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**Critical Behavior of the Ultrasonic Attenuation
for the Binary Mixture of Water and Phenol**

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the Degree of Master of Physics, Faculty of Graduate Studies, An-
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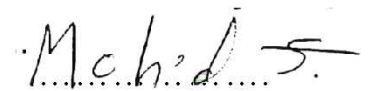
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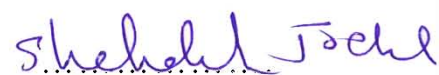
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III

Dedication

This thesis is dedicated to my dear husband, for his continued and unfailing support which makes the completion of this thesis possible.

Acknowledgment

I would like to express my thanks to several people for their contribution to the development and completion of this thesis.

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Finally my warmest thanks to my family members for their support

With respect and love.

الإقرار

أنا الموقعة أدناه مقدمة الرسالة التي تحمل العنوان:

Critical Behavior of the Ultrasonic Attenuation for the Binary Mixture of Water and Phenol

أقر بأن ما اشتملت عليه الرسالة إنما هو نتاج جهد شخصي، باستثناء ما تمت الإشارة إليه أينما ورد، وأن هذه الرسالة ككل أو كجزء منها لم يقدم من قبل لنيل أي درجة أو بحث علمي أو بحثي لدى أية مؤسسة تعليمية أو بحثية أخرى.

Declaration

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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14 / 1 / 2015

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List of Abbreviation

f	Frequency
b	Contribution of the frequency independent background absorption
S	The slope of absorption as a function of $f^{-1.06}$
C_p	Specific heat at constant pressure
C_{pc}	Specific heat at constant pressure at the critical point
C_{pb}	Background specific heat at constant pressure
g	Adiabatic coupling constant
v_c	Adiabatic sound velocity at critical temperature
t	Reduced temperature
α	Critical exponent = 0.11 ritical exponent = 0.11
α_p	Isobaric thermal expansion coefficient
α_{pc}	Critical term of isobaric thermal expansion coefficient
α_{pb}	Background term of isobaric thermal expansion coefficient
$C_p(t_f)$	Specific heat at constant pressure at a characteristic reduced temperature t_f
t_f	The reduced temperature where the attenuation has dropped to half value when it is at critical temperature.
ρ_c	Critical mass density of the binary liquid mixture
$\alpha_c(x_c, T_c)$	The absorption coefficient at critical temperature and critical concentration
$\alpha(x_c, T)$	The absorption coefficient at critical concentration and any temperature
ω^*	Experimental reduced frequency
ω^{*}	Theoretical reduced frequency
$\omega_D = \omega_0 t^{2\gamma}$	Characteristic temperature dependent relaxation rate
F(ω^{*})	The theoretical scaling function
β_T	The isothermal compressibility
β_{Tc}	The critical isothermal compressibility
β_{Tb}	The background isothermal compressibility
β_s	The adiabatic compressibility
β_{sc}	The critical adiabatic compressibility
β_{sb}	The background adiabatic compressibility
C_v	The specific heat at constant volume

X

C_{vc}	The critical specific heat at constant volume
C_{vb}	The background specific heat at constant volume
k_B	Boltzmann's constant
ξ	Correlation length
η	Dynamic shear viscosity
$z\gamma$	A critical exponent = 1.9
x_η	A critical exponent = 0.06
cP	CentiPoise
$^{\circ}\text{C}$	Celsius
K	Kelvin
x_c	Critical concentration
T_c	Critical temperature
rpm	Revolution per minute

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Abstract

The dynamic shear viscosity of the binary liquid mixture water – phenol was measured for different temperatures and concentrations using the glass capillary and digital viscometers. Dynamic shear viscosity anomaly was detected near the critical temperature $T_c = 67\text{ }^\circ\text{C}$ and the critical concentration $x_c = 33.9\%$ by weight of phenol. The specific heat at constant pressure was calculated using the two scale factor universality and found to be $241.9\text{ }\frac{\text{J}}{\text{kg K}}$. The critical and the background isobaric thermal expansion coefficients were determined and found to be $2.729 \times 10^{-3}\text{ }^\circ\text{C}^{-1}$ and $22.59 \times 10^{-3}\text{ }^\circ\text{C}^{-1}$ respectively. Ultrasonic attenuation data at 9, 15, 30 and 35 MHz were analyzed using the dynamic scaling theory of Ferrell and Bhattacharjee. The values of $\frac{\alpha_c}{f^2}$ versus $f^{-1.06}$ yields a straight line as predicted by the theory. The experimental values of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ for water – phenol were compared to the scaling function $F(\omega^*)$ and showed good agreement with the theory.

Chapter One

Introduction

The physical properties such as density and viscosity at several temperatures both for pure chemicals and their binary liquid mixtures over the whole composition range are useful for understanding of the thermodynamic and transport properties associated with heat and fluid flow. Binary liquid mixtures due to their unusual behavior have attracted considerable attention due to their importance from both theoretical and practical point of view (Ezekiel *et al*, 2012).

In the recent years, ultrasonic technique has become more powerful tool in providing information regarding the behavior of liquids and solids owing to its ability of characterizing physiochemical behavior of the medium. This type of study has increased in recent years due to industrial applications. Many researchers have undertaken these studies qualitatively through ultrasonic velocity, adiabatic compressibility and viscosity measurements (Bhandakkar *et al*, 2014).

1.1 Binary Liquid Mixture:

A binary liquid mixture is a combination of two pure liquids which can be classified as the following (Verma *et al*, 2005; Kenneth, 2003):

1. Completely miscible: when the two liquids are of the same nature, like water and alcohol.
2. Completely immiscible: when one liquid is polar while the other is not, such as water and mercury.

3. Partially miscible: if the intermolecular attractions of the two liquids are different. For example, ether and water.

In contrast to the pure liquid, in which the critical point occurs at a unique point as shown in Fig.(1.1a), the binary liquid mixtures possess a critical line which defines a locus of critical points for the mixture named the coexistence curve. At each point along this curve, the mixture exhibits a critical point with its own composition, temperature, and pressure. Fig.(1.1b) represents the common types of the coexistence curves for a binary mixture in the T - x phase diagrams at constant pressure (Iwanowski, 2007).

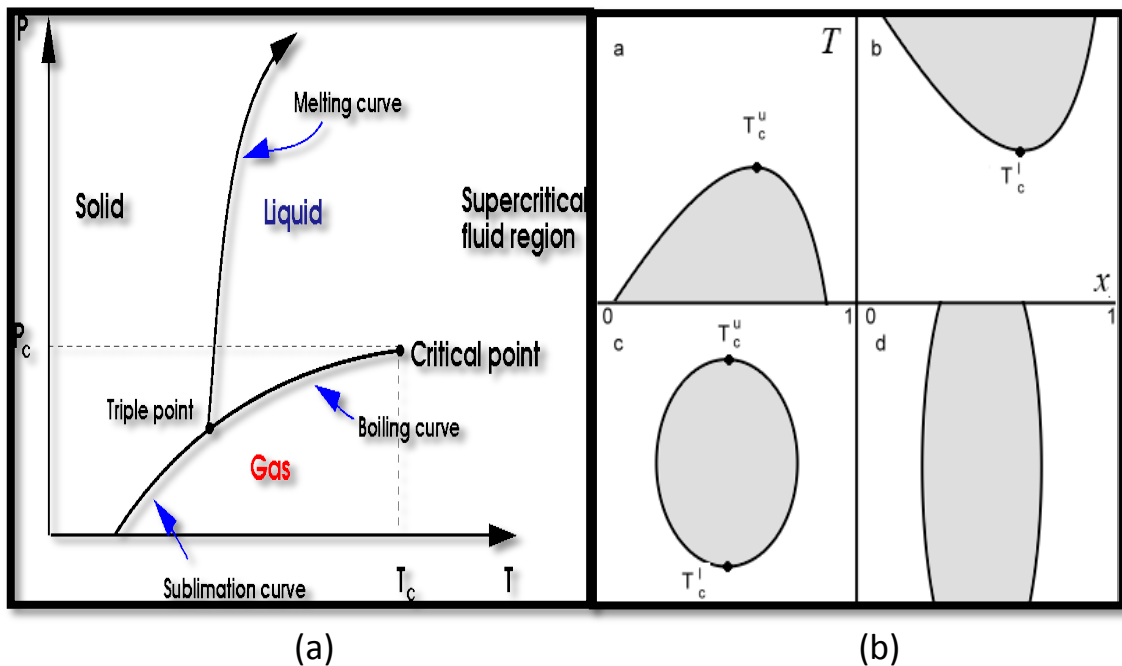


Fig.(1.1): Phase diagrams of (a) A pure liquid and (b) Binary liquid mixture at constant pressure (Iwanowski, 2007).

During the last decades, several experimental techniques have been developed in order to investigate the physical properties of pure and multi-

component fluids. For example, the dynamic light scattering technique which also known as Photon Correlation Spectroscopy (PCS) and the X-ray Photon Correlation Spectroscopy (XPCS) have been utilized to determine the size and shape of particles and to study critical fluctuations in multi-component fluids (Nurushev, 2000; Iwanowski, 2007).

Another powerful technique is the ultrasonic technique which has gained much importance in the last two decades in assessing the nature of molecular interaction and investigating the physiochemical behavior of liquid and liquid mixtures. Ultrasonic and other related thermo acoustic parameters provide useful information concerning the structure of molecules, molecular order, molecular packing, inter and intra molecular interaction (Alamelumangai and Santhi, 2014).

Three main theoretical models have been proposed to predict the fluids behavior and the sound propagation in the vicinity of the critical point:

- The dynamic renormalization-group theory: first formulated by Halperin and Hohenberg (Halperin *et al*, 1972; Hohenberg and Halperin, 1977). It allows to do predictions for the critical behavior of dynamic properties using dynamic scaling and universalities (Siggia *et al*, 1976).
- The mode coupling theory: based upon Fixmans' idea; that the critical anomalies can be comprehend as being due to nonlinear coupling between hydrodynamic modes (Fixman, 1962). It has been reformulated and developed by many authors especially Kawasaki

(Kawasaki, 1970; Ohta and Kawasaki, 1976; Shiwa and Kawasaki, 1981).

