavior was noticed in Fig.(4.4) and more clear in Fig.(4.6) which represented the relationship between dynamic shear viscosity and temperature at concentrations 32.90%, 33.90% and 35.00% by weight of phenol and temperature range from 60 °C up to 75 °C.

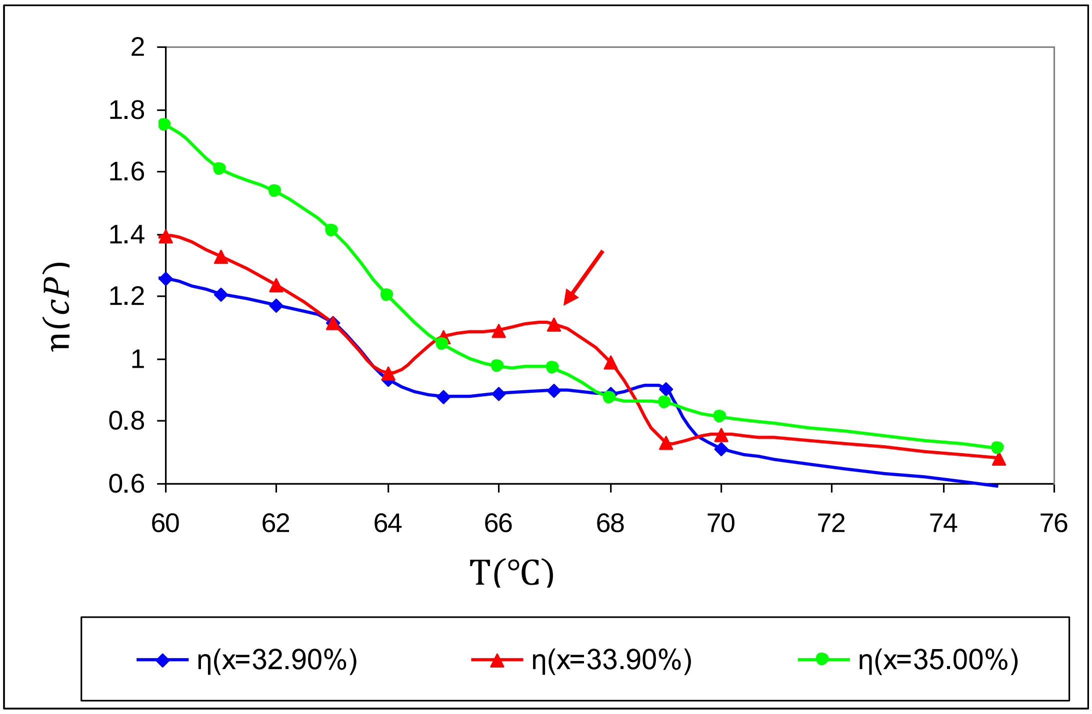

Fig. (4.6): The dynamic shear viscosity of water - phenol mixture as a function of temperature at concentrations 32.90%, 33.90% and 35.00% by weight of phenol

Fig.(4.4) and Fig.(4.6) show anomaly at concentration 33.90% by weight of phenol and temperature 67°C, this point is considered to be a critical point where water and phenol liquids become one solution (one phase).

The critical concentration and critical temperature of a binary mixture of water and phenol were measured experimentally by Krishnan (Krishnan, 1935) and determined to be 34% by weight of phenol and 69 ℃ respectively. Our values of the critical points agree with the Krishnan measured values.

Our result was also in good agreement with Howell ($T_c = 66.4$ ℃, $x_c = 34.6\%$ by weight of phenol) and Campbell ($T_c = 66.8$ ℃, $x_c = 34.5\%$ by weight of phenol) experimental results of critical temperature and critical concentration (Howell, 1932; Campbell and Campbell, 1937).

The characteristic thermal energy (kT) is equal to the phonon vibration energy ($\hbar\omega$) thus during thermal transport, the phonon frequency is

$$\omega = \frac{\mathrm{kT}}{\hbar} = 4.468 \times 10^{13}\,\mathrm{Hz}.$$

4.2 Kinematic Viscosity near Critical Point

The measured values of the mass density, kinematic and dynamic shear viscosity of a binary mixture of water and phenol at the critical concentration and around critical temperature are given in Table (4.2).

Table (4.2): The measured mass density, dynamic and kinematic shear viscosity for a binary mixture of water and phenol at critical concentration at different temperatures

T(℃)	60.0	61.0	62.0	63.0	64.0	65.0	66.0	67.0	68.0	69.0
$\eta(cP)$	1.393	1.329	1.235	1.115	0.955	1.070	1.090	1.108	0.991	0.731
$\rho(\frac{gm}{cm^3})$	0.9519	0.9438	0.9333	0.9225	0.9137	0.9180	0.9208	0.8952	0.8973	0.9260
$\nu(cSt)$	1.463	1.408	1.324	1.208	1.045	1.166	1.184	1.238	1.104	0.790

The measured mass density (Table 4.2) of water-phenol sample at critical concentration ($x_c = 0.339$ by weight of phenol) decreases as the temperature increases because the average kinetic energy of water-phenol molecules increases with temperature. At temperatures above 66.0 ℃ there is anomaly behavior.

