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Critical Behavior of the Ultrasonic Attenuation for the Binary Mixtures of Benzene-Coconut Oil and Hexane-β, β'-dichloroethyl ether

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Summary
Ultrasonic absorption and velocity measurements were made as a function of temperature for the binary mixtures of benzene-coconut oil and hexane-β, β'-dichloroethyl ether. Ultrasonic absorption at 5, 7, 10, 15, 21, and 25 MHz and above T_c is a analyzed by the dynamic scaling theory of Ferrell and Bhattacharjee (FB). The ultrasonic absorption of the two binary mixtures exhibit strong temperature and frequency dependence near T_c. Ultrasonic velocity behaves as a linearly decreasing function of temperature above the critical temperature satisfying linear relation. The observed of α_c/f^2 Vs. $f^{-1.06}$ yields a straight line as predicted by FB theory. Experimental values of α/α_c for the two binary critical mixtures are compared to the scaling function $F(\omega^*)$.

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1. Introduction

A binary mixture of two different fluids exhibits a critical temperature T_c below which the two components do not mix homogeneously in all proportions, and one can define a two-component region bounded by a coexistence curve. In many respects this is analogous to the two-phase region of a simple one-component fluid. If ultrasound propagates through a critical binary liquid at critical concentration (at which exhibits maximum ultrasonic absorption) the intensity I as a function of the propagation distance x is given by:

$$I = I_0 \, \mathrm{e}^{-2\alpha x},$$

where I_0 is the intensity at $x = 0$ and α is the absorption coefficient. There are several mechanisms responsible for the absorption in binary liquid mixtures. The so called classical absorption coefficient is due to viscosity, heat conduction, heat radiation and diffusion near the critical temperature. The excess absorption (the observed absorption minus the classical absorption value) is normally due to various relaxation processes including critical fluctuations. Any process which removes energy from the sound beam and returns it at an appreciably later time in the wave cycle causes a dissipation of acoustic energy. The processes which transfer energy from the translational mode of motion (which is sound wave) to other modes such as vibration or rotation of atoms within the molecules, or the potential energy of some structural rearrangement (chemical reaction and electrolytic processes). The molecule returns to its equilibrium state by a relaxation process, it transfers this energy back to its surrounding exponentially with time. For each relaxation process there is a characteristic relaxation time τ_D which is the time it takes the process to transfer $1/e$-th of the energy.

In this research we have examined sound propagation in the binary mixtures of benzene-coconut oil and hexane-β, β'-dichloroethyl ether (commonly called chlorex) by measuring their ultrasonic absorption. The shear viscosity of the two mixtures as a function of temperature at critical concentration has also been studied in order to determine the correlation length ξ_0 and diffusion coefficient D_0 in equation (5). In addition, the sound velocity of the benzene-coconut oil mixture at different temperatures has been measured. In the literature, there are two papers with ultrasonic measurements of these two systems [1, 2]. The absorption results in these two papers do not cover a wide range of frequencies and temperatures, which are not extensive enough to allow analysis using the dynamic scaling theory by Ferrell and Bhattacharjee (FB) [3]. In order to apply the dynamic scaling theory [3] for the study of these two systems, further absorption coefficients and velocities are measured for an extended frequency range of 5–25 MHz and the temperature range of T_c to 45 °C for benzene-coconut oil and from T_c to 35 °C for hexane-β, β'-dichloroethyl ether. The FB theory indicates that α/α_c (see equation (1) for definitions of α and α_c) should be a function of the reduced frequency ω^*, and should scale with the scaling function $F(\omega^*)$. According to the theory, α_c/f^2 vs $f^{-1.06}$ (f is the frequency in Hz) should give a straight line at the critical temperature and critical concentration of the binary liquid mixtures.