- The dynamic scaling theory: a general theory of the critical ultrasonic attenuation, established by Ferrell and Battacharjee based on an extension of the concept of the frequency-dependent specific heat; which was firstly introduced by Herzfeld and Rice in 1928 (Herzfeld and Rice, 1928; Ferrell *et al*, 1967; Ferrell and Battacharjee, 1981).

1.2. Literature Review

Continuous efforts have been made both theoretically and experimentally to study thermodynamic anomalies and sound propagation near the critical point:

The first quantitative explanation of the anomalously sound behavior (sound absorption rises to a maximum) near fluid critical point was attributed to Fixman (Fixman, 1960; 1962). His ideas were followed and modified by many scientists then used in analyzing experimental data (Kawasaki; 1969, Vincent *et al*, 1971; Mistura, 1972; Arrigo *et al*, 1977).

Dynamic scaling assumptions were suggested to predict the behavior and shape of time dependent correlation functions near the critical point of second order phase transitions (Halperin and Hohenberg, 1969).

Ultrasonic absorption in binary liquid mixtures of triethylamine with benzene, chloroform, cyclohexane and toluene at frequency 7.56 MHz has been studied using a pulse technique. Experimental values have good

agreement with theoretical evaluated values on the basis of Bauer-Sette theory (Rao and Reddy, 1977).

Ultrasonic attenuation in triethylamine and water binary mixture has been measured over the frequencies range 15 – 95 MHz and temperatures 12.5–18.0 °C. The experimental data have been analyzed on the basis of Kawasaki's mode coupling theory by Harada. As a result, he found the total sound absorption as a sum of three terms, the critical sound absorption on the basis of Kawasaki's mode coupling theory, the background relaxation part, and the classical absorption part which is due to shear viscosity (Harada, 1977).

The specific heat capacity at constant pressure C_p for the binary system triethylamine and water had been measured as a function of temperature near the critical solution temperature at the critical concentration by Thoen and his team. A very large increase in the specific heat near the critical temperature has been observed (Thoen *et al*, 1978).

Ferrell and Bhattacharjee have presented a theory of critical ultrasonic attenuation in binary liquids based on the frequency dependent specific heat. Good agreement with experiment was found, for the frequency dependence of the consolute point attenuation and for temperature dependence away from this point (Ferrell and Bhattacharjee, 1981).

Velocity and attenuation of the ultrasonic waves have been measured in complex binary mixtures of benzene and multi-component coconut oil near the critical temperature by Bhattacharya and Deo (Bhattacharya and Deo, 1983).The experimental results were analyzed in terms of theories

developed by Kawasaki and Mistura, a small velocity dispersion was observed (Bhattacharya and Deo, 1983; Kawasaki, 1970; Mistura, 1972).

The acoustic velocity and attenuation have been measured in a critical mixture of 3-methylpentane - nitroethane in the frequency range 1-17

MHz and temperature range $0.004 \leq T-T_c \leq 31.5$ K by Garland and Sanchez. The acoustic velocity varied linearly with temperature, and no critical dispersion was observed (Garland and Sanchez, 1983).

The ultrasonic velocity and attenuation have been measured for the critical binary mixture cyclohexane-nitroethane over the frequency range 3-27 MHz and the temperature range $0.01 \leq T-T_c \leq 15$ K by Sanchez and Garland. The sound velocity varied linearly with temperature and showed no critical dispersion in this frequency range. The critical attenuation was analyzed in terms of the dynamic scaling theory. The agreement between theory and experiment was good (Sanchez and Garland, 1983).

Heat capacity at constant pressure of a critical 3-methylpentane - nitroethane binary-liquid mixture has been measured using a computerized high-resolution calorimetric technique by Sanchez and his team. The critical exponent α , correlation length ξ and the amplitude of the heat capacity at constant pressure C_p were calculated. The results were in agreement with the renormalization-group theory (Sanchez *et al*, 1983).

The ultrasonic wave attenuation for the critical mixture of triethylamine and water has been measured by Fast and Yun. The experimental values were compared to the calculated values based on the dynamic scaling theory and good agreement was achieved (Fast and Yun, 1985).

P`epin and his team have reported the refractive index data for the binary mixture triethylamine and water as a function of temperature near the lower critical solution temperature. They were arrived for the first time at an unambiguous evidence for an intrinsic refractive index anomaly in critical binary liquid mixture (P`epin *et al*, 1988).

The ultrasonic velocity, absorption and shear viscosity have been measured as a function of temperature and concentration for the binary aqueous solutions of the polymer polyvinylpyrrolidone by Spickler and coworkers. The ultrasonic velocity showed a nonlinear increase with temperature and a near linear increase with concentration. Viscosity values were observed to increase monotonically with concentration and decrease with temperature (Spickler *et al*, 1989).

The ultrasonic velocity and absorption as a function of temperature, concentration, and frequency in the range (5 – 25 MHz), and shear viscosity as a function of concentration and temperature were measured for the binary liquid mixture nitrobenzene - n- hexane in the homogenous phase above the critical temperature by Abdelraziq and his team. The observed absorption at the critical point $\frac{\alpha_c}{f^2}$ vs $f^{-1.06}$ showed a straight line as predicted by the dynamic scaling theory. The ultrasonic velocity of the system at the critical concentration above the critical temperature was decreasing linearly with increasing temperature (Abdelraziq *et al*, 1990).

The temperature and frequency dependence of the ultrasonic absorption for the binary mixture of critical composition isobutyric acid and water were analyzed in terms of the dynamic scaling theory by Woermann. A

temperature and frequency dependent ultrasonic background attenuation was observed (Woermann, 1991).

Abdelraziq and his team have measured the ultrasonic velocity and absorption at the critical concentration as a function of temperature and frequency for the binary mixture carbon tetrachloride and coconut oil. They found that the absorption coefficient for the critical concentration increased with decreasing temperature until the critical temperature was approached for all frequencies. At the critical temperature and concentration the absorption coefficient $\alpha(\text{crit}, T_c)$ decreased with increasing frequency (Abdelraziq *et al*, 1992).

Viscosity measurements of the ionic binary mixture ethyl ammonium nitrate in n-octanol in the temperature range $0.01 < T - T_c < 19.4$ K near the critical consolute point were obtained by Oleinikova and Bonetti. The data were consistent with the power laws predicted by the mode coupling and dynamic renormalization group theories (Oleinikova and Bonetti, 1996).

Ultrasonic absorption was measured as a function of temperature and frequency for the binary mixture cyclohexane and aniline by Abdelraziq. The data were analyzed based on the dynamic scaling theory. It was concluded that the absorption coefficient decreased with increasing temperature. The adiabatic coupling constant has a negative sign which implies that the phase separation could be induced by a sudden decrease of the pressure (Abdelraziq, 1996).

The dynamic renormalization group theory has been applied to explain the sound behavior by Folk and Moser. Temperature and frequency

dependence of the sound velocity and absorption near the critical point in pure fluids and mixtures were investigated (Folk and Moser, 1999).

The ultrasonic absorption and velocity of the mixture perfluoromethyl cyclohexane and carbon tetrachloride have been measured by Abdelraziq. It has been found that the absorption coefficient for the critical concentration increased with decreasing temperature. The ultrasonic velocity increased with increasing frequency. The adiabatic coupling constant has a positive sign which implies that the phase separation could be induced by a sudden increase of the pressure (Abdelraziq, 1999).

Ultrasonic velocity measurements in the temperature range 211 - 303K whereas absorption coefficient measurements in the frequency range 5-43MHz and temperature range $0.15 \leq T - T_c \leq 20$ K of the benzonitrile-isooctane critical mixture were made by Hornowski and Madej. The acoustic attenuation results were compared with the scaling function of Shiwa and Kawasaki mode coupling theory, good agreement was achieved only on lower frequency range. In addition, the comparison with Kroll and Ruland renormalization group theory and Ferrell and Battacharjee dynamic scaling theory showed very good matching on all frequency range (Hornowski and Madej, 2001).

Abdelraziq used the experimental values of the critical amplitude of acoustical attenuation from the mode coupling theory of the binary liquid mixture aniline-cyclohexane to compare Hornowski's theoretical modification of the critical amplitude with Fixman, Kawasaki, Mistura and

Chaban theoretical expressions. The experimental results were found to agree well with Hornowski's model at low reduced frequencies ω^* .

On the other hand, for large values of the reduced frequency the mode coupling theory of Shiwa and Kawasaki showed poor agreement with the observed data (Abdelraziq, 2001).

Shear viscosity coefficients of the binary mixture nitroethane and 3-methylpentane were measured by Abdelraziq. The results were analyzed using the mode coupling theory while shear viscosity anomaly was detected as a function of temperature and concentration (Abdelraziq, 2002).

Abdelraziq confirmed the relationship called two scale factor universality by studying different binary liquid mixtures nitrobenzene - n-hexane, methanol - n-heptane, methanol - cyclohexane and nitrobenzene - n-heptanes. He calculated the universal quantity R for each mixture (Abdelraziq, 2003).

Ultrasonic velocity studies were carried out at a frequency of 2 MHz using ultrasonic pulse echo system on cresols in ethylacetate at constant temperature of 311K by Nayakulu and his team. A linear correlation between the ultrasonic velocity and acidity constant was detected (Nayakulu *et al*, 2005).

Ultrasonic absorption and velocity measurements were made as a function of temperature and frequency for the binary mixtures; benzene - coconut oil and hexane - β , β' -dichloethyl ether by Abdelraziq. The ultrasonic absorption was found to exhibit a strong temperature and frequency dependence near the critical temperature while the ultrasonic velocity

behaved as a linearly decreasing function of temperature above the critical value (Abdelraziq, 2005).

The densities and viscosities of binary liquid mixtures of diethyl oxalate and dimethyl malonate with polar and nonpolar solvents, ethanol, acetone, chloroform, carbon tetrachloride, benzene and toluene were measured as a function of composition of the corresponding binary mixtures. It was found that viscosity deviations of binary mixtures of diethyl oxalate with ethanol, acetone, chloroform, and carbon tetra chloride were positive over the entire range of composition, while for its mixtures with benzene and toluene the deviations were negative over the entire range of composition. For the binary mixtures of diethyl malonate with acetone, chloroform, carbon tetrachloride and benzene the viscosity deviations were positive over the entire range of composition, while the values were negative for its mixtures with ethanol and toluene (Diwedi and Singh, 2007).