The kinematic and dynamic shear viscosities of a binary mixture of water and phenol have been plotted as a function of temperature at critical concentration ($x_c = 33.90\%$ by weight of phenol) as shown in Fig.(4.7).

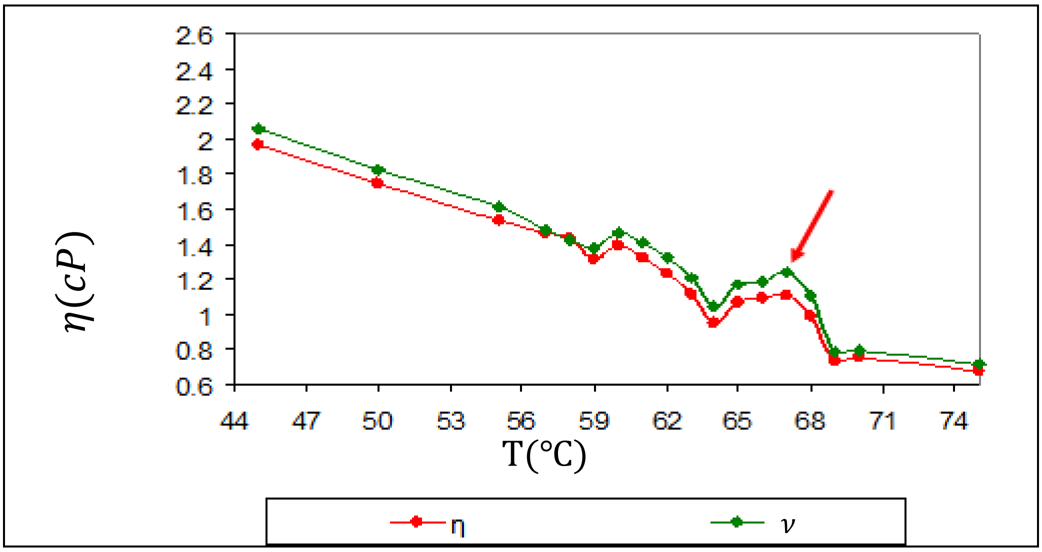

Fig. (4.7): The dynamic and kinematic shear viscosity of water-phenol mixture as a function of temperature at critical concentration

The kinematic shear viscosity curve (Fig.(4.7)) shows anomalous behavior around the critical point. The value of kinematic shear viscosity at critical point ($\nu =$ 1.238 cSt) is higher than the value of the dynamic shear viscosity ($\eta =$ 1.108 cP).

4.3 Viscosity – Concentration Dependence

The dynamic shear viscosities of water and phenol binary mixture as a function of concentration have been plotted in Fig. (4.8) for six different temperatures 32 °C, 40°C, 50°C, 55°C, 60°C and 67°C.

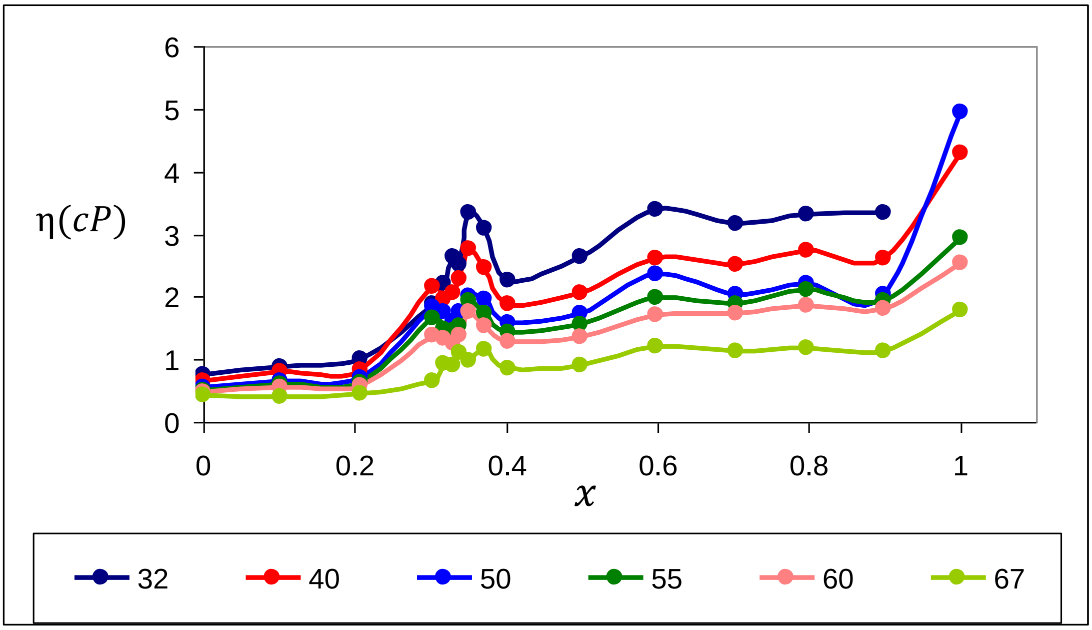

Fig. (4.8): The dynamic shear viscosity of water-phenol mixture as a function of concentration at temperatures 32.0, 40.0, 50.0, 55.0, 60.0 and 67.0 °C

In general, the dynamic shear viscosity η of a binary mixture of water and phenol increases as the concentration (by weight of phenol) increases, except in the region around the critical concentration ($x \sim 0.339$ by weight of phenol) there is anomaly behavior.