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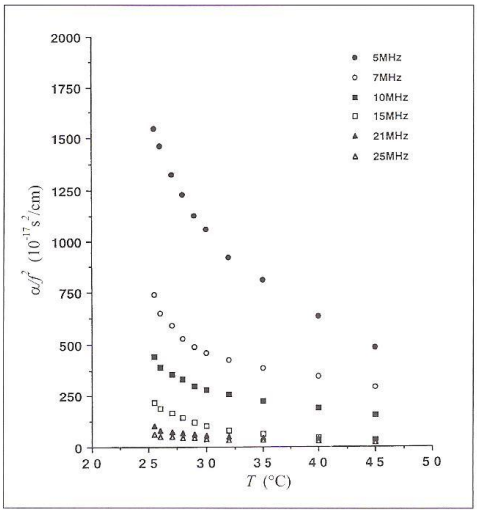

Figure 1. Temperature dependence of absorption for the critical binary mixture of benzene-coconut oil.

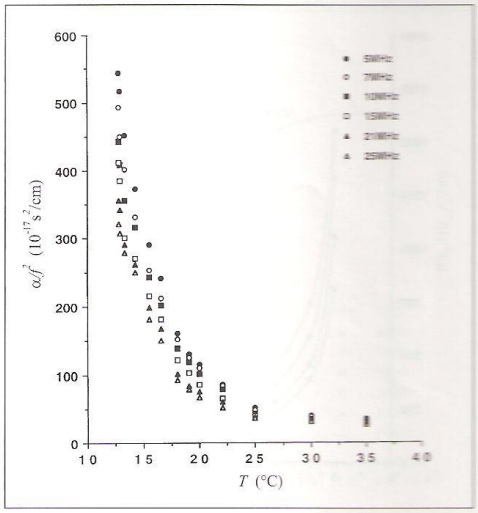

Figure 2. Temperature dependence of absorption for the critical binary mixture of hexane-chlorex.

2. Experimental Technique

The purified liquids benzene, hexane and β,β'-dichloroethyl ether were obtained from Fisher Scientific. The coconut oil was from Fry Scientific and of a pure lab-grade. The chemicals were used without any further purification. The absorption and velocity were made with a Matec pulse-echo system that generates a train of ultrasonic pulses through the temperature-controlled test cell. The setup and operational procedures are discussed in our previous papers [4, 5, 6, 7, 8, 9, 10]. The shear viscosity was measured by using a Brookfield Digital DV-I+ Viscometer. The measurement range of DV-I+ (in centipoise or miliPascal seconds) is determined by the rotational speed of the spindle, the size and shape of the spindle, the container the spindle is rotating in, and the full scale torque of the calibrated spring [10]. To control the temperature of the sample cell, a water jacket is provided with inlet and outlet attached with a thermostatic device consisting of a constant temperature water bath and a water-pump circulating system. The thermostatic control error was $\pm 0.01\,^{\circ}$C. Attention must be paid to maintaining a steady temperature. Difficulties are due mainly to irregularities in peak structure arising from the temperature fluctuations near critical temperature. These were minimized and the diffraction effects were not a problem in our measurements [4, 11].

3. Results and Analysis

The binary mixture benzene-coconut oil has an upper critical temperature T_c of 25.4 °C ±0.1 °C and the critical composition x_c is 60.37% by weight of benzene [1]; for hexane and β,β'-dichloroethyl ether T_c = 12.7 °C and x_c = 46.08% by weight of hexane [2]. In Figures 1 and 2, the temperature dependence of the absorption for the binary mixtures of benzene-coconut oil and hexane-β,β'-dichloroethyl ether, respectively, at 5, 7, 10, 15, 21,and 25 MHz are shown. The error in the absorption measurements was less than 4%. It is observed that α/f^2 decreases with temperature at all frequencies, studied. Figures 3 and 4 indicate the frequency dependence of α/f^2 at various temperatures for the two binary mixtures.