The viscosity and density of a binary liquid mixture of diacetone alcohol with benzene and chlorobenzene were measured by Kubendran and Baskaran. It was concluded that as the temperature increases the intermolecular interactions between molecules become weak. It was observed that intermolecular interactions were more with chlorobenzene than with benzene (Kubendran and Baskaran, 2008).

Ultrasonic velocity, absorption and shear viscosity were measured in aqueous solution of poly ethylene glycol by Singh and his team. The ultrasonic velocity was observed to increase with temperature at a given concentration, while as the concentration was increased at a given

temperature the velocity was increased. The shear viscosity is decreasing with increasing temperature (Singh *et al*, 2008).

Ultrasonic velocity, density, and viscosity were measured for the solutions of polyvinyl acetate by Singh and Bhatt. Various acoustical parameters were calculated; like adiabatic compressibility, acoustic impedance, and ultrasonic attenuation. The study revealed that ultrasonic velocity and acoustic impedance decreased with temperature, and increased with concentration. The adiabatic compressibility increased with temperature and decreased with concentration. Ultrasonic attenuation decreased with temperature and increased with concentration (Singh and Bhatt, 2010).

Ultrasonic velocity and density were determined experimentally for the binary liquid mixtures of methyl methacrylate with alcohols by Vadamar and his team. The presence of dipole - dipole interactions was observed throughout the study due to the polar nature of alcohols (Vadamalar *et al*, 2011).

The ultrasonic attenuation spectra of nitrobenzene - hexane mixture of critical composition was analyzed in the frequency range 10 KHz- 1 GHz by Mirzaev and Kaatze. They showed that at agiven temperature the spectra revealed a none critical relaxation term, in addition to the critical contribution (Mirzaev and Kaatze; 2012).

Sandhya and his team compared the experimental ultrasonic sound velocity in binary liquid mixtures with theoretical models of Nomoto, Van Deal, and Vangal model at different temperatures. Their results supported theoretical models (Sandhya, 2013).

The ultrasonic velocity, viscosity, and density were measured for the binary system tetrahydrofuran - ethanol. The measured values were used to evaluate different acoustical parameters, such as adiabatic compressibility, intermolecular free length, acoustical impedance, and internal pressure (Bhandakkar *et al*, 2014).

The dynamic shear viscosity for the binary mixtures of coconut oil with carbon tetrachloride and benzene was measured as a function of temperature by Kittany and Abdo. Debye momentum cut off was calculated using the mode coupling theory (Kittany, 2014; Abdo, 2014).

Dynamic shear viscosity for the binary mixture methanol and cyclohexane for different temperatures and concentrations was measured by Omar. The dynamic scaling theory of Ferrell and Bhattacharjee has been applied to calculate the adiabatic coupling constant g , isobaric thermal expansion coefficient α_p and diffusion coefficient D (Omar, 2014; Ferrell and Bhattacharjee, 1981).

1.3. Objectives and Motivations

Ultrasonic spectroscopy is an excellent non-destructive technique for characterizing the various aspects of physiochemical behavior of liquid mixtures. It helps in bringing out the facts, which can have positive implications of both industry as well as the theory of building process.

The binary mixture phenol C_6H_5OH and water has great importance. There is no adequate information about its physical properties that lead us to study some of its properties.

The main goal of this work is to study the ultrasonic attenuation for the binary mixture of water and phenol at the critical temperature and concentration applying the dynamic scaling theory. The critical temperature and concentration will be determined. Viscosity of the binary mixture will be measured as a function of temperature; in addition the specific heat and isobaric thermal expansion coefficient will be calculated.

1.4. Thesis Organization

The thesis is divided into five chapters. Chapter one is the introduction which presents the definition of binary liquid mixtures and also provides a review of ultrasonic studies in those mixtures reported in the literature.

Chapter two discusses the dynamic scaling theory of Ferrell and Bhattacharjee which was applied for analyzing the ultrasonic data.

Methodology of the work is given in chapter three, while the measured data and the results in chapter four. In chapter five conclusions are displayed.

Chapter Two

Theory

Ultrasonic is the branch of acoustics dealing with sound above the audible range (Rajathi *et al*, 2011). Depending on the frequency, ultrasound is divided into three classes, namely power ultrasound (20–100 kHz), high frequency ultrasound (100 kHz–1 MHz), and diagnostic ultrasound (1–500 MHz) (Wu *et al*, 2012).

2.1 Ultrasonic Attenuation

Ultrasound is attenuated as it travels through a medium. Two main mechanisms contribute to ultrasound attenuation: absorption and scattering. There are three sources of absorption: viscosity, heat conduction, and inter molecular processes.

Viscous absorption occurs when ever there is a relative motion between adjacent portions of the medium, such as during shear deformation or the compressions and expansions that accompany the sound transmission. Heat conduction absorption results from the conduction of thermal energy from higher temperature condensations to lower temperature rarefactions. Molecular processes leading to absorption include the conversion of kinetic energy of the molecules into; stored potential energy as in structural rearrangement or rotational and vibrational energies for poly atomic molecules(Kinsler,1999).

2.2The Dynamic Scaling Theory

Ferrell and Bhattacharjee have presented a general theory of the critical ultrasonic attenuation, based on an extension of the concept of frequency-dependent specific heat.

The total attenuation, at the critical temperature, can be expressed as:

$$\frac{\alpha(\text{crit}, T_c)}{f^2} = S f^{-1.06} + b \quad (2.2.1)$$

Where, b is the contribution of the frequency independent background absorption ($\frac{\alpha(\text{back})}{f^2}$). The value of S is given by:

$$S = \left[\frac{\pi^2 C_{pc} g^2 v_c \acute{\alpha}}{2z\gamma T_c C_p^2(t_f)} \right] \left[\frac{\alpha \omega_0}{2\pi} \right]^{z\gamma} \quad (2.22)$$

Where, $\acute{\alpha} = 0.11$ and $z\gamma = 1.9$, both are critical exponents, and $C_p(t_f)$ is the specific heat at a characteristic reduced temperature t_f which can be approximated by $t = \left(\frac{T - T_c}{T_c} \right)$ value at which $\frac{\alpha(\text{crit}, T_c)}{f^2}$ for a given frequency is one-half its value at T_c , $a = \frac{\omega}{\omega_0} t_f^{-1.9}$ is dimensionless scaling factor of order unity, ω_0 is characteristic temperature dependent relaxation rate, g is adiabatic coupling constant, v_c is adiabatic sound velocity at T_c , C_{pc} is the critical amplitude in the expression for specific heat at constant pressure of a mixture of critical composition which is:

$$C_p = C_{pc} t^{-\acute{\alpha}} + C_{pb} \quad (2.2.3)$$

C_{pb} is the background specific heat at constant pressure (Abdelraziq, 1999; Iwanowski, 2007).

The adiabatic coupling constant g is a dimensionless parameter describing the magnitude of coupling between critical density fluctuations and the propagating sound wave; it was introduced by Ferrell and Bhattacharjee and is given by:

$$g = \rho_c C_p \left(\frac{dT_c}{dP} - \frac{T \alpha_p}{\rho C_p} \right) \quad (2.2.4)$$

$$\text{Or} \quad g = \left(\frac{C_{pb} \alpha_{pc} T_c}{C_{pc}} \right) - \alpha_{pb} T \quad (2.2.5)$$

Where, ρ_c is the critical density, α_p is isobaric thermal expansion coefficient which can be expressed as:

$$\alpha_p = \alpha_{pc} t^{\alpha'} + \alpha_{pb} \quad (2.2.6)$$

Where, α_{pc} and α_{pb} are the critical and background terms of isobaric thermal expansion coefficients respectively (Iwanowski, 2007; Bhattacharjee *et al*, 2010).

The theory indicates that $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ should be a function of the reduced frequency ω^* and the relation takes the form:

$$\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)} = F(\omega^*) = (1 + \omega^{*-0.5})^{-2} \quad (2.2.7)$$

Where, $F(\omega^*)$ is the theoretical scaling function predicted by Ferrell and Bhattacharjee. $\alpha(x_c, T)$ is the absorption coefficient at the critical concentration and temperature T above the critical temperature and $\alpha_c(x_c, T_c)$ is the absorption coefficient at critical temperature and critical concentration.

The expression of the reduced frequency ω^* is:

$$\omega^* = \frac{\omega}{\omega_D} = \frac{2\pi f}{\omega t^{z\gamma}} \quad (2.2.8)$$

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

$$\omega_D = \frac{K_B T}{3\pi \eta \xi^3} = \frac{K_B T_c}{3\pi \eta_0 \xi_0^3} t^{z\gamma} = \omega_0 t^{z\gamma} \quad (2.2.9)$$

$$\omega_0 = 2D_0 \xi_0^{-2} \quad (2.2.10)$$

Where, K_B is the Boltzmann's constant, ξ is the correlation length and η is the dynamic shear viscosity, ξ_0 is the critical amplitude of the

correlation length, η_0 is the noncritical part of the dynamic shear viscosity and D_0 is the diffusion coefficient in $\frac{\text{cm}^2}{\text{s}}$.

The correlation length is given by:

$$\xi = \xi_0 t^{-\gamma} \quad (2.2.11)$$

and the shear viscosity is also given by:

$$\eta = \eta_0 t^{-x_\eta \gamma} \quad (2.2.12)$$

Where, x_η is a critical exponent = 0.06, $x_\eta \gamma$ is a critical exponent = 0.037 and $\gamma = 0.64$, (Arrigo *et al*, 1977; Ferrell and Bhattacharjee, 1981; Abdelraziq, 1999).

The isothermal β_T and adiabatic β_s compressibilities and specific heat at constant volume C_v can be expressed under the assumptions that all the quantities are expressed as power laws of the form:

$$\beta_T = \beta_{Tc} t^{-\alpha} + \beta_{Tb} \quad (2.2.13)$$

$$\beta_s = \beta_{sc} t^{-\alpha} + \beta_{sb} \quad (2.2.14)$$

$$C_v = C_{vc} t^{-\alpha} + C_{vb} \quad (2.2.15)$$

Where, β_{Tc} is the critical isothermal compressibility, β_{sc} is the critical adiabatic compressibility, C_{vc} is the critical specific heat at constant volume, β_{Tb} is the background isothermal compressibility, β_{sb} is the background adiabatic compressibility and C_{vb} is the background specific heat at constant volume (Abdelraziq, 1999).