4.4 The Mode - Coupling Theory

The mode-coupling theory of Kawasaki, Perl and Ferrell (Kawasaki, 1970; Perl and Ferrell, 1972) can be used to determine the background viscosity η_0, the constant A, Debye momentum cutoff q_D and the intermolecular force range L at critical concentration and above critical temperature.

4.4.1 The Background Viscosity η_0

The measured data of the dynamic shear viscosity at critical concentration $x_c = 33.90\%$ and above critical temperature $67℃ < \mathrm{T} \leq 68.7℃$ are given in Table (4.3).

Table (4.3): The dynamic shear viscosity measurements at critical concentration and above critical temperature

T(℃)	**T(K)**	$\boldsymbol{T}$	$\boldsymbol{\eta(cP)}$	**ln (t)**	**ln ($\boldsymbol{\eta}$)**
67.2	340.35	0.000588	0.928	-7.43882	-0.07383
67.4	340.55	0.001176	0.911	-6.74568	-0.09316
67.6	340.75	0.001764	0.900	-6.34021	-0.10427
67.8	340.95	0.002352	0.889	-6.05253	-0.11716
68.4	341.55	0.004116	0.862	-5.49291	-0.14770
68.7	341.85	0.004998	0.847	-5.29876	-0.16591

The linear relationship between the natural logarithmic of dynamic shear viscosity $\ln(\eta)$ and the natural logarithmic of reduced temperature ln (t) has been represented in Fig. (4.9).

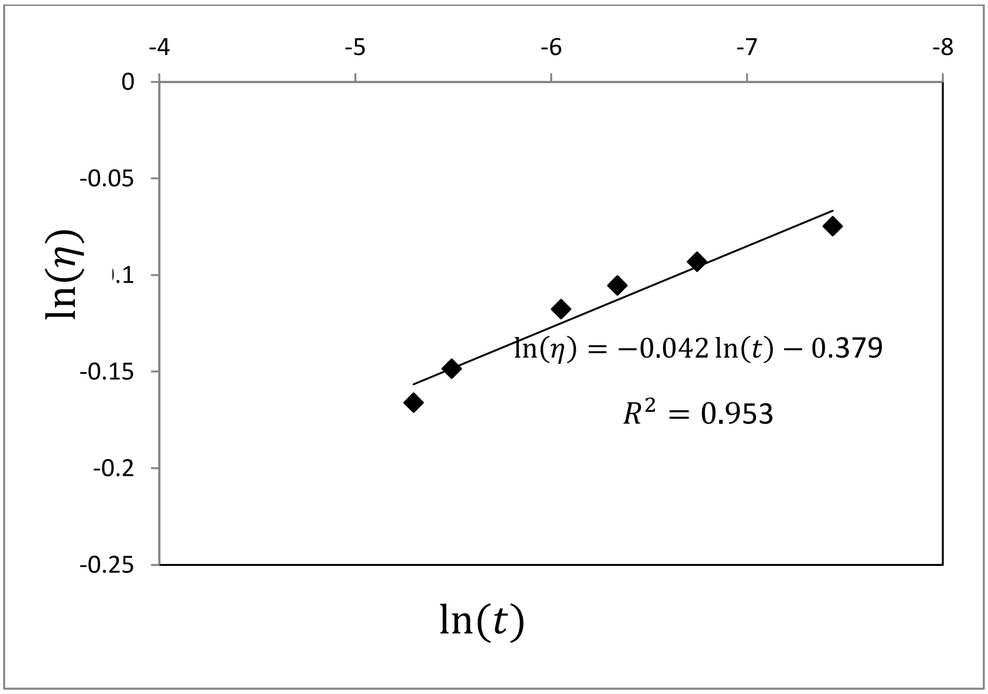

Fig.(4.9): ln(η) Vs. ln(t)

According to the equation $\eta = \eta_0 t^{-x_\eta \nu}$, the natural logarithmic of dynamic shear viscosity can be written as the follows:

$$\ln(\eta) = -x_\eta \nu \ln(t) + \ln(\eta_0) \tag{4.3}$$

By comparing equation (4.3) with the result in Fig.(4.9), the value of critical exponents constant $x_\eta \nu$ is presented by the slope of the curve which equal to 0.042. It is consistent with the theoretical value ($x_\eta \nu = 0.04$) (Klein and Woermann, 1978). The intercept of the curve represented the natural logarithmic of the background viscosity $\ln(\eta_0) = -0.379$, the value of the background viscosity (noncritical part of the measured shear viscosity) equals to 0.684 cP. This means that above $\eta = 0.684$ cP the long wavelength modes was dissipate (page 29).

The power law of the dynamic shear viscosity equation (2.7) and the theoretical value of critical exponent are used to calculated the background viscosity by plotting η Vs. $t^{-0.04}$, as shown in Fig.(4.10).