Table I. The experimental ultrasonic critical velocity u_c, intercept c, and the temperature coefficient of velocity A at different frequencies for the benzene-coconut oil and hexane-β,β'-dichloroethyl ether critical mixtures.

f (MHz)	u_c (m/s)	A (m/s °C)	c (m/s)
	Benzene-coconut oil T_c = 25.4 °C		
5	1330.3	-3.38	1416.1
7	1333.4	-3.36	1418.7
10	1336.8	-3.43	1423.9
15	1340.8	-3.48	1429.2
21	1345.3	-3.40	1431.7
25	1347.7	-3.36	1433.0
	hexane-chlorex T_c = 12.7 °C		
5	1161.8	-4.26	1215.9
7	1166.6	-4.32	1221.5
10	1171.7	-4.21	1225.2
15	1179.1	-4.31	1233.8
21	1185.6	-4.30	1240.2
25	1192.0	-4.27	1246.2

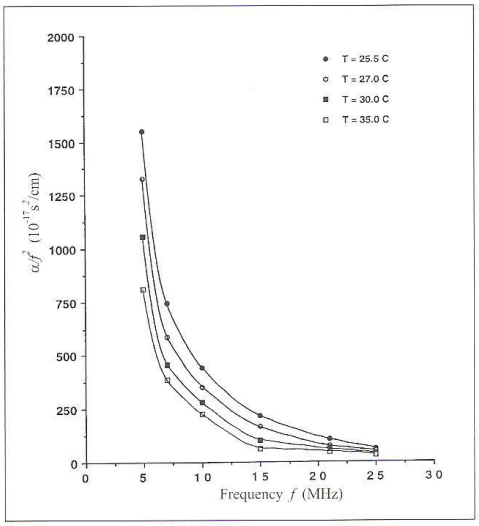

Figure 3. Frequency dependence of absorption at various temperatures for the benzene-coconut oil mixture.

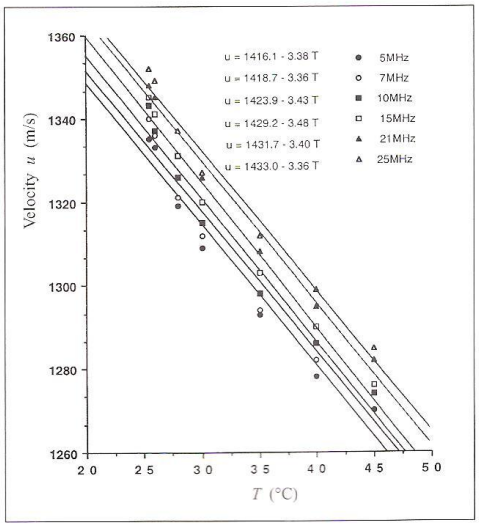

Figure 5. The ultrasonic velocity versus temperature at critical concentration at different frequencies for benzene–coconut oil mixture.

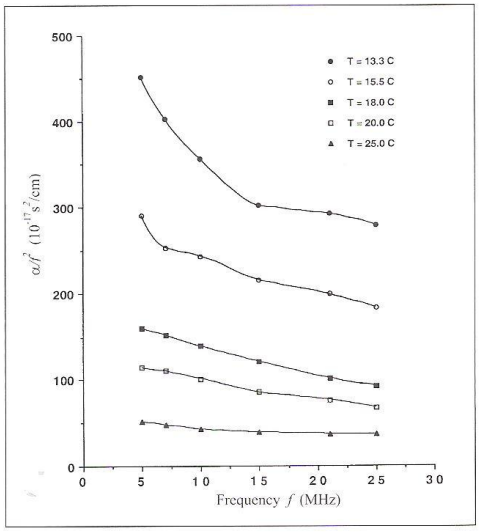

Figure 4. Frequency dependence of absorption at various temperatures for the hexane-chlorex mixture.