Chapter Three

Methodology

The samples of the binary liquid mixture water - phenol were prepared in the laboratory at different concentrations from 0% to 100% by weight of phenol. Viscosity measurements were carried out with the glass capillary and digital viscometer. The experimental data were analyzed and the critical temperature and concentration were obtained.

The glass equipments were cleaned using hot soapy water and then rinse with distilled water before and after use. Acetone and ethanol were used for a final rinse of urgently needed glasses to help drying them quickly.

3.1 Water and Phenol

1. Water: distilled water was used in this work, it is a colorless liquid without odor or taste with the molecular formula H_2O . The molecular structure of water is shown in Fig.(3.1).

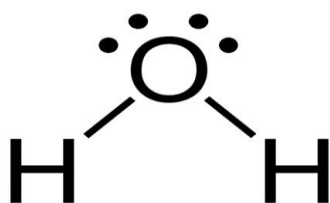


Fig.(3.1): Molecular structure of water.

2. Phenol: with purity (99.5% - 100%) was used in this work without any further purification. Phenol is an organic compound with the molecular formula $C_6 H_5 OH$, it looks as white crystalline at room temperature smelling of disinfectant. The molecular structure of phenol is shown in Fig.(3.2).

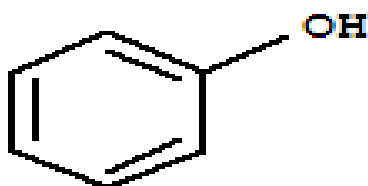


Fig.(3.2): Molecular structure of phenol.

Some physical properties of water and phenol are given in Table 3.1 (Pilato, 2010).

Table 3.1: Some physical properties of water and phenol

Property	Water	Phenol
Molar Mass	18.015 g /mol	94.1 g /mol
Color	White solid, Colorless liquid	White solid, Colorless liquid
Density (20°C)	0.9982 g / cm ³	1.071g / cm ³
Melting Point	0 °C	40.9 °C
Boiling Point	100 °C	181.8 °C
Flash Point	—	82 °C
Solubility in water	Universal solvent	8.3 g/100 mL (20 °C)

3.2 Experimental Apparatus

3.2.1 Density Measurements:

The following instruments were used to measure the density:

- HX-Z Electronic Balance (Fig.3.3.a): it is used to measure the mass of the liquid with accuracy ± 0.0005 .
- The 10 ml glass Picnometer (Fig.3.3.b): it is used to measure the accurate volume of the liquid.

The densities for both liquids water and phenol were measured separately at room temperature, and the mixture density was measured at temperature range $32 \leq T \leq 75$ °C for each concentration.



Fig.(3.3): (a) Electronic Balance (b) Picnometer to measure the density.

The density ρ is given by:

$$\rho(\text{gm/cm}^3) = \frac{\text{mass of the liquid}}{\text{volume of the liquid}}$$

The mass of the liquid is the resultant of subtraction of the filled and empty Picnometer measured masses.

The measured average densities for the two liquids at temperature 32 °C are $\rho_{phenol} = 1.1437 \text{ gm/cm}^3$ and $\rho_{water} = 0.9987 \text{ gm/cm}^3$.

3.2.2 Viscosity Apparatus:

Two models of viscometers were used:

- A Brookfield Digital Viscometer Model DV-I+ (Fig.3.4.a) with accuracy $\pm 1\%$ has been used; with set of seven spindles (RV Spindle Set) and UL-ADAPTER. The speeds of the rotated spindle are in two sets. The first set is 0.0, 0.5, 1.0, 2.0, 2.5, 4, 5, 10, 20, 50 and 100 rpm, the second set is 0.0, 0.3, 0.6, 1.5, 3, 6, 12, 30 and 60 rpm. The spindles measure viscosity range from 100 cP up to

1.33×10^7 cP. The UL-spindles measure viscosity range from 1cP up to 2000 cP (Brookfield, 1999).

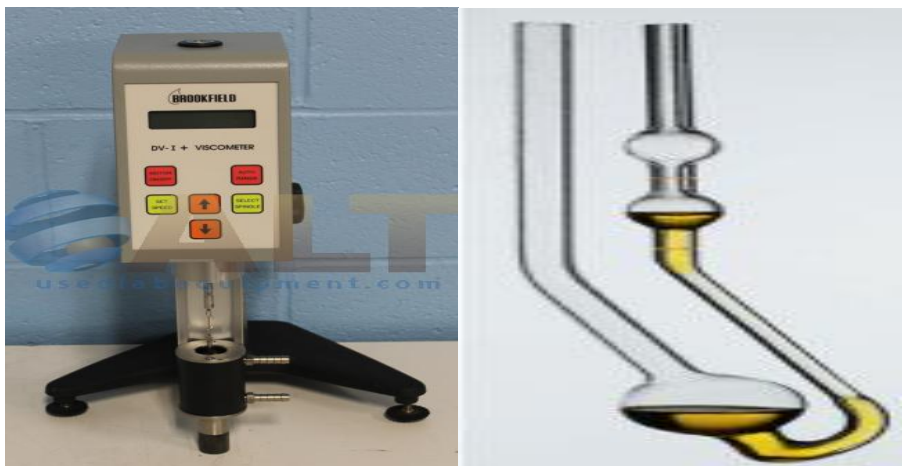
- A Glass Capillary Viscometer (Fig.3.4.b) with accuracy of $\pm 0.2\%$ consists of a U-shaped glass tube having a capillary inside it with glass bulbs. The lower bulb is filled with sample and then the sample suctioned into the double bulb side by pipette filler (Fig.3.5).

The viscosity of the sample was determined by measuring the time of flow between timing marks using an accurate stop watch with accuracy ± 0.00005 .

The viscosity is determined using the formula:

$$\eta_1 = \eta_2 \frac{\rho_1 t_1}{\rho_2 t_2}$$

Where, η_1 , ρ_1 , t_1 are the dynamic shear viscosity, mass density and time of flow of the reference liquid which is water. The values, η_2 , ρ_2 , t_2 are the dynamic shear viscosity, mass density and time of flow of the binary liquid mixture water - phenol (Generalic, Eni., 2014).



(a) **(b)**
Fig. (3.4): (a) Brookfield Digital Viscometer Model DV-I+
 (b) The Glass Capillary Viscometer.



Fig. (3.5): Pipette Filler

3.2.3 Temperature Apparatus

- Julabo F25-MV Refrigerated and Heating Circulator (Fig.3.6.a) with accuracy $\pm 1\%$ is used to control the temperature of the samples in the UL-ADAPTER and Capillary viscometer.
- The temperature of the samples was measured using a Digital Prima long Thermometer (Fig.3.6.b) with accuracy $\pm 1\%$ and it measures temperature from -50 to $+100$ °C.



(a)



(b)

Fig.(3.6): (a) Julabo F25-MV Refrigerated and Heating Circulator

(b) Digital Prima Long Thermometer.

3.3 Samples Preparation:

The mass concentration was calculated using the formula:

$$x_{\text{phenol}}\% = \frac{\text{mass of phenol}}{\text{mass of the mixture}} = \frac{(\rho v)_{\text{phenol}}}{(\rho v)_{\text{phenol}} + (\rho v)_{\text{water}}}$$

Where, ρ is the density and v is the volume.

The total volume of the sample was 20 ml, and the concentrations were 0.00%, 10.17%, 20.80%, 30.20%, 32.90%, 33.90%, 35.00%, 37.10%, 40.20%, 49.80%, 59.70%, 70.40%, 79.77%, 89.90% and 100.00% by weight of phenol.

Chapter Four

Results and Analysis

4.1 Viscosity Measurements

The dynamic shear viscosity values for the binary liquid mixture water and phenol have been measured three times for different concentrations (0.00% - 100.00%) by weight of phenol at temperature range (32-75°C) and the average viscosities are given in Tables 4.1 and 4.2.

Table (4.1): Dynamic viscosity measurements at different temperatures and concentrations (0.00% - 37.00%) for the binary liquid mixture water - phenol.

η (cP)									
T (°C)	0.00%	10.17%	20.80%	30.20%	31.80%	32.90%	33.90%	35.00%	37.10%
32	0.765	0.87	0.99	1.87	2.20	2.62	2.50	3.33	3.09
40	0.653	0.80	0.83	2.14	1.95	2.05	2.28	2.75	2.46
45	0.596	0.71	0.75	2.36	1.96	1.88	1.96	2.45	2.20
50	0.547	0.64	0.71	1.83	1.75	1.61	1.75	2.00	1.94
55	0.504	0.60	0.63	1.66	1.48	1.46	1.53	1.92	1.73
57	0.489	0.56	0.60	1.40	1.41	1.35	1.46	1.72	1.75
58	0.481	0.55	0.58	1.34	1.32	1.32	1.43	1.72	1.68
59	0.474	0.55	0.59	1.30	1.33	1.25	1.31	1.69	1.59
60	0.467	0.55	0.57	1.37	1.31	1.25	1.39	1.74	1.53
61	0.460	0.53	0.55	1.25	1.29	1.21	1.32	1.60	1.40
62	0.453	0.53	0.54	1.13	1.23	1.17	1.23	1.53	1.54
63	0.447	0.50	0.53	0.90	1.18	1.11	1.11	1.41	1.38
64	0.440	0.43	0.46	0.95	1.01	0.93	0.95	1.19	1.25
65	0.434	0.42	0.46	0.85	0.98	0.87	1.07	1.04	1.16
66	0.428	0.41	0.46	0.73	0.97	0.88	1.09	0.97	1.22
67	0.422	0.40	0.45	0.64	0.94	0.90	1.10	0.97	1.14
68	0.416	0.40	0.44	0.66	0.92	0.88	0.99	0.87	1.07
69	0.410	0.40	0.37	0.56	0.87	0.90	0.73	0.85	1.10
70	0.404	0.36	0.38	0.54	0.84	0.70	0.75	0.81	1.00
75	0.378	0.35	0.37	0.50	0.72	0.58	0.68	0.70	0.89

Table (4.2): Dynamic viscosity measurements at different temperatures and concentrations (40.20% - 100.00%) for the binary liquid mixture water – phenol.

