An-Najah National University

Faculty of Graduate Studies

Intermolecular Force Range and Noncritical Shear Viscosity of the Critical Binary Mixture of Benzene -Coconut Oil

By

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III
Dedication

To the candles that burned yesterday to light up my life today, my father and mother, as well as, to my dear husband, who shared my life and supporting me all the time.

This thesis also dedicated to my mother- in- law, who helped me to accomplish this work, also to whose have endured my absence from them for extended periods, my children Jana and Omar.

To my brothers and sisters, to my family with love

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V

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الاقرار

أنا الموقعة أدناه مقدمة الرسالة التي تحمل العنوان:

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Intermolecular Force Range and Noncritical Shear Viscosity of the Critical Binary Mixture of Benzene -Coconut Oil

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اقر بان ما اشتملت عليه هذه الرسالة إنما هو نتاج جهدي الخاص ، باستثناء م ا تمت الإشارة إليه حيثما ورد، وان هذه الرسالة ككل من أو جزء منها لم يقدم من قبل لنيل أية درجة أو بحث علمي أو بحثي لدى أية مؤسسة تعليمية أو بحثية أخرى.

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<u>Declaration</u>

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.

Student's Name: اسم الطالبة:

Signature: التوقيع:

Date: التاريخ:

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List of Abbreviations

A	The mode coupling theory constant
atm	Atmospheric pressure
c	Specific heat
cP	Centipoises
C_{PC}	Critical amplitude of the specific heat at constant pressure
cSt	Centistokes
gm	Gram
H	Heat
k_B	Current
I	Joule's constant
J	Kelven Degree
K	Boltzmann's constant
Kg	Kilogram
L	Intermolecular force range
Molal	Mole/Liter
MPa	Mega Pascal
N	Newton
Q	Volume flow rate
q_D	Debye momentum cutoff constant
R	Pipe radius
R_ζ	Universal quantity
R^2	Coefficient of determination
s	Seconds
t	Time
T_c	Critical temperature
$T^{`}_{c}$	Pressure derivation of the critical temperature along the critical line
ν	The kinematic viscosity
V	Volt
W	Work
α	Critical exponent
α_{Pc}	Critical amplitude of the thermal expansion at constant pressure
ɤ	Ratio between specific heat at constant pressure and specific heat at constant volume
Δp	Pressure difference
ΔT	Temperature difference
$\dfrac{\partial y}{\partial x}$	Rate of deformation or velocity gradient
ζ_o	Correlation length

η	Shear viscosity
η_o	Noncritical part of the measured shear viscosity
υ	Critical exponent
ρ	Mass density of the liquid
ρ_c	Mass density at the critical temperature
τ	The reduced temperature
x_c	Critical concentration
$x_\eta \upsilon$	Critical exponent
ς	Shear stress

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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 T_c 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 $\eta_o = 1.30\text{cP}$. The Debye momentum cutoff q_D is found to be $0.358\,\text{Å}^{-1}$, the constant A = 0.052 ± 0.003, the correlation length $\zeta_o = 1.02\text{Å}$, and the intermolecular force range L= 9.90Å by using the mode coupling approach. The forces are weak because of the small value of the viscosity.

Chapter One

Introduction

1.1 Liquids and Binary Mixtures

The states of matter in nature are solid, liquid, gas and plasma. The liquid is able to flow and take the shape of a container. Some liquids can be compressed, but others resist compression. Liquid does not disperse to fill every space of a container, and maintains a fairly constant density. A distinctive property of the liquid state is surface tension (Hansen and Mc Donald, 2006).

Most systems in nature and technology are mixtures of differently sized particles. Each binary mixture is a mixture that contains two constituents. In chemistry, a mixture is a material system made up of two or more different substances which mixed but are not combined chemically. A mixture refers to the physical combination of two or more substances on which the identities are retained and mixed in the form of solutions, suspensions, and colloids. Examples of binary mixtures are benzene - coconut oil, nitrobenzene - n-hexane, and methanol - cyclohexane (Atkins and Pauli, 2002; Abdelraziq *et al*, 1989).

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 (Abdelraziq, 2005; Yusur Kittany, 2014).

1.2 Previous Studies

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" (Symon and Keith, 1971). 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 (Lide, 2005).

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 factors such as temperature, shear rate, catalyst, pressure, and molecular weight concentration (Lide, 2005; Tajweed Niarat *et al,* 2014). There are two types of fluid viscosity known as dynamic (or absolute) and kinematic viscosity. These types were studied for different aqueous solutions; for example Kestin and his group tabulated values of dynamic and kinematic viscosity of potassium chloride and sodium chloride on temperature range 25-150 ºC, pressure range 0.1-35 MPa and concentration range 0-5 molal (Kestin *et al*, 1981). Tat and Gerpen studied the kinematic viscosity of biodiesel and it blends with diesel fuel from close to their melting point to 100°C, the results indicate that their viscosity of blend biodiesel is getting higher than biodiesel (Tat and Van Gerpen, 2002). Sparks and his group measured the dynamic viscosity and density of a fluid by using the micromachined resonating tube

of a fluid enabling the calculation of the kinematic viscosity (Sparks *et al*, 2009).

Barnett studied the relation between the dynamic viscosity η of liquid and their chemical nature (Barnett *et al*, 1896). De Guzman proposed three-constants form to represent liquid viscosity as a function of temperature (De Guzman, 1913). Vogel proposed a three-constant representation (Vogel, 1921). Goletz then applied the temperature dependence proposed by Vogel over the full range from the normal boiling temperature to the freezing point and generalized an Antoine type equation (Goletz and Tassiou, 1977). Poling presented power law form of liquid viscosity as a function of temperature and represented liquid viscosity in the polynomial form (Poling *et al*, 1987).