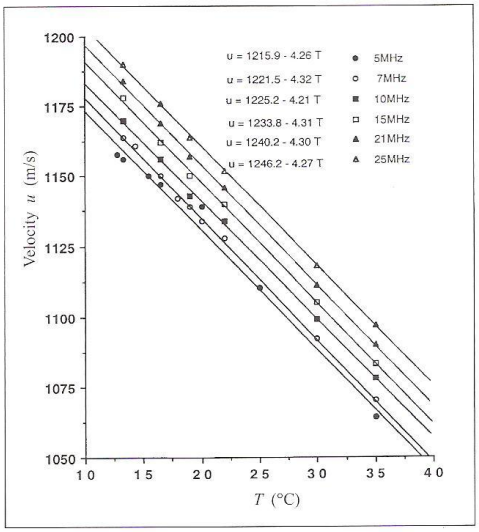

Figure 6. The ultrasonic velocity versus temperature at critical concentration at different frequencies for the hexane-chlorex mixture.

The temperature and frequency dependence of ultrasonic velocity of benzene-coconut oil and hexane-β,β'-dichloroethyl ether mixtures at critical concentration are shown in Figures 5 and 6. The ultrasonic velocity behaves as a linearly decreasing function of temperature above the critical temperature satisfying a linear relation u = c + A T. The c's and A's of linear relations for the two binary critical mixtures at different frequencies are given in Table I. A similar behaviour relating to velocity and temperature was reported by Fenner and Bowen [12], Arrigo and Sette [13], Fast and Yun [14] Garland and Sanchez [15] and also by our previous work [5, 7, 8, 9], in different liquid mixtures.

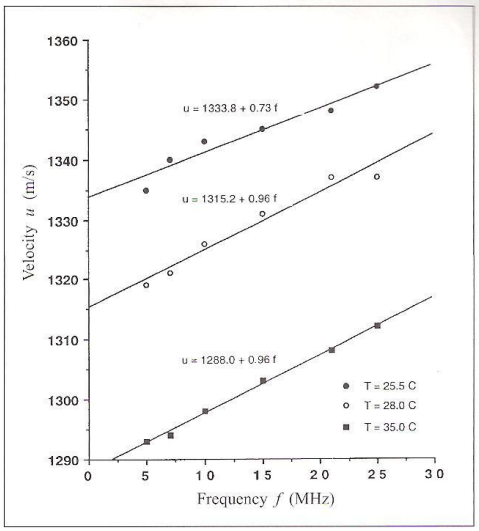

Figure 7. Frequency dependence of velocity for the critical mixture of benzene–coconut oil.

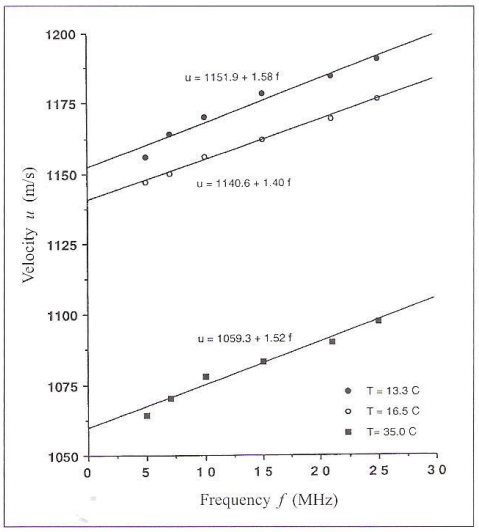

Figure 8. Frequency dependence of velocity for the critical binary mixture of hexane-chlorex.

The critical mixtures of benzene-coconut oil and hexane-β,β'-dichloroethyl ether exhibit a very small dispersion as shown in Figures 7 and 8. This dispersion appears to be unreal and may be due to the large scatter in velocity data measurements. The velocity for both critical mixtures increases with increasing frequency (Figures 7 and 8). The error in the velocity measurements was less than 0.2%.