η (cP)	40.20%	49.80%	59.79%	70.40%	79.77%	89.90%	100.00%
T (°C)							
32	2.25	2.64	3.39	3.15	3.31	3.33	Solid
40	1.87	2.07	2.60	2.50	2.73	2.61	4.28
45	1.86	1.89	2.59	2.26	2.44	2.35	3.67
50	1.58	1.72	2.35	2.02	2.21	2.03	3.29
55	1.41	1.56	1.97	1.87	2.10	1.93	2.93
57	1.34	1.46	1.89	1.86	2.02	1.85	2.68
58	1.29	1.48	1.81	1.74	2.00	1.82	2.70
59	1.25	1.42	1.77	1.73	1.90	1.81	2.73
60	1.28	1.34	1.70	1.72	1.85	1.81	2.53
61	1.15	1.26	1.64	1.72	1.90	1.80	2.45
62	1.16	1.24	1.58	1.63	1.78	1.76	2.42
63	1.03	1.17	1.49	1.61	1.59	1.64	2.28
64	0.87	0.98	1.29	1.24	1.38	1.35	2.17
65	0.94	0.98	1.24	1.16	1.21	1.34	1.84
66	0.90	0.93	1.21	1.14	1.20	1.14	1.82
67	0.85	0.90	1.20	1.12	1.17	1.12	1.79
68	0.83	0.89	1.16	1.11	1.16	1.08	1.71
69	0.84	0.85	1.17	1.10	1.17	1.07	1.69
70	0.82	0.80	1.09	1.00	1.05	1.01	1.77
75	0.75	0.71	1.01	0.88	0.925	0.946	1.60

Tables 4.1 and 4.2 show that in general at each concentration of phenol, the dynamic shear viscosity of the binary liquid mixture water - phenol decreases as the temperature increases. It is also observed that some values of the dynamic shear viscosity were increased other than decreased with increasing temperature, that mostly due to contamination or invisible particles adhere to the capillary wall, that leading to an increase of the measured flow times (section 3.2.2).

The dynamic shear viscosities of water - phenol are plotted as a function of temperature at different concentrations of phenol (10.17% - 90%) and shown in Figs. (4.1) – (4.3).

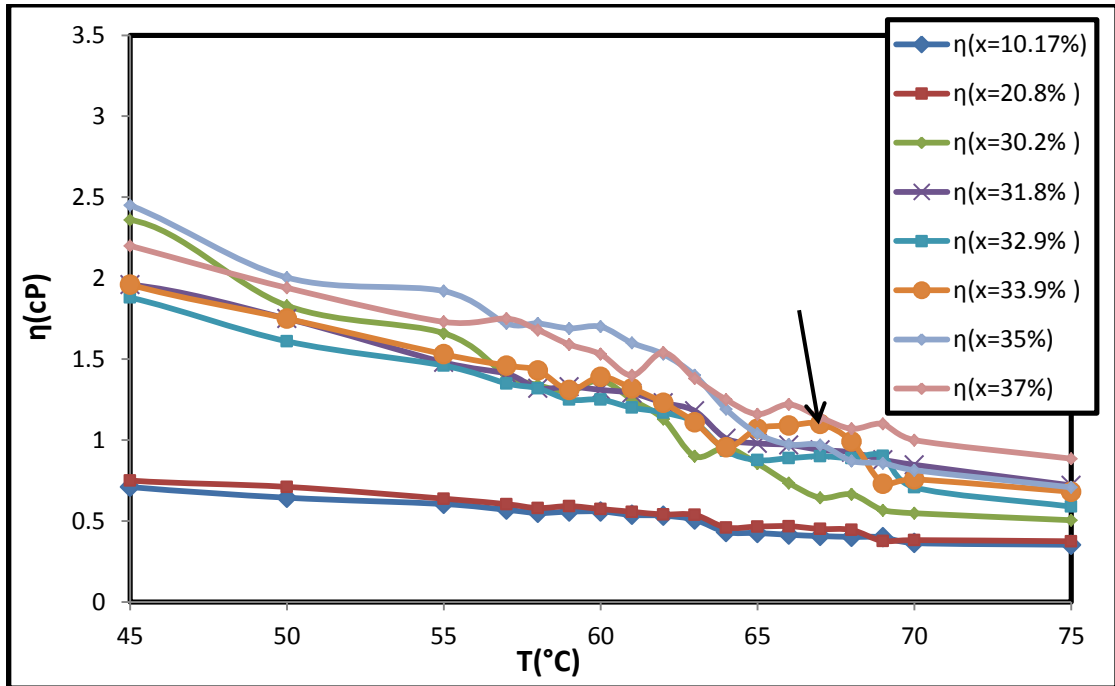


Fig. (4.1): The dynamic shear viscosity of water - phenol mixture as a function of temperature at (10.17 - 37%) by weight of phenol.

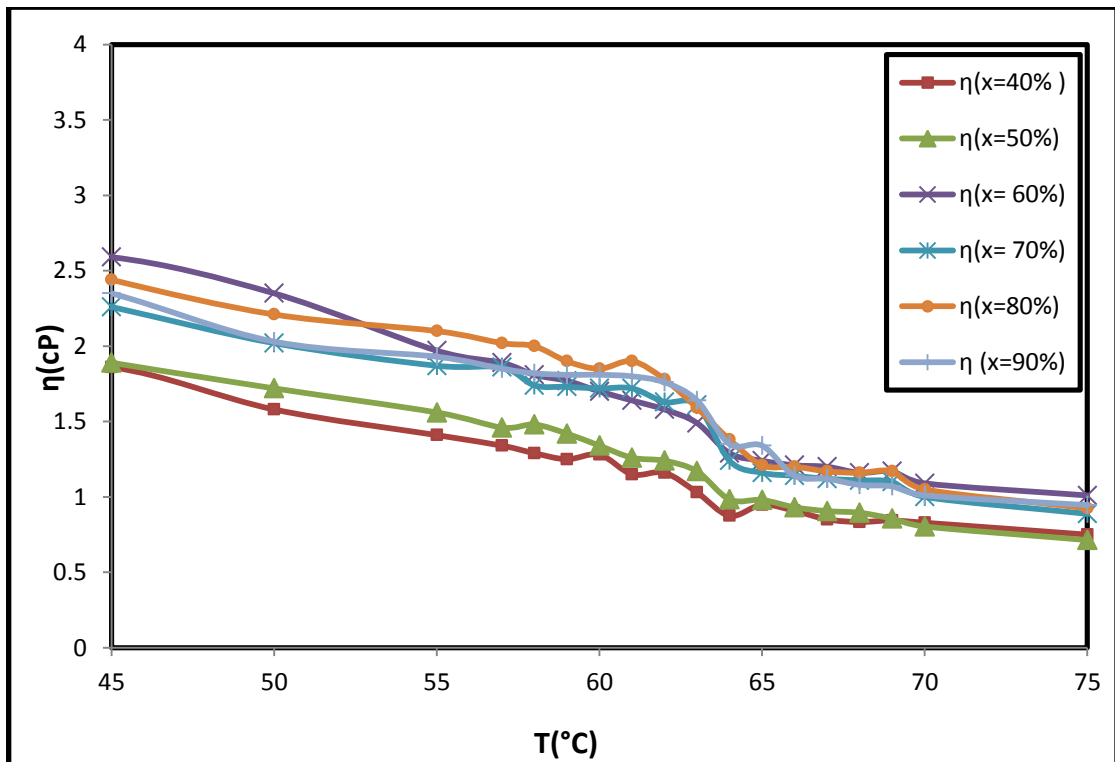


Fig.(4.2): The dynamic shear viscosity of water - phenol mixture as a function of temperature at (40% - 90%) by weight of phenol.

The anomaly can be detected by plotting 32.90%, 33.90% and 35.00% by weight of phenol at temperature range (60.0 – 75.0°C) as shown in Fig.(4.3):

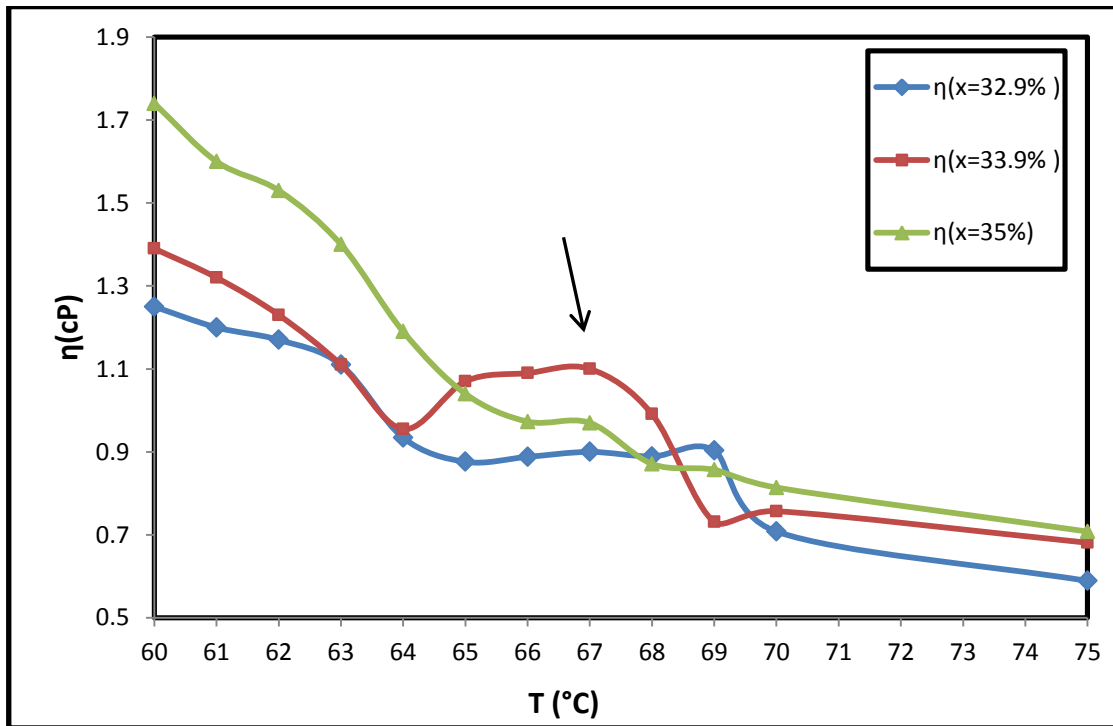


Fig.(4.3): The dynamic shear viscosity of water - phenol mixture as a function of temperature at 32.90%, 33.90% and 35.00% by weight of phenol.