A form for absolute and kinematic viscosities and the constants of their equations are presented by Natarajan (Natarajan and Viswanath, 1989). Dutt made a study on 100 liquids and obtained the constants of Natarajan form to them (Dutt and Prasad, 2004).

Some researchers studied the viscosity of different materials such oils, and water as a function of temperature, intensity of light, pressure, molecular weight and density, as Bridgman who developed a method by which the relative viscosity of liquids may be determined over a wide range of pressures at various temperatures. The method has been applied to 43 liquids in the pressure range between atmospheric and 12,000 kg/cm at 30°C and 75°C (Bridgman, 1925; Tajweed Niarat *et al,* 2014; Tajweed Niarat *et al,* 2013; Tajweed Niarat, 2014; Ahmad Bahti, 2014). Another

study by Bakshi, gave an empirical relation between velocity of sound in a liquid and viscosity (Bakshi and Parthasarathy, 1953).

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 (Fixman, 1962; Abdelraziq, 1989; Diya Qasem, 2014). Katti and Chaudhri measured viscosities of binary mixtures of Benzyl Acetate - Dioxane, Aniline - m-cresol (Katti and Chaudhri, 1964). Viscosities and densities of 14 binary nonelectroyle systems were measured at 25 ℃ by Herice and Brewer (Herice and Brewer, 1967). 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 (Teja and Rice, 1981). Viscosities and densities of 68 aromatic hydrocarbons binary liquid systems were studied at 298.15 K by Chevaller and his group (Chevaller *et al*, 1990).

The existence of shear viscosity anomaly has been experimentally observed in some binary liquid systems near the critical temperature and concentration (Senger, 1972; Yamada and Kawasaki, 1975; Ohta and Kawasaki, 1976; Abdelraziq, 1990; Abdelraziq, 1992; Abdelraziq *et al,* 1999; Abdelraziq, 1996; Abdelraziq, 2000; Abdelraziq, 2001; Abdelraziq, 2002; Saja Omar, 2014). 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 mixture (D'Arrigo *et al*, 1977). Nieuwoudt and Sengers made a reevaluation for the critical

exponent of viscosity for binary mixtures near consolute point (Nieuwoudt and Sengers, 1989).

Abdelraziq measured the shear viscosity coefficient of nitrothane - 3-methelpentane binary mixture (Abdelraziq, 2002). Domanska and Krolikowska determined the densities and viscosity correlations for five different binary mixtures of the ionic liquids (Domanska and Krolikowska, 2012). The tracer diffusion coefficients of some binary mixtures like tritiated water and acetonitrile in water - acetonitrile have been determined at several temperatures (Easteal, 1980).

Binary mixtures have been studied from several aspects. Many thermodynamic properties of binary liquid mixtures were studied. Frank and Evans studied the entropy in binary mixtures (Frank and Evans, 1945). Excess volumes and excess heats of three binary mixtures were measured by Rao and Naidu (Rao and Naidu, 1976). McDonald made application to calculate the thermodynamic properties as excess enthalpy, volume and Gibbs free energy of 12–6 liquids of Lennard-Jones binary mixtures (McDonald, 2006).

The relationships between refractive index, molar volume and molar refraction, and the corresponding mixing properties of binary mixtures were clarified by Brocos and his group (Brocos *et al*, 2003).

Kumar and his group showed that binary liquid mixtures shape ideal systems in which to evaluation of some predictions of the modern theories of critical phenomena. Kumar, Krishnamurthy, and Gopal exhibited critical review of recent experimental studies of equilibrium critical phenomena in

binary liquid mixtures (Kumar *et al*, 1983). The critical behavior of binary liquid mixtures was examined by small angle neutron scattering for binary mixtures of protonated and perdeuterated 1, 4-polybutadiene and was shown to display an upper critical solution temperature by Bates and his group (Bates *et al*, 1985).

Ternary mixtures are a three-part mixture of some substance, the viscosity and density has been experimentally measured of ternary liquid mixtures. D'Arrigo and his group measured shear viscosity in the ternary liquid mixtures as water-benzene-ethanol and water-diethylether-ethanol near 25 ºC and 1 atm (D'Arrigo *et al*, 1977). The viscosity and density of other ternary mixture of di (2-ethylhexyl) phosphoric acid-benzene-toluene were measured at different temperatures by Swain and Chakravotty (Swain *et al*, 1999). Meng measured and predicted densities and viscosities of aqueous binary and ternary solutions at temperatures rang (20-60) °C (Meng, 2000). 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 (Swinney and Henry, 1973). Ohta and Kawasaki were studied the dynamic critical phenomena of classical liquids by using the mode coupling theories. They also investigated the vertex corrections and non-Vfarkoffian effects (Ohta and Kawasaki, 1976).

The sound absorption and dispersion were studied numerically near the critical point in a critical binary mixture depending on Kawasaki mode-coupling theory. They also showed that the mode-coupling approach breaks

down over a large range of temperatures and frequencies (Harada *et al*, 1980*)*.

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 (Gotze, 1998).

Kob and Anderson tested the mode coupling theory for super cooled binary mixture (Lennard-Jones) and they calculated Van Hove correlation function (Kob and Anderson, 2008).

1.3 Objectives of the Study

The main goals of this study are:

1. To measure the critical temperature and critical concentration of benzene - coconut oil binary mixture.

2. To determine the mentioned below constants of benzene - coconut oil binary mixture using the mode coupling theory. These constants are:

    1. The noncritical part of the measured shear viscosity η_o.

    2. The correlation length ζ_o.

    3. The intermolecular force rang L (Debye Parameter).

    4. The Debye momentum cutoff q_D constant.

    5. The constant A.

1.4 Motivation

The interest in studying the binary liquid mixtures of these days increased because of the presence of its distinctive characteristics from the rest of the liquids, benzene – coconut oil binary mixture is one of them.