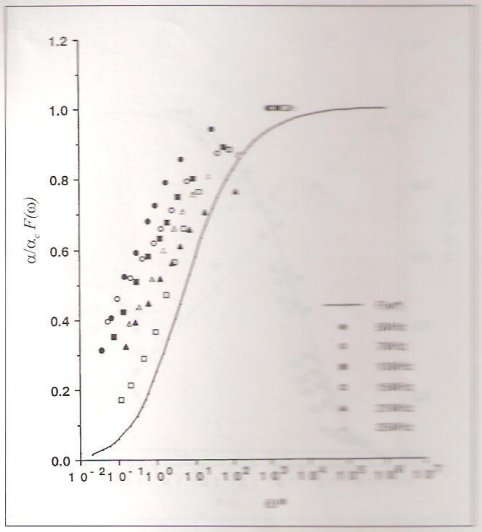

Figure 9. A plot of α/α_c as a function of the reduced frequency ω^* with scaling function $F(\omega^*)$ for benzene-coconut oil mixture.

According to the dynamic scaling theory (FB) [3, 14, 15, 16, 17], the expression for the critical term of the absorption as a function of reduced frequency ω^* is

$$\alpha/\alpha_c = \alpha(x_c,\omega,T)/\alpha(x_c,\omega,T_c) = F(\omega^*), \quad (1)$$

where $\alpha(x_c,\omega,T)$ is the absorption term at critical concentration x_c and at temperature T and $\alpha(x_c,\omega,T_c)$ is the absorption term at critical concentration x_c and at critical temperature T_c and

$$F(\omega^*) = (1+1/\sqrt{\omega^*})^{-2} \quad (2)$$

is the predicted scaling function. The dimensionless reduced frequency ω^* is defined by

$$\omega^* = \omega/\omega_D, \quad (3)$$

where the characteristic temperature-dependent relaxation rate ω_D is given by [15, 18]

$$\omega_D = \omega_0 t^{1.93}. \quad (4)$$

Here $t = (T-T_c)/T_c$ is the reduced temperature and ω_0 is

$$\omega_0 = 2D_0\xi_0^{-2} = (k_B T_c/3\pi\eta_0\xi_0^3), \quad (5)$$

where D_0 is the diffusion coefficient, ξ_0 is the correlation length, kB k_B designates Boltzmann's constant and η_0 is the shear viscosity coefficient.

In Figures 9 and 10, the experimental values of α/α_c vs ω^* are shown along with the theoretical scaling function $F(\omega^*)$. The data of benzene-coconut oil critical mixture (Figure 9) were fitted to the theoretical curve using a value

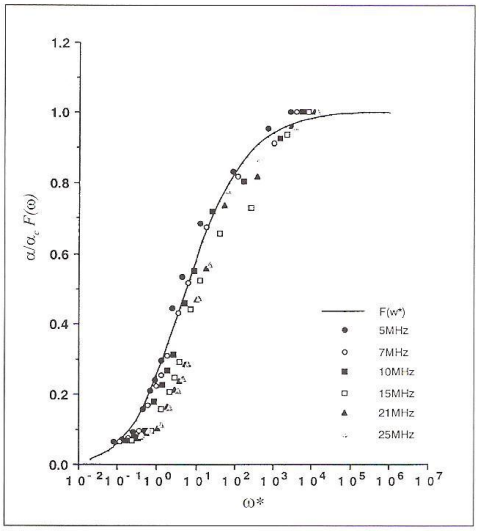

Figure 10. A plot of α/α_c as a function of the reduced frequency ω^* with scaling function $F(\omega^*)$ for hexane-chlorex mixture.