Figs. (4.1) and (4.3) show an observed anomaly of the dynamic shear viscosity of water - phenol mixture at concentration $x_c = 33.90\%$ by weight of phenol and temperature $T_c = 67.0^\circ\text{C}$, where the value of the viscosity is 1.10 cP.

Our experimental results showed a good agreement with Howell experimental results in the critical temperature which was found to be $T_c = 66.4^\circ\text{C}$ but the critical concentration was found by Howell to be $x_c = 34.6\%$ by weight of phenol (Howell, 1932). The result was also in good agreement

with Krishnan experimental result of the critical temperature $T_c = 69^\circ\text{C}$ where the concentration $x_c = 34\%$ by weight of phenol (Krishnan, 1935). Our result was also in good agreement with Campbell measurements of the critical temperature which is 66.8°C . The concentration value was found to be 34.5% by weight of phenol (Campbell Alan N. and Campbell A. Jean R., 1973).

The dynamic viscosity for concentrations 0.00% and 100.00% of phenol are plotted as a function of temperature in Fig.4.4:

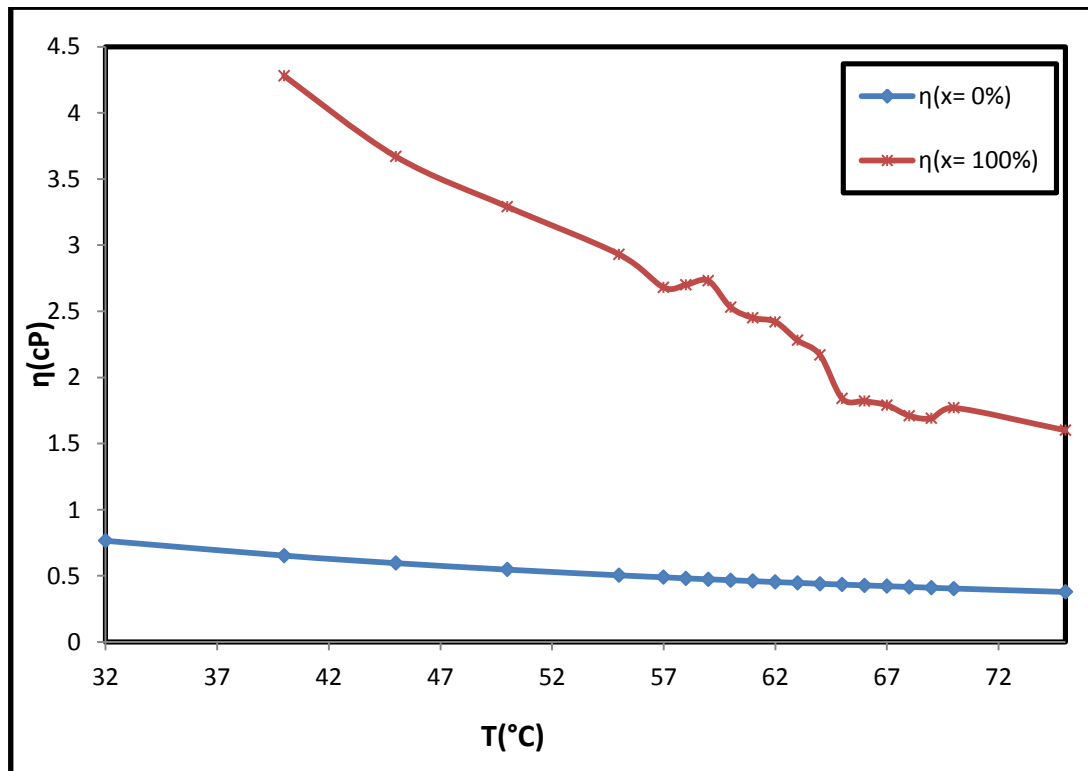


Fig.(4.4): The dynamic shear viscosity of the pure liquids water and phenol as a function of temperature .

Fig.(4.4) shows the dynamic shear viscosity of pure liquids, water ($x = 0.00\%$), and phenol ($x = 100.00\%$).

4.2 Calculation of Specific Heat of the Critical Mixture

The Two Scale Factor Universality is used to find the specific heat at constant pressure at the critical temperature c_{pc} of water - phenol binary mixture.

The universal constant R is given by:

$$R = \xi_0 \left\{ \frac{\alpha' \rho_c c_{pc}}{K_B} \right\}^{1/3} = 0.27 \quad (4.2.1)$$

Where: $\alpha' = 0.11$, $K_B = 1.3806 \times 10^{-23}$ (J/K), and $\xi_0 = 2.2 \times 10^{-10}$ m (Jacobs, 1986; Abdelraziq, 2015 to be published).

The density ρ_c of the critical mixture water - phenol was measured at different temperatures above the critical point as mentioned in section 3.2.1. The results are given in Table (4.3).

Table (4.3): The measured densities at different temperature above the critical point of the binary liquid mixture water- phenol.

T (°C)	ρ ($\frac{\text{gm}}{\text{cm}^3}$)
67.0	0.959
67.2	0.954
67.4	0.949
67.6	0.938
67.8	0.928
68.4	0.922
68.7	0.918

The critical density ρ_c at critical temperature and critical concentration equals to $0.959 \frac{\text{gm}}{\text{cm}^3}$. Using equation 4.2.1 the critical specific heat at constant pressure for the critical mixture water - phenol is calculated to be $241.9 \frac{\text{J}}{\text{kg K}}$.

4.3 Calculation of Isobaric Thermal Expansion Coefficient

The isobaric thermal expansion coefficient α_p can be calculated using the relation:

$$\alpha_p = \frac{1}{v} \left(\frac{dv}{dT} \right)_p \quad (4.3.1)$$

Where,

v is the volume, T is the temperature and P is the pressure. Equation 4.3.1 could be expressed in another form by applying $v = \frac{m}{\rho}$.

$$\alpha_p = -\frac{1}{\rho} \left(\frac{d\rho}{dT} \right)_p = \rho \left(\frac{d\rho^{-1}}{dT} \right) \quad (4.3.2)$$

Where, m is the mass and ρ is the density.

The values of the density and its reciprocal at different temperatures above the critical point are presented in Table (4.4).

Table (4.4): The mass density and its reciprocal values at different temperatures for the critical mixture water and phenol.

T(°C)	Mass density ρ (gm/cm ³)	ρ^{-1} (cm ³ /gm)
67.2	0.954	1.054
67.4	0.949	1.053
67.6	0.938	1.066
67.8	0.928	1.077
68.4	0.922	1.084
68.7	0.918	1.089

The reciprocal of the density is fitted with the corresponding temperatures and the slope $\left(\frac{d\rho^{-1}}{dT} \right)$ is determined from Fig.(4.5).

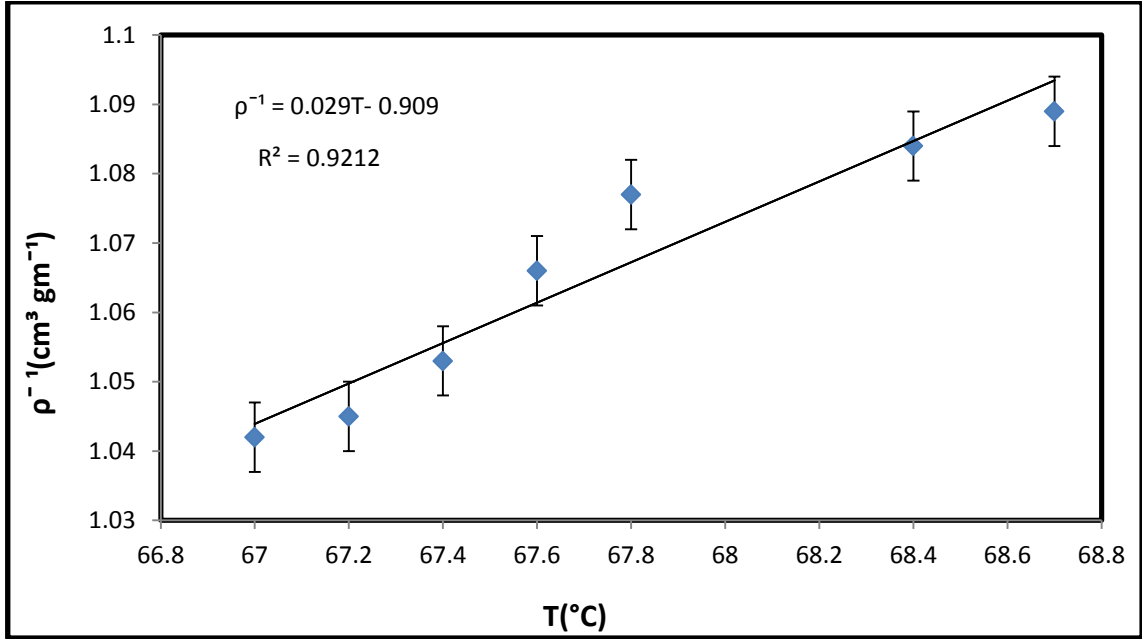


Fig.(4.5): The reciprocal of density for the critical mixture water-phenol as a function of temperature.

The slope from Fig.(4.5) is $\frac{d\rho^{-1}}{dT} = 0.029 \frac{\text{cm}^3}{\text{gm}^\circ\text{C}}$.

The critical and the background isobaric thermal expansion coefficients can be determined by linear fitting of the isobaric thermal expansion coefficient α_p versus $t^{-\alpha'}$, where $t = \frac{T-T_c}{T_c}$ and $\alpha' = 0.11$ depending on the relation:

$$\alpha_p = \alpha_{pc}t^{-\alpha'} + \alpha_{pb} \quad (4.3.3)$$

The values of α_p are calculated using equation (4.3.1) and the data are presented in Table (4.5).

Table (4.5): The isobaric thermal expansion coefficient at different temperatures for the critical mixture of water and phenol.

T(°C)	$t^{-\alpha'}$	α_p (°C ⁻¹)
67.2	1.895	0.0276
67.4	1.756	0.0275
67.6	1.670	0.0272
67.8	1.627	0.0269
68.4	1.530	0.0267
68.7	1.498	0.0266

The data of the isobaric thermal expansion coefficient α_p are plotted versus $t^{-\alpha}$ as shown in Fig.(4.6).