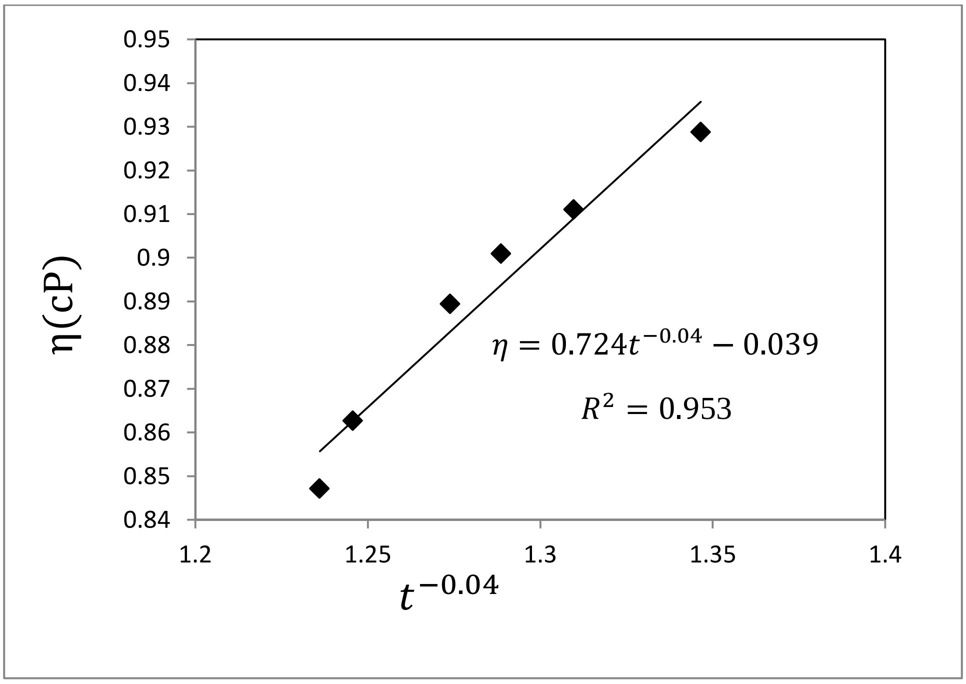

Fig.(4.10): The measured dynamic shear viscosity at critical concentration and above critical temperature Vs. $t^{-0.04}$

The linear relationship between η and $t^{-0.04}$ has been represented in Fig.(4.10). The value of the background viscosity of a binary mixture of water and phenol has been calculated from the slope $\eta_0 = 0.724$ cP, this value is much close to the η_0 value which equals to 0.684cP that found by using Fig.(4.9).

4.4.2 The Constant A

The value of constant A at critical concentration and above critical temperature can be determined by using the mode coupling theory of Kawasaki, Peral and Ferrell, equation (2.6).

Where $\frac{\Delta\eta}{\eta}$ is the relative anomalous shear viscosity, q_D is the Debye momentum cutoff and ξ is the average correlation length of the concentration fluctuations which is given by the power law, equation 2.8: ($\xi = \xi_0 t^{-\nu}$), with ξ_0 being the correlation length and equals to 2.2 Å for water and phenol binary mixture (Abdelraziq, 2015).

The values of the anomalous shear viscosity and the average correlation length of a binary mixture of water and phenol have been calculated at critical concentration and above critical temperature; the results are given in Table (4.4).

Table (4.4): The dynamic shear viscosity measurements and the logarithm of the average correlation length at critical concentration and above critical temperature

T(°C)	T(K)	t	ξ	$\ln(\xi)$	η	$\frac{\Delta\eta}{\eta}$
67.2	340.35	0.000588	257.0589	5.549305	0.928	0.263
67.4	340.55	0.001176	164.9580	5.105691	0.911	0.248
67.6	340.75	0.001764	127.2550	4.846193	0.900	0.240
67.8	340.95	0.002352	105.8557	4.662077	0.889	0.230
68.4	341.55	0.004116	73.98946	4.303923	0.862	0.206
68.7	341.85	0.004998	65.34381	4.179663	0.847	0.191