Benzene – coconut oil binary mixture can be used as an alternative fuel, which made from vegetable oils such as coconut oil, soybean, cotton or Jatropha mixed with petroleum or conventional diesel.

This mixture has been studied using the dynamic scaling theory by Bhattacharya and Deo (Bhattacharya and Deo, 1983), and Abdelraziq (Abdelraziq, 2005), but has not been studied using the mode coupling theory.

1.5 Lay Out

In chapter 2, the theoretical background of the experimental work will be discussed; such as the theory of viscosity for pure and binary liquids, the mode coupling theory. In chapter 3, the methodology of the experiment: the characteristics of the components, experimental apparatus, procedure, and statistical analysis. The measured data will be given in chapter 4, the critical point will be determined and the viscosity data near the critical point will be analyzed using the mode coupling theory. A comparison between our results and literatures also is done. In chapter 5, the conclusion of this work and future works will be displayed.

Chapter Two

Theory

2.1 Viscosity

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 (Symon and Keith, 1971).

The shear viscosity of a fluid can be expressed in two distinct forms:

1. The dynamic viscosity:

    The dynamic viscosity is defined as the ratio of shear stress (force over cross section area) to the rate of deformation.

    $$\eta = \frac{\text{ţ}}{\frac{\partial y}{\partial x}} \tag{2.1}$$

    Where η is the dynamic viscosity in (Pa.s), ţ is shear stress ($\frac{N}{m^2}$), and $\frac{\partial y}{\partial x}$ is rate of deformation or velocity gradient (shear rate) ($\frac{1}{s}$) (Dutt *et al*, 2007).

    The dynamic viscosity unit is used to be centipoise (cP) which is 10^{-2} of Poise.

2. Kinematic viscosity:

    The kinematic viscosity is the dynamic viscosity η divided by the mass density of the fluid ρ.

    $$v = \frac{\eta}{\rho} \tag{2.2}$$

Where, ν is kinematic viscosity in centistokes (cSt), ρ is mass density in $\frac{g}{cm^3}$ (Dutt *et al*, 2007).

2.2 Pure Liquid Viscosity

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 (Dutt *et al*, 2007).

The relationship that explains the relation between the dynamic viscosity and the temperature is in the form of an Arrhenius-type equation (Clements *et al*, 2006).

$$\eta = \eta_o \, e^{\frac{E_\eta}{RT}} \qquad (2.3)$$

Where η is the measured dynamic shear viscosity, η_o is the dynamic shear viscosity at some reference temperature, E_η 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;

$$Ln(\eta) = \check{A} + \frac{B}{T} \qquad (2.4)$$

Where $\check{A}$ and B are constants determined empirically.

At temperatures above the normal boiling point, the $\ln(\eta)$ versus $\frac{1}{T}$ relationship becomes nonlinear and is described by a number of semi-empirical methods.

The relationship that explains the relation between the dynamic viscosity and the pressure is expressed in terms of an equation known as Poiseuille's law (Tuchinsky, 1976)

$$Q = \frac{\pi R^4 \Delta p}{8 \eta l} \qquad (2.5)$$

$$\eta = \frac{\pi R^4 \Delta p}{8 l Q}$$

Where Q is volume flow rate, Δp is pressure difference, R is pipe radius, l is pipe length, and η is dynamic viscosity

2.3 Dynamic Viscosity of Mixtures

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)(Dutt *et al*, 2007).

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

2.3.1 The Mode Coupling Theory

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 (Horst, 2012).

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

The mode coupling theories were studied by Kawasaki in 1970, by Mistura in 1971 and by Perl and Ferrell in 1972 (Kawasaki, 1970; Mistura, 1972; Perl and Ferrell, 1972). 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 form from a field-theoretic perspective.They reviewed the mode coupling theory successes and failures (Reichman and Charbonneau, 2005).

The critical anomaly of the shear viscosity coefficient by Kawasaki and Perl and Ferrell is given according to the law (Perl and Ferrell, 1972):

$$\frac{\eta - \eta_o}{\eta} = \frac{\Delta\eta}{\eta} = A \ Ln \ \zeta + A \ Ln \ q_D \qquad (2.6)$$

Where η_o 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 (D'Arrigo $et \ al$, 1977), 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

$$\eta = \eta_0 \, \tau^{\,-x_\eta v} \qquad\qquad (2.7)$$

Where τ is the reduced temperature, which is given by $\dfrac{T - T_c}{T_c}$, v is critical exponent $= 0.64$(D'Arrigo *et al*, 1977; Klein and Woermann, 1978), $x_\eta v$ 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 (Matthaeus *et al*, 1971). The correlation length is the average distance between two molecules at temperatures near the critical temperature.

The correlation length expression is given by

$$\zeta = \zeta_0 \, \tau^{-v} \qquad\qquad (2.8)$$

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 $\eta = \eta_0$ (Buchingham, 1937).

The intermolecular force range L (Debye parameter) can be calculated from light-scattering experiment by the relation (Klein. H. *et al,* 1978)

$$L = 2.45 \, \zeta_0 T_c^{\frac{\gamma - 1}{2}} \qquad\qquad (2.9)$$

Where γ is the ratio between specific heat at constant pressure and specific heat at constant volume, $\gamma = 1.25$ (Klein. H. *et al,* 1978).

2.4 Two – Scale – Factor Universality

The two – scale – factor universality has been used in modern theories to explain the critical phenomena of binary liquid mixtures by predict R_ζ(Hohenberg *et al*, 1976). This quantity is defined as

$$R_\zeta = \zeta_o[\alpha \frac{\rho_c C_{pc}}{k_B}]^{\frac{1}{d}} = \zeta_o[\alpha \frac{T_c \alpha_{pc}}{k_B T_c^`}]^{\frac{1}{d}} \qquad (2.10)$$

Where R_ζ is the universal quantity and it is equal 0.270 (Bervillier, 1976), 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(Abdelraziq, 2003).