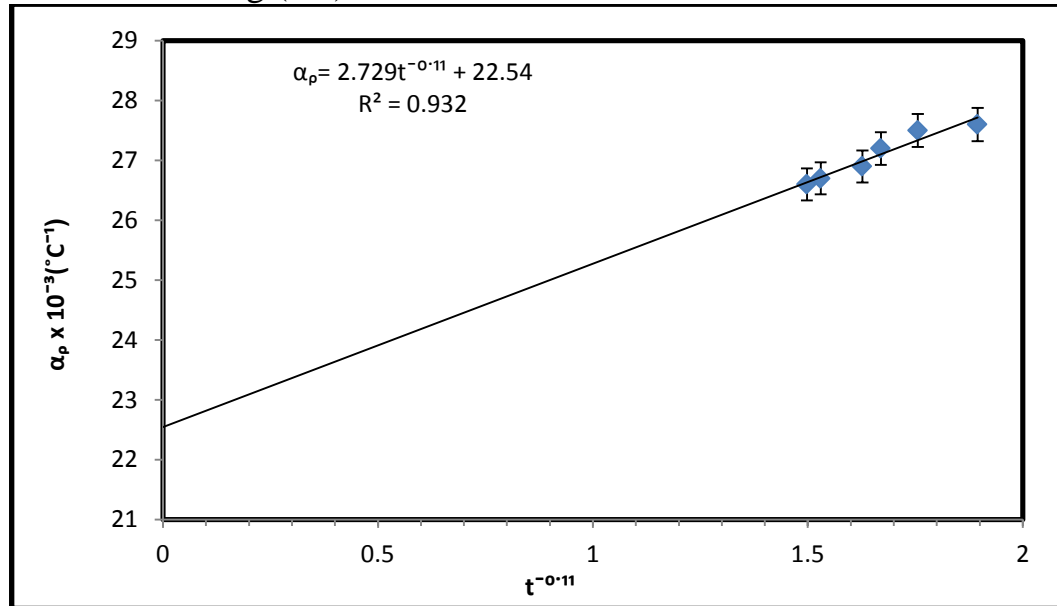


Fig.(4.6): The isobaric thermal expansion coefficient for the critical mixture water - phenol as a function of $t^{-0.11}$.

The slope of Fig.(4.6) represents the critical isobaric thermal expansion coefficient $\alpha_{pc} = 2.729 \times 10^{-3} \text{C}^{-1}$, and the intercept of the line represents the background isobaric thermal expansion coefficient $\alpha_{pb} = 22.54 \times 10^{-3} \text{C}^{-1}$ according to the equation (4.3.3), where:

$$\alpha_p = 2.729 \times 10^{-3} t^{-0.11} + 22.54 \times 10^{-3}$$

4.4 Ultrasonic Attenuation Results

The ultrasonic absorption measurements were made at the frequencies 9, 15, 30 and 35 MHz at the critical concentration of water - phenol mixture above the critical temperature. The data are given in Table (4.6):

Table (4.6): The absorption coefficient at different temperatures and frequencies 9, 15, 30, and 35 MHz for the critical mixture water - phenol.

T (°C)	$\frac{\alpha}{f^2} \times 10^{-12}$ At 9 MHz	$\frac{\alpha}{f^2} \times 10^{-12}$ At 15MHz	$\frac{\alpha}{f^2} \times 10^{-12}$ At 30MHz	$\frac{\alpha}{f^2} \times 10^{-12}$ At 35MHz
67.0	0.802	0.244	0.044	0.030
67.2	0.765	0.235	0.042	0.029
67.5	0.753	0.231	0.040	0.028
68.0	0.685	0.204	0.038	0.026
69.0	0.666	0.191	0.036	0.024
71.0	0.555	0.162	0.033	0.021
73.0	0.493	0.151	0.028	0.019
76.0	0.425	0.142	0.026	0.017
80.0	0.407	0.133	0.024	0.016

The temperature dependence of the absorption coefficient of the critical mixture water - phenol at different frequencies is shown in Fig.(4.7).

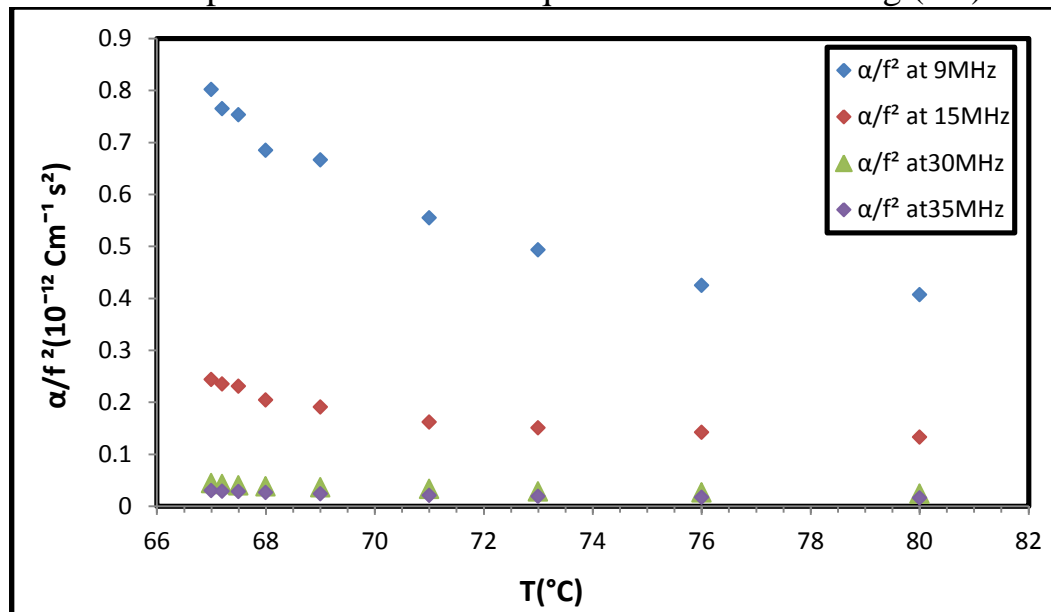


Fig.(4.7): The absorption coefficient $\frac{\alpha}{f^2}$ as a function of temperature for the critical mixture water - phenol at different frequencies.

Fig.(4.7) shows the ultrasonic absorption coefficient for the critical mixture water - phenol at different temperatures, at frequencies 9, 15, 30, and 35 MHz. It can be seen from Fig.(4.7) that the ultrasonic absorption

coefficient for the critical concentration decreases as the temperature increases for all frequencies.

The absorption coefficient at the critical temperature and concentration decreases with increasing frequency.

The dynamic scaling theory proposed a linear relation between $\frac{\alpha_c}{f^2}$ and $f^{-1.06}$ for different frequencies at the critical concentration and temperature. The values of the critical absorption coefficient $\frac{\alpha_c}{f^2}$ at different frequencies are given in Table (4.7):

Table (4.7): Values of the critical absorption coefficient at different frequencies for the critical mixture of water and phenol.

f(MHz)	$f^{-1.06}$ ($10^{-8} \text{ s}^{1.06}$)	$\frac{\alpha_c}{f^2}$ ($10^{-12} \text{ cm}^{-1} \text{ s}^2$)
9	4.251	0.802
15	2.473	0.59
30	1.186	0.42
35	1.007	0.33

The data of the absorption coefficient at the critical temperature and concentration $\frac{\alpha_c}{f^2}$ as a function of $f^{-1.06}$ are shown in Fig.(4.8).

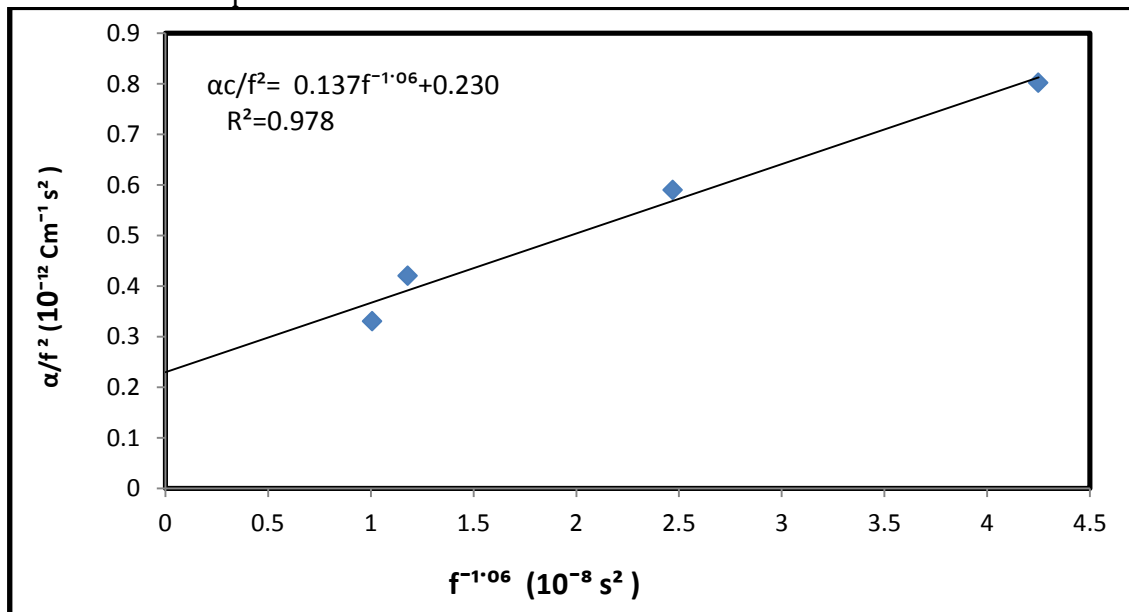


Fig.(4.8): The absorption coefficient $\frac{\alpha_c}{f^2}$ for the critical mixture water - phenol as a function of $f^{-1.06}$.