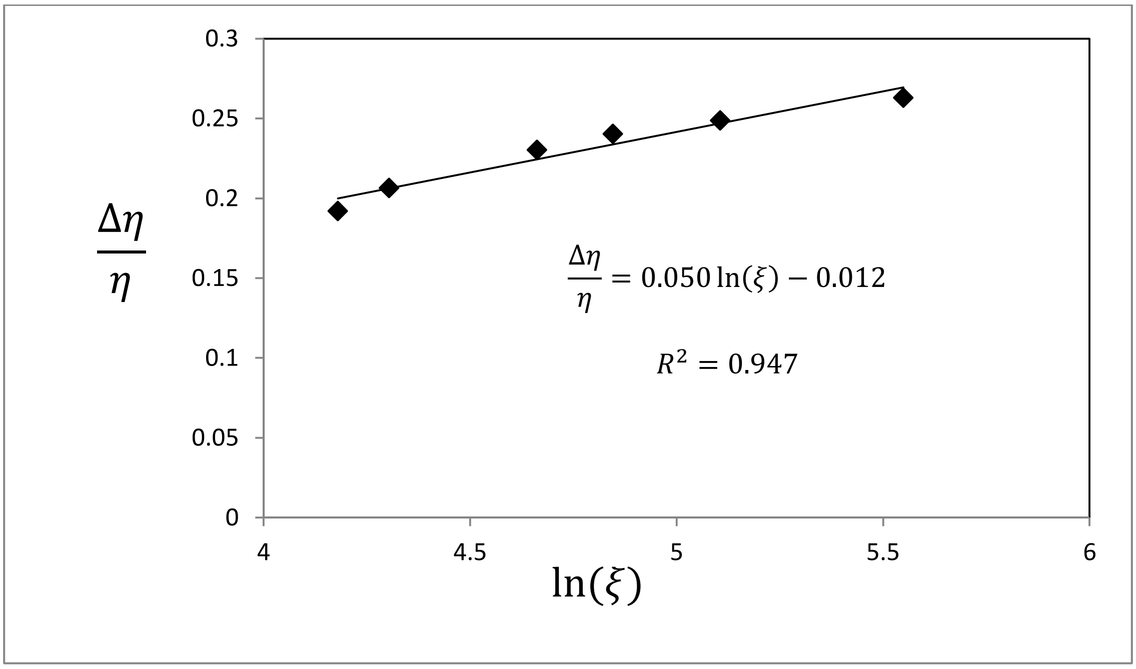

Fig.(4.11): The anomalous shear viscosity at critical concentration and above critical temperature Vs. the logarithmic of average correlation length

Fig.(4.11) shows the linear relationship between the anomalous shear viscosity and the logarithmic of the average correlation length with slope equals to A value, which is 0.050.

The value of the MCT constant A was calculated for many different binary liquid mixtures, for example, Swinney and Henry, D'Arrigo and his team and Abdelraziq group determined the value of A experimentally as 0.054, 0,056 and 0,058 respectively (Swinney and Henry, 1973; D'Arrigo *et al*, 1979; Abdelraziq *et al*, 1996).

The measured value of the MCT constant A is in a good agreement with the theoretical value which equals to 0.054 (D'Arrigo *et al*, 1997). The value of the constant A doesn’t depend on the type of mixtures.

4.4.3 Debye Momentum Cutoff q_D

The value of Debye momentum cutoff (the upper cutoff wave length) q_D can be calculated by using the intercept of Fig. (4.11) and the measured value of the MCT constant A. By using equation (2.6), the value of q_D is $0.7866\ \text{Å}^{-1}$, this mean that all modes with momentum value greater than $0.7866\ \text{Å}^{-1}$ are not allowed. Above this value the mixture will lose its properties and separated into multiphase mixture.

Determining Debye momentum cutoff of water-phenol binary mixture supports the MCT of the critical viscosity.

4.4.4 The Intermolecular Force Range L (Debye Parameter)

The intermolecular force range L (Debye parameter) has been determined by using equation (2.9): ($L = 2.45\ \xi_0 T_c^{(\gamma-1)/2}$), where ξ_0 is the correlation length equals to 2.2 Å (Abdelraziq, 2015), T_c (= 340.15 K) is the critical temperature of a binary mixture of water and phenol and γ is the critical exponent ($\gamma = 1.25$).

Substituting the values of ξ_0, T_c and γ in equation (2.9), L value is found to be 11.169 Å. The large value of the Debye parameter L indicates that the mutual forces between water and phenol molecules in a binary mixture considered as a weak attractive force.

The origin of this force is due to interaction between positive and negative ions in addition similar type of ions that cause columbic type force. Particularly there exist attractive columbic forces between phenol hydrogen and water oxygen also between phenol oxygen and water hydrogen that are clearly mention in section 3.1.2.

4.5 Two-Scale-Factor Universality

Specific heat capacity at constant pressure at critical temperature can be calculated by using the two scale factor universality (Hohenberg *et al*, 1976).

$$R_\zeta = \xi_0[\alpha \frac{\rho_c C_{pc}}{K_B}]^{\frac{1}{d}} = 0.270 \tag{4.3}$$

Where R_ζ is the universal quantity, α is the critical exponent equals to 0.11, ρ_c is the mass density at the critical temperature ($\rho_c = 0.8952\ \frac{gm}{cm^3}$ for binary mixture of water and phenol, calculated in this work), K_B = 1.3806× $10^{-23} J/K$ is the Boltzmann's constant, d = 3 is the dimension and ξ_0 is the correlation length equals to 2.2 Å for water – phenol binary mixture.

Substituting all the previous parameters in equation (4.3). The value of specific heat at constant pressure at critical temperature equals to 259.16 $\frac{J}{kg.K}$.

Chapter Five

Discussion

The critical temperature and critical concentration of a binary liquid mixture of water and phenol were measured, the results were $T_c = 67.0\ ℃$ and $x_c = 0.3390$ by weight of phenol, these measured values of critical points are agreed with Howell, Krishnan and Campbell experimental results (Howell, 1932; Krishnan, 1935; Campbell and Campbell, 1937).

The background shear viscosity η_0 for a binary liquid mixture of water and phenol around critical points was calculated to be 0.684 cP . This value is larger than the background shear viscosity of nitroethane-3-methylpentane ($\eta_0 = 0.358$ cP) (Abdelraziq, 2002), and it is smaller than the values of the background viscosity of CCl_4-coconut oil ($\eta_0 = 2.59$ cP) (Kittany, 2014), benzene-coconut oil ($\eta_0 = 1.30$ cP) (Abdo, 2014) and nitrobenzene-n-heptane ($\eta_0 = 0.70$ cP)(Abdelraziq *et al*, 1997) binary mixtures. The difference in η_0 value are due to the physical properties (density, viscosity and intermolecular force) of each binary mixture.