The specific heat at constant pressure c_p is given by

$$c_p = c_{pc} \tau^{-\alpha} + c_{pb} \qquad (2.11)$$

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

Chapter three

Methodology

High-purified samples of benzene and coconut oil were used, in this work. The shear viscosities from 18 to 42 ℃ 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.

3.1 The Characteristics of the Binary Mixture Components

1. Benzene :

Benzene is an organic chemical compound with the molecular formula C_6H_6. Its molecule is composed of 6 carbon atoms joined in a ring, with 1 hydrogen atom attached to each carbon atom. Benzene molecules contain only carbon and hydrogen atoms, so it is classified as a hydrocarbon. Benzene is a colorless and highly flammable liquid (Lide, 2005). High-purified benzene (99%) was used.

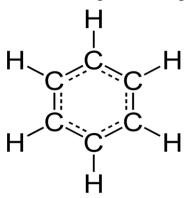The molecular structure of benzene is given in Fig.(3.1).

Fig.(3.1): The molecular structure of benzene.

2. Coconut oil:

The scientific name for coconut is Cocos nucifera. Coconut oil is an edible oil extracted from the kernel or meat of matured coconuts harvested from the coconut palm. It has various applications in food, medicine, and industry. It is slow to oxidize, because of its high saturated fat content (Bruce, 2005). High-purified coconut oil was used in this work.

The characters of benzene and coconut oil are shown in Table (3.1):

Table (3.1): Physical and chemical properties of benzene and coconut oil.

Property	Benzene (Winek *et al*, 1971)	Coconut oil (Krishna *et al,* 2010)
Color	Colorless	Colorless(T>30ºC), white (T<30ºC).
Molecular weight	78.11 gm/mole	-
Melting point	5.5ºC (41.9ºF)	24ºC
Density (20ºC)	0.879 gm/cm^3	0.921 gm/cm^3
Solubility in water (25ºC)	1.8 gm/Liter	Insoluble
Viscosity (25 ºC)	0.604 cP	55.4 cP

3.2 Experimental Apparatus

1- The instrument that was used to measure the viscosity of the critical mixture of benzene and coconut oil is Brookfield Viscometer Model DV-I+:

The viscometer has a set of seven spindles RV SPINDLE SET and UL-ADAPTER with accuracy ±1%. It has two rotational speeds set of the spindles. The first set: 0.0, 0.5, 1, 2, 2.5, 4, 5, 10, 20, 50 and 100 rpm. The second set: 0.0, 0.3, 0.6, 1.5, 3, 6, 12, 30 and 60 rpm. The spindles measure viscosity range from 100 up to 1.33×10^7 cP, The UL-spindles measure

viscosity range from 1 up to 2000 cP (Manual No.M/92-021-K1098) Fig.(3.2).

Fig.(3.2): The Brookfield Viscometer Model DV-I+.

2- Digital Prima Long Thermometer is used to measure the temperature; its accuracy is ±1% with measures temperature range is from −20°C to +100°C Fig.(3.3).

Fig.(3.3): The Digital Prima Long Thermometer.

3- Fried Electric model WB-23 was used to measured and control the temperature of the samples to a specific temperature Fig.(3.4).

Fig.(3.4): The Fried Electric model WB-23.

4- Pycnometer (Density bottle) is used to measure the density Fig.(3.5).

Fig.(3.5): The Pycnometer.

5- The analytical balance HR-200 with accuracy of ± 0.00005% was used to measure the mass Fig.(3.6).

Fig.(3.6): The analytical balance HR-200.

6- Calorimeter set is used to measure the heat capacity Fig.(3.7).

Fig.(3.7): Calorimeter set.

The set has calorimeter, digital voltmeter (accuracy in the DC volts range is ±1 %), digital ammeter (accuracy of the ammeter is ±1 %), and power supply.

3.3 Procedure

Samples were prepared from 0% benzene by weight to 100% benzene by weight using the analytical balance HR-200.

1- Binary liquid mixture (coconut oil – benzene) samples preparation.

Concentrations were prepared from 0% benzene by weight to 100% benzene by weight, the first is pure coconut oil (0% benzene), the last is pure benzene (100% benzene) and the samples were prepared by following procedure:

The concentration of benzene by weight in the binary mixture =

$$x_{Benzene} = \frac{\rho_{Benzene}*V_{Benzene}}{\rho_{Benzene}*V_{Benzene}+ \rho_{Coconut\ oil}*V_{Coconut\ oil}} \times 100\% \quad (3.1)$$

Where $\rho_{Benzene}$ is the density of benzene, $V_{Benzene}$ is the volume of benzene, $\rho_{Coconut\ oil}$ is the density of coconut oil, and $V_{Coconut\ oil}$ is the volume of coconut oil

A one component volume was fixed to find the other.

2- Viscosity measurements.

The viscosity of liquids was measured by different instruments, capillary viscometers, orifice viscometers, high temperature high shear rate viscometers, rotational viscometers, falling ball viscometers, vibrational viscometers, and ultrasonic viscometers (Dutt *et al*, 2007).

The Brookfield Rotational Viscometer Model DV-I+ was used to measure the viscosity of the critical mixture of benzene-coconut oil samples. The viscosity of each sample was measured at temperature range from 18ºC to 42ºC.

3- Heat capacity measurement.

The binary mixture was weighed before putting in the calorimeter. The calorimeter was directly connected with power supply, digital ammeter, digital voltmeter set. The current values were changed using the variable resister. The reading of the digital ammeter and the digital voltmeter were taken at temperature, the time needed to reach to the specified temperature was read by the timer.