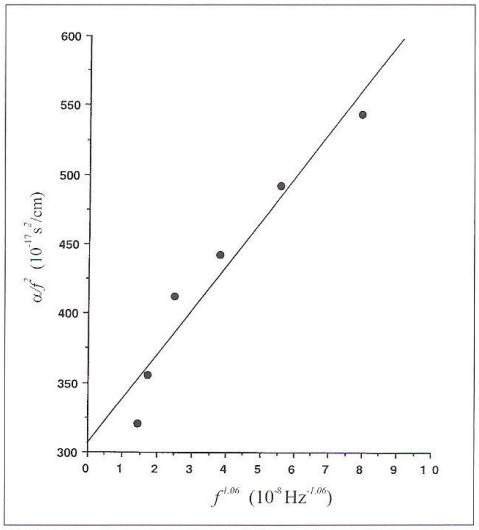

Figure 12. α_c/f^2 versus $f^{-1.06}$ at T_c and critical mixture of hexane–chlorex for this work.

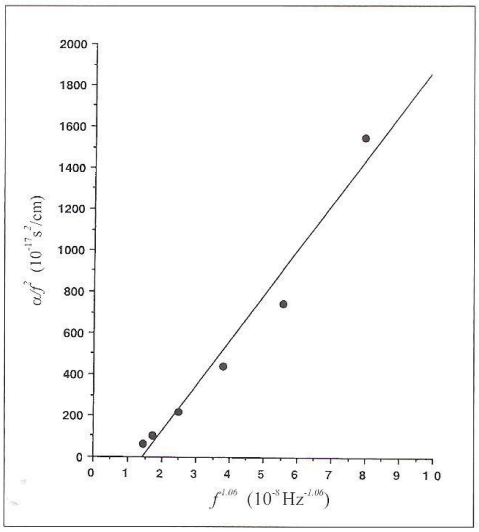

Figure 11. α_c/f^2 versus $f^{-1.06}$ at T_c and critical mixture of benzene–coconut oil for this work.

of $\omega_0 = 14.90 \cdot 10^{10} \pm 0.21 \cdot 10^{10}$ Hz (a least-square fit of the computed data ω_D at various temperatures given by reference [1] yields the value of ω_0]. The data of hexane-β,β'-dichloroethyl ether, (Figure 10) were fit using a value of $\omega_0 = 5.42 \cdot 10^{10} \pm 0.32 \cdot 10^{10}$ Hz [19].

According to dynamic scaling theory (FB) [3, 15]

$$\alpha_c/f^2 = Sf^{-1.06} + b, \tag{6}$$

where α_c/f^2 is the absorption coefficient at critical concentration and critical temperature T_c, the quantity $b = \alpha(\text{background})/f^2$ is the frequency-independent value of Navier-Stokes term [20], and S is a constant independent of frequency. Figures 11 and 12 show plots of α_c/f^2 vs $f^{-1.06}$ for benzene-coconut oil and hexane-β,β'-dichloroethyl ether critical mixtures, respectively. A least-square fit of Figure 11 yields a slope of $220 \cdot 10^{-9}\,\text{cm}^{-1}\text{s}^{0.94}$ and intercept (background term) of $-322 \cdot 10^{-17}\,\text{cm}^{-1}\text{s}^2$. The velocity and attenuation of ultrasonic waves have been measured in complex binary mixture of benzene-coconut oil by Bhattacharya and Deo [1] near the critical temperature in the low MHz region. The ultrasonic absorption data is given for the frequencies 1, 2, 3, and 5 MHz. Their experimental results were analysed in terms of theories developed by Kawasaki [21] and Mistura [22]. Bhattacharya and Deo's data were plotted in Figure 13, this (according to FB theory) yields a slope of $295 \cdot 10^{-9}\,\text{cm}^{-1}\text{s}^{0.94}$ and intercept of $-733 \cdot 10^{-17}\,\text{cm}^{-1}\text{s}^2$. For hexane-$\beta,\beta'$-dichloroethyl ether critical mixture (Figure 12), the slope is $32 \cdot 10^{-9}\,\text{cm}^{-1}\text{s}^{0.94}$ and the intercept (background term) is $306 \cdot 10^{-17}\,\text{cm}^{-1}\text{s}^2$. Our values (absorption, slope, and intercept) show good agreement with Bhattacharya and Deo's data. For example of benzene-coconut oil, at frequency 5 MHz and $T = 26.0\,^\circ\text{C}$ his measurement of absorption is $1480 \cdot 10^{-17}\,\text{cm}^{-1}\text{s}^2$, and our measurement at the same frequency and temperature is $1472 \cdot 10^{-17}\,\text{cm}^{-1}\text{s}^2$. Bains and Breazeale [2] have measured the ultrasonic absorption of hexane-β,β'-dichloroethyl ether critical mixture at 1, 3, 5, 7, and 9 MHz. Their work was analysed using the Fixman's theory. According to FB theory their data (Figure 14) yield a slope of $61 \cdot 10^{-9}\,\text{cm}^{-1}\text{s}^{0.94}$ and intercept of