The value of the MCT constant A at critical concentration and above critical temperature was found to be 0.050 which is in a good agreement with the theoretical value (A = 0.054) and it doesn't depend on the type of mixture. The Debye momentum cutoff was calculated to be 0.7866 $Å^{-1}$, the values of background shear viscosity, Debye momentum cutoff and the constant A for some binary mixtures are shown in table (5.1).

The intermolecular force range L (Debye parameter) was calculated to be 11.17 Å. This large value indicates that the mutual force between water and phenol molecules in a binary mixture considered as a week attractive force.

Table (5.1): The measured values of background shear viscosity and Debye momentum cutoff of some binary liquid mixtures

Binary liquid mixture	Background viscosity (η_0)	Debye momentum cutoff (q_D)	Intermolecular force range (L)	Constant A
Water – phenol	0.684 cP	0.7866 $Å^{-1}$	11.17 Å	0.0500
CCl_4 - coconut oil	2.590 cP	0.1260 $Å^{-1}$	5.50 Å	0.0538
benzene - coconut oil	1.300 cP	0.3580 $Å^{-1}$	9.90 Å	0.0520
Nitroethane - 3-methylpentane	0.358 cP	2.0000$Å^{-1}$	9.30 Å	0.054
nitrobenzene - n-heptane	0.700 cP	0.2750 $Å^{-1}$	10.65 Å	0.058

The measured and calculated parameters for water and phenol binary mixture which are found in this research and other works are summarized in table (5.2).

Table (5.2): Summary of the measured results in this work and previous works

Parameter	This work	Previous works
T_c (°C)	67. 0	69.0^a 66.4^b 66.8^c
x_c	0.3390	$0.340^{\,a}$ $0.346^{\,b}$ $0.345^{\,c}$
η_0 (cP)	0.684	-
q_D ($Å^{-1}$)	0.7866	-
L (Å)	11.17	-
C_{pc} ($\frac{J}{k_g.K}$)	259.16	-
A	0.050	-

Where, $^{(a)}$:(Krishnan, 1935), $^{(b)}$: (Howell, 1932), $^{(c)}$:(Campbelland Campbell,1937).

Chapter Six

Conclusions and Future Work

The dynamic shear viscosity around critical points (at critical concentration and above critical temperature) of a binary mixture of water and phenol was studied by using the mode coupling theory.

The inversely relationships between dynamic shear viscosity and temperature for water and phenol samples of different concentrations were studied. The anomalous behavior of the dynamic shear viscosity for this binary liquid mixture was detected at temperature equals to 67.0 ℃ and concentration equals to 33.90% by weight of phenol.

The values of the background viscosity η_0, the intermolecular force range and the MCT constant A were determined for water and phenol binary mixture the results were 0.684 cP, 11.17 Å and 0.050 ± 0.005 respectively. The Debye momentum cutoff was found to be 0.7866 $Å^{-1}$, this mean that all modes with momentum value greater than 0.7866 $Å^{-1}$ are not allowed and above this value the mixture will lose its properties and separated into multiphase mixture.

The critical amplitude of specific heat at constant pressure was calculated using the two scale factor universality, the result was $C_{pc} = 259.16 \frac{J}{Kg.K}$.

As a future work, the thermal expansion coefficient, heat capacity at constant pressure, refractive index and susceptibility for a binary mixture of water and phenol can be measured at critical concentration and above critical temperature. In addition, the dynamic shear viscosity of different types of binary liquid mixtures can be measured around critical point, it can be used to find the MCT parameters (the background viscosity η_0, the constant A, Debye momentum cutoff q_D and the intermolecular force range L or Debye parameter).

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Appendix A

The measured data of the dynamic shear viscosity and the mass density of a water and phenol samples at different concentrations (from 0.00% to 100.00% by weight of phenol) and different temperatures ($32℃ \leq T \leq 75℃$) are listed in tables (A.1) and (A.2)

Table (A.1): The dynamic shear viscosity measurements for a binary mixture of water and phenol at different temperatures and concentrations