The following equation is used to find the work done on the critical binary mixture to calculate Joule`s constant.

$$\text{Work} = \text{current} \times \text{volt} \times \text{time}$$

$$W = I \times V \times t \qquad (3.2)$$

Eq.(3.3) is used to calculate Joule`s constant

$$J = \frac{W}{H} \qquad (3.3)$$

Where W is the work, I is the current, V is the volt, t is the time, J is joule`s constant, and H is the quantity of heat.

Joule`s constant of benzene-coconut oil mixture (70% benzene by weight) was calculated by using two – scale – factor universality and Eq. (3.4), then H was estimated.

The specific heat c_b of the binary mixture at different ΔT was calculated by using:

$$H = (m_o\, c_o + m_s\, c_s + m_h c_h + m_b c_b)\, \Delta T \qquad (3.4)$$

Where: m_o = mass of calorimeter cup, m_s = mass of stirring rod, m_h= mass of heater, m_b= mass of the binary mixture, c_o = specific heat of calorimeter cup, c_s specific heat of stirring rod, c_h specific heat of heater, c_b specific heat of the binary mixture at critical concentration, and ΔT temperature difference.

3.4 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.

Chapter Four

Results and Discussion

4.1 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 (18-42ºC) as given in Table (4.1).

Table (4.1): The dynamic viscosity values of benzene - coconut oil samples of different concentrations (0%-100%) by weight of benzene at temperature range (18-42ºC).

T (ºC)	η(cP) 0%*	η(cP) 10%*	η(cP) 20%*	η(cP) 30%*	η(cP) 40%*	η(cP) 50%*	η(cP) 60%*	η(cP) 70%*	η(cP) 80%*	η(cP) 90%*	η(cP) 100%*
18	61.5	26.8	13.4	9.6	4.61	3.58	2.62	1.41	0.96	0.96	0.77
19	55.1	25.5	12.9	9.44	4.48	3.58	2.65	1.41	0.96	0.96	0.77
20	51.7	24.3	12.5	9.01	4.35	3.52	2.59	1.34	0.96	0.96	0.77
21	48.6	23.1	12.1	8.75	4.22	3.39	2.56	1.3	0.96	0.96	0.77
22	45.2	22	11.6	8.38	4.1	3.33	2.50	1.3	0.96	0.96	0.77
23	42.7	20.9	11.2	8.11	3.97	3.2	2.47	1.28	0.93	0.96	0.77
24	40	20	10.8	7.95	3.84	3.14	2.43	1.26	0.93	0.94	0.77
25	37.9	18.9	10.4	7.57	3.71	3.01	2.37	1.25	0.93	0.93	0.77
26	35.6	18	10	7.36	3.65	2.88	2.37	1.31	0.93	0.9	0.77
27	33.6	17	9.73	7.04	3.58	2.82	2.37	1.22	0.93	0.9	0.77
28	31.6	16.3	9.34	6.83	3.46	2.75	2.30	1.18	0.93	0.87	0.77
29	29.4	15.6	8.96	6.56	3.39	2.69	2.24	1.16	0.93	0.87	0.77
30	27.9	14.7	8.77	6.29	3.26	2.62	2.24	1.15	0.93	0.87	0.77
31	26.9	14.1	8.45	6.03	3.2	2.56	2.24	1.15	0.93	0.87	0.77
32	24.9	13.4	8.13	5.76	3.14	2.5	2.24	1.12	0.93	0.83	0.735
33	23.3	12.7	7.87	5.55	3.07	2.43	2.21	1.09	0.93	0.83	0.735
34	21.9	12.2	7.62	5.33	2.94	2.37	2.18	1.09	0.9	0.83	0.735
35	20.5	11.7	7.3	5.23	2.88	2.3	2.14	1.09	0.9	0.83	0.735
36	19.1	11.1	7.04	5.07	2.82	2.27	2.12	1.05	0.9	0.83	0.7
37	17.9	10.6	6.78	5.01	2.75	2.24	2.11	1.04	0.9	0.8	0.7
38	16.7	10	6.59	4.69	2.69	2.18	2.07	1.02	0.9	0.8	0.7
39	15.9	9.54	6.49	4.59	2.62	2.15	2.05	0.99	0.87	0.8	0.7
40	15.3	9.09	6.27	4.48	2.5	2.08	1.92	0.96	0.87	0.8	0.7
41	14.6	8.7	6.08	4.37	2.43	2.05	1.86	0.93	0.83	0.8	0.67
42	14.4	8.32	5.95	4.35	2.43	1.98	1.79	0.9	0.83	0.8	0.63

* Concentration by weight of benzene

The dynamic viscosity of benzene - coconut oil of different concentrations (0%-80%) benzene as a function of temperature are plotted in Figs.(4.1) – (4.3).

Fig.(4.1): The dynamic viscosity of benzene – coconut oil as a function of temperature at concentrations (00%-40%) by weight of benzene.
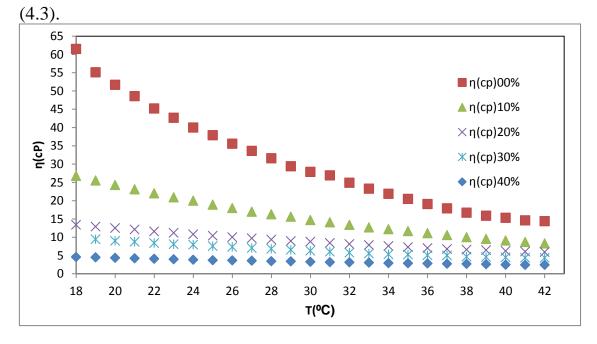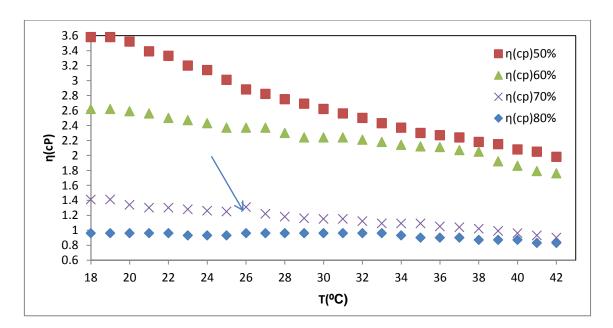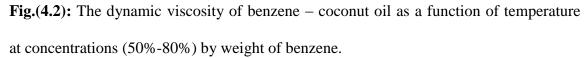

Fig.(4.2): The dynamic viscosity of benzene – coconut oil as a function of temperature at concentrations (50%-80%) by weight of benzene.