Fig.(4.8) shows a linear relation of $\frac{\alpha_c}{f^2}$ versus $f^{-1.06}$ as predicted by the dynamic scaling theory. The slope of Fig.(4.8) is $0.137 \times 10^{-8} \text{ cm}^{-1}\text{s}^{0.94}$ and the intercept which represents the frequency independent background part of $\frac{\alpha_c}{f^2}$ is $0.230 \times 10^{-12} \text{ cm}^{-1}\text{s}^2$. The slope in Fig.(4.8) can be defined as:

$$S = \left[\frac{\pi C_{pc} g^2 v_c \dot{\alpha}}{2z\gamma T_c C_p^2(t_f)} \right] \left[\frac{\alpha \omega_0}{2\pi} \right]^{\dot{\alpha}/z\gamma} \quad (4.4.1)$$

The temperature independent relaxation rate ω_0 is determined from the following equation:

$$\omega_D = \omega_0 t^{z\gamma} = \frac{K_B T_c}{3\pi\eta_0 \xi_0^3} t^{z\gamma} \quad (4.4.2)$$

$$\text{Where, } \omega_0 = \frac{1.3806 \times 10^{-23} \times 338.95}{3\pi \times 0.684 \times 10^{-3} \times (2.2 \times 10^{-10})^3} = 6.817 \times 10^{10} \text{ s}^{-1}$$

The dynamic scaling theory proposed that the ratio of the absorption coefficient at the critical concentration and temperature T, $\alpha(x_c, T)$ to the absorption coefficient at the critical concentration and critical temperature $\alpha_c(x_c, T_c)$ should be a function of the reduced frequency ω^* and should scale with the theoretical scaling function $F(\omega^*)$ according to the equation:

$$\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)} = F(\omega^*) = (1 + \omega^{*-0.5})^{-2} \quad (4.4.3)$$

$$\text{Where, } \omega^* = \frac{\omega}{\omega_D} = \frac{2\pi f}{\omega t^{z\gamma}} \quad (4.4.4)$$

The ratio $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ and ω^* are calculated at different frequencies. The results are given in Table (4.8).

Table (4.8): Values of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ at different frequencies for the critical

mixture water - phenol.

f (MHz)	ω^* experimental	$\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$	$\omega^{*'}theoretical$	F($\omega^{*'}$)
9	51.88	0.80	10^{-10}	9.9×10^{-11}
	9.094	0.67	10^{-9}	9.9×10^{-10}
	2.444	0.53	10^{-8}	9.9×10^{-9}
	0.6553	0.43	10^{-7}	9.9×10^{-8}
	0.1749	0.24	10^{-6}	9.9×10^{-7}
	0.0811	0.17	10^{-5}	9.9×10^{-6}
	0.0374	0.08	10^{-4}	9.8×10^{-5}
	0.0186	0.001	10^{-3}	9.8×10^{-4}
15	86.47	0.81	10^{-2}	0.008
	15.15	0.71	10^{-1}	0.05
	4.074	0.53	1	0.25
	1.092	0.42	10	0.57
	0.2915	0.30	10^2	0.82
	0.1351	0.21	10^3	0.93
	0.0625	0.12	10^4	0.98
	0.0310	0.09	10^5	0.99
30	173.28	0.85	10^6	0.99
	30.319	0.70	10^7	0.99
	8.149	0.50	10^8	0.99
	2.184	0.40	10^9	0.99
	0.5832	0.21	10^{10}	0.99
	0.2703	0.16	10^{11}	0.99
	0.1250	0.09	10^{12}	0.99
	0.0621	0.05	10^{13}	0.99
35	202.16	0.85	10^{14}	0.99
	35.37	0.71	10^{15}	0.99
	9.507	0.51	10^{16}	0.99
	2.549	0.42	10^{17}	0.99
	0.6804	0.27	10^{18}	0.99
	0.3154	0.21	10^{19}	0.99
	0.1458	0.099	10^{20}	0.99
	0.0725	0.081	10^{21}	0.99

These experimental data are taken from (Abdelraziq, 2015 to be published)

The experimental data of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ of the critical mixture water - phenol are plotted as a function of the reduced frequency ω^* and fitted to the theoretical curve of $F(\omega^*)$ as shown in Fig.(4.9).

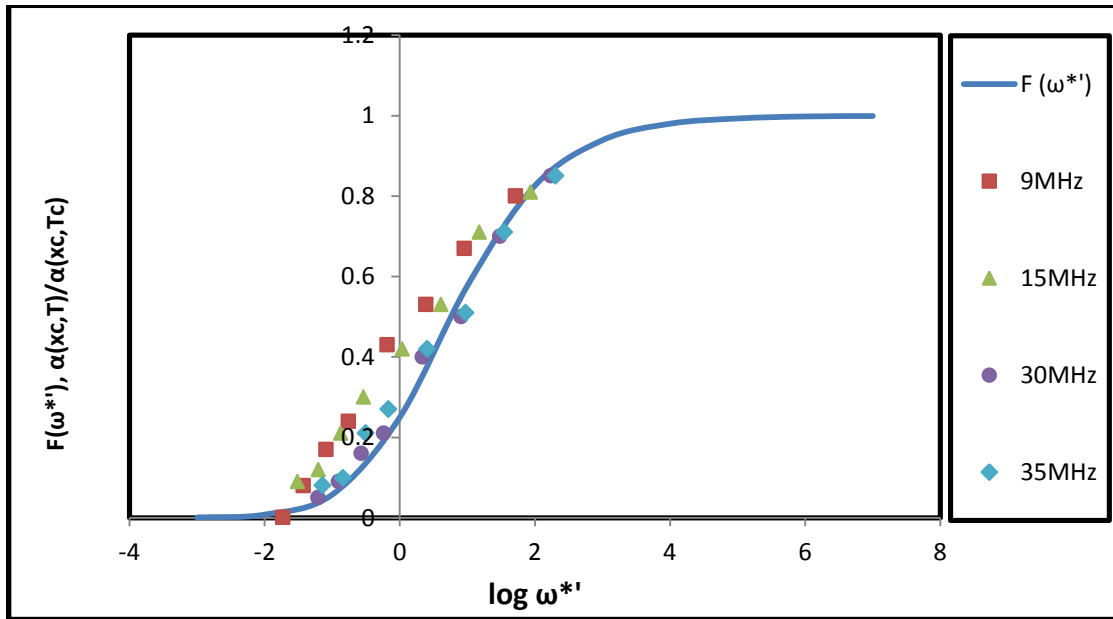


Fig.(4.9): The experimental values of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ as a function of the reduced frequency ω^* along with the theoretical scaling function $F(\omega^*)$.

The experimental data of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ as function of the reduced frequency ω^* are in good agreement with the theoretical scaling function $F(\omega^*)$ as assumed by Ferrell and Bhattacharjee (Ferrell and Bhattacharjee, 1981).

Chapter Five

Conclusion

The anomalous behavior of the dynamic shear viscosity for the binary liquid mixture water – phenol was detected at the critical point. The critical point temperature and concentration were determined to be 67°C and 33.9% by weight of phenol respectively. Our experimental results showed a good agreement with Howell, Krishnan and Campbell experimental results (Howell, 1932; Krishnan, 1935; Campbell Alan N. and Campbell A. Jean R., 1973).

The specific heat at constant pressure at the critical temperature and concentration C_{pc} was calculated using the Two Scale Factor Universality. The critical and the background thermal expansion coefficients α_{pc} , α_{pb} were determined.

The ultrasonic attenuation coefficient for the critical mixture water - phenol were plotted as a function of temperature, at frequencies 9, 15, 30, and 35 MHz. The ultrasonic absorption coefficient for the critical concentration decreases as the temperature increases for all frequencies. The absorption coefficient at the critical temperature and concentration decreases with increasing frequency. The results showed a linear relation of $\frac{\alpha_c}{f^2}$ versus $f^{-1.06}$ as predicted by the dynamic scaling theory.

The critical and background parts of the attenuation coefficient were determined. The experimental data of $\frac{\alpha(x_c, T)}{\alpha_c(x_c, T_c)}$ as function of the reduced frequency ω^* are in good agreement with the theoretical scaling function

$F(\omega^*)$ as assumed by Ferrell and Bhattacharjee (Ferrell and Bhattacharjee, 1981).

Water-phenol mixture can be studied by using the renormalization group theories as a future work to be compared with the mode coupling theories.

The values of different properties which are found in this work and other works are given in Table (5.1).

Table 5.1: The values of different properties which are found in this work and other works.

The measured and calculated value	Our work	Other works
x_c^*	33.9%	34.6% ^(a) 34.5% ^(b) 34.0% ^(c)
T_c^*	67°C	66.4°C ^(a) 66.8°C ^(b) 69.0°C ^(c)
C_{pc}^{**}	$241.9 \frac{J}{Kg K}$	—
ρ_c^*	$0.959 \frac{gm}{cm^3}$	—
α_{pc}^{**}	$2.729 \times 10^{-3} \text{ } ^\circ\text{C}^{-1}$	—
α_{pb}^{**}	$22.59 \times 10^{-3} \text{ } ^\circ\text{C}^{-1}$	—
S^{**}	$0.137 \times 10^{-8} \text{ cm}^{-1} \text{ s}^{0.94}$	—
b^{**}	$0.230 \times 10^{-12} \text{ cm}^{-1} \text{ s}^2$	—

^(a)(Howell, 1932), ^(b)(Campbell Alan N. and Campbell A. Jean R., 1973),

^(c)(Krishnan, 1935), * measured value, ** calculated value.

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جامعة النجاح الوطنية
كلية الدراسات العليا

السلوك الحرج للاضحلال الصوتي للخليط الثنائي الماء والفينول

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قدمت هذه الأطروحة استكمالاً لمتطلبات الحصول على درجة الماجستير في الفيزياء بكلية الدراسات العليا في جامعة النجاح الوطنية في نابلس، فلسطين.

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ب
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المخلص

في هذه الدراسة تم قياس اللزوجة للخليط السائل الثنائي الماء- الفينول باستخدام مقياسي اللزوجة الرقمي والزجاجي الشعري. و تم تحديد شذوذ اللزوجة عند درجة الحرارة الحرجة ($T_c=67.0^\circ\text{C}$) والتركيز الحرج بنسبة (33.90 %) من وزن الفينول. تم حساب الحرارة النوعية ومعامل التمدد الحراري عند ثبات الضغط للخليط الثنائي الحرج. تم تحليل بيانات الاضمحلال فوق الصوتي عند الترددات 9، 15، 30 و 35 ميگاهرتز باستخدام (Dynamic scaling theory). وتم تحديد العلاقة الخطية بين قيم ($f^{-1.06}$ و $\frac{\alpha_c}{f^2}$) كما تنبأت النظرية. وقد تم مقارنة القيم العملية $\frac{\alpha}{\alpha_c}$ مع الاقتران النظري $F(\omega^*)$ ووجد توافق جيد بين القيم العملية والنظرية.