T (℃)	η(cP) 0.00%	η(cP) 10.17%	η(cP) 20.80%	η(cP) 30.20%	η(cP) 31.80%	η(cP) 32.90%	η(cP) 33.90%	η(cP) 35.00%	η(cP) 37.10%	η(cP) 40.20%	η(cP) 49.80%	η(cP) 59.79%	η(cP) 70.40%	η(cP) 79.77%	η(cP) 89.90%	η(cP) 100.0%
32	0.765	0.871	0.995	1.875	2.204	2.625	2.504	3.330	3.095	2.255	2.648	3.396	3.157	3.317	3.334	Solid*
40	0.653	0.806	0.839	2.147	1.959	2.058	2.285	2.758	2.460	1.879	2.070	2.600	2.502	2.735	2.614	4.280
45	0.596	1.074	1.139	3.549	2.940	2.830	2.950	3.684	3.306	2.794	2.848	3.994	3.391	3.671	3.534	3.678
50	0.547	0.644	0.711	1.832	1.758	1.617	1.750	2.008	1.948	1.586	1.724	2.359	2.029	2.211	2.037	4.937
55	0.504	0.605	0.638	1.664	1.487	1.463	1.537	1.923	1.738	1.418	1.568	1.979	1.876	2.104	1.938	2.934
57	0.489	0.569	0.604	1.408	1.413	1.351	1.467	1.722	1.757	1.340	1.463	1.896	1.863	2.025	1.859	2.681
58	0.481	1.015	1.071	2.433	2.390	2.399	2.608	3.128	3.052	2.344	2.700	3.292	3.175	3.637	3.307	2.705
59	0.474	0.558	0.592	1.306	1.335	1.255	1.319	1.698	1.592	1.249	1.421	1.775	1.736	1.909	1.815	4.968
60	0.467	0.559	0.574	1.374	1.318	1.256	1.393	1.747	1.536	1.283	1.343	1.703	1.720	1.857	1.182	2.537
61	0.460	0.538	0.557	1.251	1.291	1.206	1.329	1.605	1.407	1.150	1.268	1.641	1.726	1.909	1.801	2.459
62	0.453	0.533	0.540	1.136	1.232	1.172	1.235	1.535	1.540	1.156	1.241	1.588	1.636	1.781	1.768	2.421
63	0.447	0.508	0.538	0.900	1.181	1.113	1.115	1.409	1.386	1.037	1.174	1.498	1.611	1.596	1.649	2.288
64	0.440	0.432	0.460	0.952	1.010	0.934	0.955	1.199	1.253	0.875	0.983	1.294	1.243	1.380	1.357	2.170
65	0.434	0.426	0.466	0.855	0.981	0.876	1.070	1.046	1.163	0.948	0.980	1.244	1.165	1.212	1.343	1.849
66	0.428	0.415	0.468	0.735	0.970	0.888	1.090	0.973	1.228	0.908	0.931	1.215	1.142	1.203	1.149	1.822
67	0.422	0.407	0.451	0.644	0.940	0.900	1.108	0.970	1.141	0.851	0.906	1.201	1.120	1.172	1.123	1.790
68	0.416	0.402	0.445	0.664	0.924	0.889	0.991	0.871	1.075	0.834	0.895	1.160	1.116	1.163	1.088	1.719
69	0.410	0.400	0.376	0.567	0.878	0.903	0.731	0.857	1.104	0.843	0.856	1.177	1.103	1.168	1.074	1.693
70	0.404	0.369	0.383	0.548	0.847	0.708	0.757	0.814	1.000	0.829	0.804	1.090	1.000	1.052	1.018	1.779
75	0.378	0.353	0.375	0.505	0.720	0.589	0.681	0.708	0.885	0.750	0.714	1.012	0.888	0.925	0.946	1.604

* Concentration by weight of phenol

* phenol viscosities measured above T=40℃, under this temperature the liquid phenol becomes solid

Table (A.2): The mass density measurements for a binary mixture of water and phenol at different temperatures and concentrations