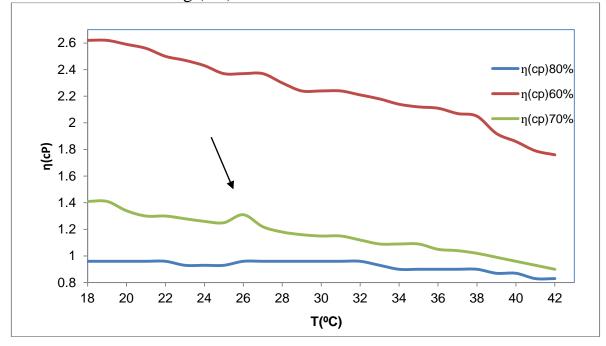The anomaly can be noticed by plotting 60%, 70%, and 80% by weight of benzene as shown in Fig.(4.3).

Fig (4.3): Dynamic viscosity at different temperatures for 60%, 70%, and 80% by weight of benzene.

Figs. (4.2) and (4.3) show a cusp at $x_c = 0.700$ by weight of benzene and $T_c = 26.00°C$. 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 T_c is 25.40 °C and the critical concentration x_c is 0.6037 by weight of benzene (Battacharya and Deo; 1983). This will affect the value of the noncritical part of the shear viscosity η_o.

4.2 The Noncritical Part Shear Viscosity η_o

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 $\eta = \eta_o\, \tau^{-x_\eta \upsilon}$, where x_η and υ are critical exponents. These dynamic shear viscosity are displayed in Table (4.2).

Table (4.2): The dynamic shear viscosity values above the critical temperature $T_c = 26.00^oC$.

$\eta(cP)$	$T(^oC)$	$T\text{-}T_c$	$\tau = \dfrac{T-T_c}{T_c}$	$\ln(\eta)$	$\ln(\tau)$
1.54	26.2	0.2	0.0077	0.432	-4.867
1.47	26.4	0.4	0.0154	0.385	-4.174
1.50	26.6	0.6	0.0231	0.406	-3.769
1.50	26.8	0.8	0.0308	0.406	-3.481
1.54	27.0	1.0	0.0385	0.432	-3.258
1.47	27.2	1.2	0.0462	0.385	-3.076
1.41	27.4	1.4	0.0538	0.344	-2.922
1.44	27.6	1.6	0.0615	0.365	-2.788
1.41	27.8	1.8	0.0692	0.344	-2.670
1.41	28.0	2.0	0.0769	0.344	-2.565
1.44	28.2	2.2	0.0846	0.365	-2.470
1.41	28.4	2.4	0.0923	0.344	-2.383
1.41	28.6	2.6	0.1000	0.344	-2.303
1.41	28.8	2.8	0.1077	0.344	-2.228
1.41	29.0	3.0	0.1154	0.344	-2.159
1.44	29.2	3.2	0.1231	0.365	-2.095

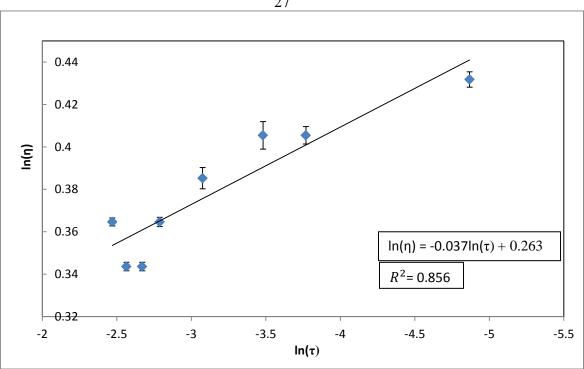Ln(η) is plotted against ln(τ) to find the value of the critical exponent $x_\eta\, \upsilon$ as shown in Fig (4.4).

Fig. (4.4): Ln(η) vs. Ln(τ), using power law $\eta = \eta_o t^{-x_\eta v}$.

Fig (4.4) shows that the values of the shear viscosity fit the power law

$$\eta = \eta_o\, \tau^{-x_\eta v}.$$

$$\ln(\eta) = \ln(\eta_o) - x_\eta\, v\, \ln(\tau)$$

The constant $x_\eta v$ is the slope which equals to 0.037 ± 0.003 it is in good agreement with the critical exponent $x_\eta v$ of D'Arrigo and Klein and Woermann which equal 0.037 ± 0.003 (D'Arrigo *et al*, 1977; Klein and Woermann, 1978).

The intercept equals $\ln(\eta_o) = 0.263$. The noncritical part of the measured shear viscosity $\eta_o = 1.30$cP; η_o was found by Abdelraziq to be 2.76cP (Abdelraziq, 2005). The noncritical part of the measured shear viscosity η_o is small compared to Abdelraziq`s result because of the different in values of critical temperature T_c and critical concentration x_c .

$\eta = \eta_o \, \tau^{-x_\eta \, \nu}$ is used to calculate the noncritical part of the measured shear viscosity η_o by plotting η vs. $\tau^{-0.037}$.