T (℃)	$\rho(\frac{gm}{cm^3})$0.00%	$\rho(\frac{gm}{cm^3})$10.17%	$\rho(\frac{gm}{cm^3})$20.80%	$\rho(\frac{gm}{cm^3})$30.20%	$\rho(\frac{gm}{cm^3})$31.80%	$\rho(\frac{gm}{cm^3})$32.90%	$\rho(\frac{gm}{cm^3})$33.90%	$\rho(\frac{gm}{cm^3})$35.00%	$\rho(\frac{gm}{cm^3})$37.10%	$\rho(\frac{gm}{cm^3})$40.20%	$\rho(\frac{gm}{cm^3})$49.80%	$\rho(\frac{gm}{cm^3})$59.79%	$\rho(\frac{gm}{cm^3})$70.40%	$\rho(\frac{gm}{cm^3})$79.77%	$\rho(\frac{gm}{cm^3})$89.90%	$\rho(\frac{gm}{cm^3})$100.0%
32	0.9950	0.9811	0.9123	0.9640	0.9882	0.9441	0.9414	1.0490	0.9518	0.9454	0.9817	0.9877	0.9963	1.0378	1.0347	Solid*
40	0.9921	0.9810	0.9070	0.9639	0.9870	0.9420	0.9493	1.0571	0.9510	0.9462	0.9920	0.9807	0.9959	1.0368	1.0315	1.0440
45	0.9910	0.9801	0.9139	0.9691	0.9840	0.9546	0.9559	1.0814	0.9536	0.9386	0.9986	0.9792	0.9935	1.0357	1.0294	1.0530
50	0.9880	0.9770	0.9210	0.9650	0.9839	0.9524	0.9610	1.0721	0.9606	0.9593	1.0253	1.0158	0.9890	1.0322	1.0284	1.0534
55	0.9851	1.0008	0.9427	0.9580	0.9820	0.9464	0.9524	1.0740	0.9539	0.9602	1.0193	1.0151	1.0061	1.0281	1.0317	1.0527
57	0.9847	0.9670	0.9236	0.9653	0.9914	0.9410	0.9876	1.0531	0.9601	0.9658	0.9667	1.0161	1.0059	1.0250	1.0352	1.0521
58	0.9842	0.9783	0.9481	0.9707	0.9510	0.9563	1.0065	1.0611	0.9545	0.9555	1.0079	1.0108	0.9978	1.0174	1.0293	1.0514
59	0.9837	0.9752	0.9477	0.9650	1.000	0.9364	0.9531	1.0795	0.9504	0.9554	0.9980	1.0027	0.9928	1.0223	1.0335	1.0552
60	0.9832	0.9880	0.9303	0.9786	0.9950	0.9452	0.9519	1.0680	0.9559	0.9542	0.9869	0.9900	0.9972	1.0169	1.0316	1.0242
61	0.9826	0.9805	0.9322	0.9775	0.9920	0.9375	0.9438	1.0555	0.9491	0.9471	0.9871	0.9814	1.0045	1.0769	1.0355	1.0229
62	0.9821	0.9770	0.9229	0.9712	0.9840	0.9386	0.9333	1.0555	0.9384	0.9591	0.9921	0.9718	1.0095	1.0312	1.0343	1.0317
63	0.9816	0.9758	0.9488	0.9710	0.9890	0.9469	0.9225	1.0555	0.9566	0.9451	0.9865	0.9686	1.0126	1.0238	1.0327	1.0305
64	0.9810	0.9708	0.9565	0.9820	0.9860	0.9445	0.9137	1.0555	0.9629	0.9495	0.9748	0.9725	1.0001	1.0226	1.0338	1.0295
65	0.9805	0.9775	0.9648	0.9726	0.9610	0.8975	0.9180	0.9368	0.9543	0.9558	0.9889	0.9607	0.9913	1.0168	1.0324	1.0294
66	0.9800	0.9878	0.9741	0.9828	0.9860	0.9471	0.9208	0.9368	0.9370	0.9478	0.9750	0.9667	1.0017	1.0175	1.0301	1.0285
67	0.9790	0.9860	0.9452	0.9750	0.9890	0.9318	0.8952	0.9368	0.9396	0.9378	0.9760	0.9700	0.9935	1.0117	1.0341	1.0374
68	0.9789	0.9830	0.9660	0.9676	0.9920	0.9257	0.8973	0.9368	0.9462	0.9331	0.9666	0.9640	1.0012	1.0206	1.0274	1.0367
69	0.9780	0.9804	0.8884	0.9621	0.9850	0.9215	0.9260	0.9368	0.9253	0.9301	0.9786	0.9738	1.0003	1.0114	1.0286	1.0357
70	0.9777	0.9243	0.9430	0.9448	0.9750	0.8542	0.9549	0.9325	0.9290	0.9364	0.9825	0.9612	0.9993	1.0164	1.0275	1.0493
75	0.9748	0.9860	0.9521	0.9478	0.9690	0.8912	0.9520	0.9435	0.9510	0.9390	0.9648	0.9835	0.9977	1.0183	1.0261	1.0484

* phenol viscosities measured above T=40℃, under this temperature the liquid phenol becomes solid

جامعة النجاح الوطنية

كلية الدراسات العليا

اعتماد اللزوجة على التركيز و درجة الحرارة للخليط الثنائي للماء والفينول في نظرية الإزدواج

إعداد

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إشراف

أ.د. عصام راشد عبد الرازق

د. محمد أبو جعفر

قدمت هذه الأطروحة استكمالا لمتطلبات الحصول على درجة الماجستير في الفيزياء بكلية الدراسات العليا في جامعة النجاح الوطنية في نابلس – فلسطين

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إشراف

أ.د. عصام راشد عبد الرازق

د. محمد أبو جعفر

الملخص

في هذا البحث تم قياس معامل اللزوجة للخليط الثنائي الماء ($\mathrm{H_2O}$) و الفينول ($\mathrm{C_6H_5OH}$) عند درجات حرارة مختلفة ($32.0\ ^\circ\mathrm{C} \le T \le 75.0\ ^\circ\mathrm{C}$) و تراكيز مختلفة (من0.00%إلى 100.00%من وزن الفينول) باستخدام مقياس اللزوجة الزجاجي الشعري (glass capillary viscometer) و مقياس اللزوجة الرقمي (Brookfield viscometer model DV−I+). لقد تم تحديد درجة الحرارة الحرجة و التركيز الحرج وكانت النتيجة 67.0 °C و 33.90% من وزن الفينول على التوالي. وقد استخدمت نظرية الازدواج (MCT) لحساب قيم كل من معامل اللزوجة $\eta_0 = 0.684$ cP، قطع زخم ديباي $q_\mathrm{D} = 0.786\ \mathrm{Å}^{-1}$ والثابتِ A في نظرية الازدواج وكان يساوي 0.050. وقد تم حساب قوى الربط بين جزيئات الماء و الفينول في المخلوط الثنائي و كانت النتيجة L = 11.17 Å ، هذه القيمة الكبيرة تشير الى أن القوى المتبادلة بين جزيئات الخليط الثنائي هي قوى تجاذب ضعيفة. تم قياس السعة الحرجة للحرارة النوعية عند ثبوت الضغط وعند التركيز الحرج وحول درجة الحرارة الحرجة $C_\mathrm{pc} = 259.16 \frac{\mathrm{J}}{\mathrm{kg.K}}$ باستخدام قانون المقياس العالمي للمخاليط الثنائية.