The measured dynamic shear viscosity η at the critical concentration is plotted versus $\tau^{-0.037}$ as shown in Fig. (4.5).
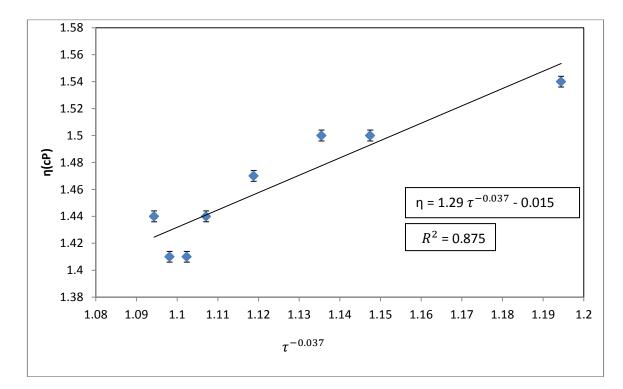

Fig. (4.5): The shear viscosity η versus $\tau^{-0.037}$.

The noncritical part of the measured shear viscosity η_o from the slope is found to be 1.29 cP, and it is very close to the value which is found by using Fig.(4.4).

4.3 Calculation of the Correlation Length ζ_o

Two – scale – factor universality is used to find the correlation length ζ_o of the benzene-coconut oil binary liquid mixture Eq. (2.10).

$$R_\zeta = \zeta_o[\alpha \frac{\rho_c C_{PC}}{k_B}]^{\frac{1}{d}} = \zeta_o[\alpha \frac{T_c \alpha_{PC}}{k_B \, T`_c}]^{\frac{1}{d}} = 0.270$$

Where critical exponent α = 0.11, dimension d = 3, mass density at the critical temperature $\rho_c = 0.878 \frac{g}{cm^3}$ which found in this work, where Boltzmann's constant is given by $k_B = 1.38 \times 10^{-23} \frac{J}{k}$.

The correlation length ζ_o for benzene-coconut oil binary liquid mixture is calculated and it is equal 1.98Å. The correlation length ζ_o for benzene-coconut oil binary liquid mixture was calculated at 25.4°C by Abdelraziq and it is equal 1.02Å (Abdelraziq, 2005).

The value of correlation length ζ_o 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°C, 0.7000 by weight of benzene) respectively. While in Abdelraziq`s work were (25.40°C, 0.6037 by weight of benzene) respectively.

The critical amplitude of the specific heat at constant pressure of the benzene - coconut oil mixture c_{pc} is calculated to be 84.30 $(\frac{J}{Kg.K})$ by using Eq. (2.10), and ζ_o which was found by Abdelraziq.

The specific heat at constant pressure of the benzene - coconut oil mixture c_{pc} is then used in Eq. (3.4) to find the quantity of heat H.

The value of Joule`s constant of the benzene - coconut oil binary liquid mixture equals 4.37 $\frac{Calori}{Joule}$ by using Eq. (3.3).

The work W, the quantity of heat H, the ammeter readings and the voltmeter reading of Joule`s constant experiment are given in Table (4.3).

The Joule`s constant J of the benzene - coconut oil binary liquid mixture which equal 4.37 $\frac{Calori}{Joule}$ is larger than Joule`s constant of water because of

most of the quantity of heat H is contributed from binary mixture which
contains 70% by weight of benzene.

Table (4.3): Joule`s constant experiment data.

T(ºC)	ΔT(ºC)	I(Ampere)	V(Volt)	t(Sec)	W(Joule)	$H = \frac{W}{J}$(Calori)
26.0	2.7	1.11	4.66	112	579.33	45.48
26.4	3.1	1.11	4.66	122	631.06	49.54
26.8	3.5	1.10	4.64	134	683.94	53.69
27.0	3.7	1.10	4.64	140	714.56	56.10
27.6	4.3	1.10	4.62	157	797.87	62.64
28.6	5.3	1.10	4.59	193	974.46	76.50
29.0	5.7	1.09	4.57	209	1041.09	81.73
29.2	5.9	1.09	4.57	216	1075.96	84.47

The specific heat at constant pressure (c_p) of the benzene - coconut oil
binary mixture is calculated from the value of the quantity of heat H for
temperatures from 26.2 – 29.2ºC. Eq. (2.11) is used to find the specific heat
at constant pressure of the benzene - coconut oil mixture c_{Pc} by plotting c_P
versus $\tau^{-0.11}$ which is shown in Fig.(4.6).

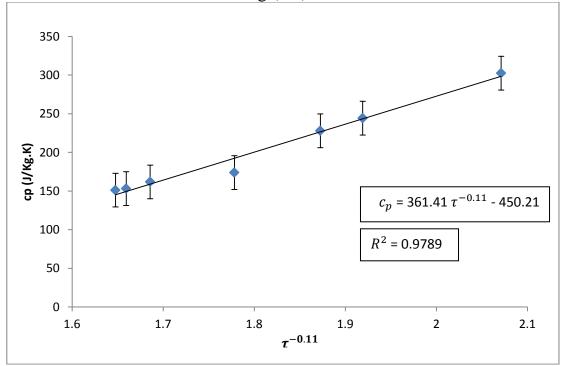$c_p = 361.41\ \tau^{-0.11} - 450.21$

$R^2 = 0.9789$

Fig. (4.6): The specific heat at constant pressure c_P versus $\tau^{-0.11}$.

Using Eq. (2.11), the critical amplitude of specific heat at constant pressure of the benzene-coconut oil mixture c_{pc} can be calculated.

The slope equals $c_{pc} = 361.41 \frac{J}{Kg.K}$, and the intercept is the critical background of specific heat at constant pressure of the benzene - coconut oil mixture $c_{pb} = 450.21 \frac{J}{Kg.K}$.

The critical amplitude of specific heat at constant pressure c_{pc} is used to calculate ζ_0.

4.4 The Intermolecular Force Range L (Debye Parameter)

The intermolecular force range L (Debye parameter) is calculated by using Eq. (2.9), Where ζ_0 equals to 1.98Å, γ is the ratio between specific heat at constant pressure and specific heat at constant volume, $\gamma = 1.25$ (Klein. H *et al*, 1978), and the critical temperature T_c is 26.00ºC.

The intermolecular force range L (Debye parameter) equals 9.90Å. 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 = 9.90Å gives an indication that the forces between the components of the benzene – coconut oil binary mixture is weak attractive force.

4.5 Calculation of the Constant A

The constant A of the mode coupling theory has a theoretical value 0.054 (D'Arrigo *et al*, 1977). The value of A can be determined by using Eq. (2.5), Where η_0 is the noncritical part shear viscosity which is calculated in section (4.2) to be 1.30 cP, $\frac{\Delta\eta}{\eta}$ is the relative anomalous dynamic shear

viscosity, ζ is the average correlation length of the concentration fluctuations which is given by Eq. (2.8). The relative anomalous shear viscosity $\frac{\Delta\eta}{\eta}$ versus the logarithm of the average correlation length $\ln\zeta$ is plotted in Fig.(4.7) at the critical concentration and above the critical temperature.
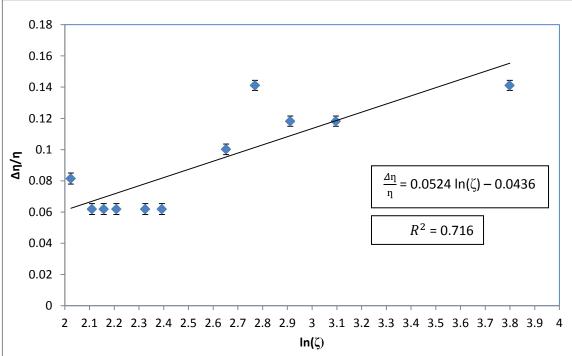

Fig.(4.7): $\frac{\Delta\eta}{\eta}$ versus the logarithm of the average correlation length $\ln \zeta$.

Fig.(4.7) shows that the relation between $\frac{\Delta\eta}{\eta}$ and $\ln\zeta$ is a linear relation with the slope equals to A value, which is 0.052 ± 0.003.

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 (Swinney and Henry, 1973; D'Arrigo *et al*, 1977; Abdelraziq, 2002; and Abdelraziq *et al*, 1996).

4.6 Determination of the Debye Momentum Cutoff q_D

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

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.

4.7 Results Summary

The results of the parameters which are found in this work and others work are summarized and given in Table (4.4).

Table (4.4): Summary of the measured and calculated parameters in this work and others work.

Parameter	This work	Previous studies
T_c	26.00°C	25.40°C[a]
x_c	0.700	0.6037[a]
η_o	1.30 cP	2.76 cP[b]
ζ_o	1.98 Å	1.02 Å[b]
L (Debye Parameter)	9.90 Å	-
q_D	$0.358 \, Å^{-1}$	-
c_{pc}	$361.41 \, \frac{J}{Kg.K}$	-
c_{pb}	$450.21 \frac{J}{Kg.K}$	-
J	$4.37 \, \frac{Calori}{Joule}$	-
A	0.052 ± 0.003	0.054[c] 0.056[d] 0.058[e] 0.054[f] 0.0538[g]

Where, a: (Battacharya and Deo; 1983), b: (Abdelraziq, 2005), c: (Swinny and Henry, 1973), d: (D'Arrigo *et al* , 1977), e: (Abdelraziq *et al,* 1996), f: (Abdelraziq, 2002), g: (Yusur Kittany, 2014).

Chapter Five

Conclusion 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_o has been determined to be 1.98 Å, the intermolecular force range L (Debye parameter) has been obtained to be 9.90 Å. 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 $0.358 \, Å^{-1}$.

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 Å; 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_o of the binary mixture is also smaller than the dynamic viscosity η_o of CCl_4 - coconut oil binary mixture.

Yusur calculated the intermolecular force range L for the binary mixture CCl_4 and coconut oil to be 5.5 Å (Yusur Kittany, 2014). Abdelraziq found the intermolecular force range L for the binary mixture of Nitroethane and 3-Methylpentane and it was 10.65 Å (Abdelraziq, 2002).

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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كلية الدراسات العليا

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إعداد

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إشراف

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الملخص

تم قياس معامل اللزوجة للخليط الثنائي ل البنزين و زيت جوز الهند باستخدام جهاز قياس اللزوجة الرقمي. وتم قياس اللزوجة لجميع التراكيز فوق درجة الحرارة الحرجة. وتم أخذ القراءات للزوجة للخليط عند التركيز الحرج و درجة الحرارة الحرجة (X_c, T_c)، وتم تحليل النتائج فوق درجة الحرارة الحرجة T_c باستخدام نظرية الإزدواج. وقد ظهر الشذوذ في اللزوجة الحرجة كإقتران مع درجة الحرارة والتركيز. و قد تم قياس قيمة الجزء غير الحرج من اللزوجة الحرجة بالقرب من درجة الحرارة الحرجة لتكون $\eta_o = 1.30cP$ وتم حساب قطع الزخم ديباي q_D وكان يساوي $0.358\ Å^{-1}$، و الثابت $A = 0.052 \pm 0.003$ ، و طول الإرتباط $\zeta_o = 1.02Å$ ، وقوة ربط الجزيئات (عامل ديباي) $L = 9.90Å$ باستخدام نظرية الإزدواج، ويظهر أن قوى الربط ضعيفة لأن قيمة اللزوجة صغيرة.