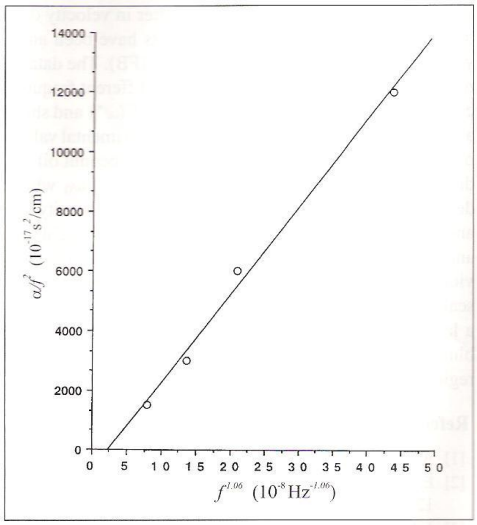

Figure 13. α_c/f^2 versus $f^{-1.06}$ at T_c and critical mixture of benzene–coconut oil for Bhattacharya and Deo's data.

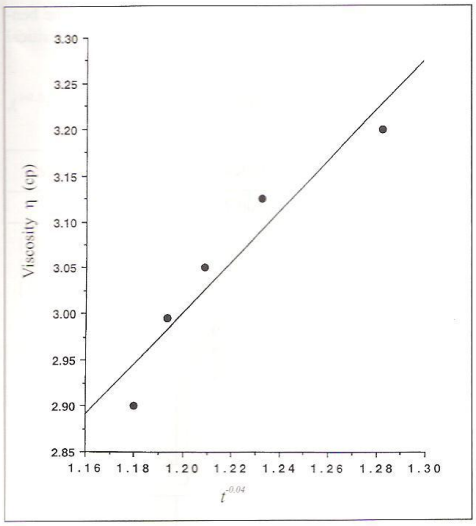

Figure 15. The measured values of the shear viscosity η versus $t^{-0.04}$ for the critical mixture of benzene–coconut oil.

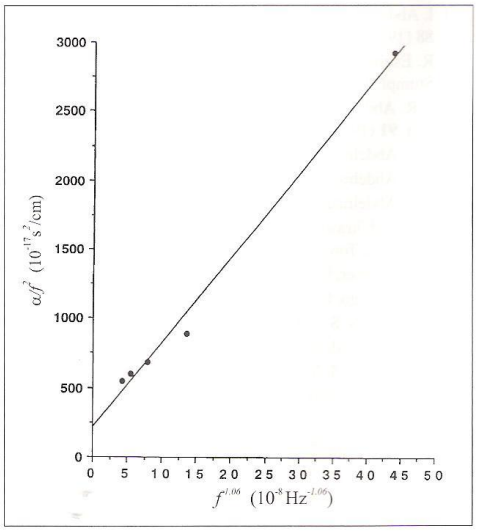

Figure 14. α_c/f^2 versus $f^{-1.06}$ at T_c and critical mixture of benzene–coconut oil for Bains and Brezeale's data.

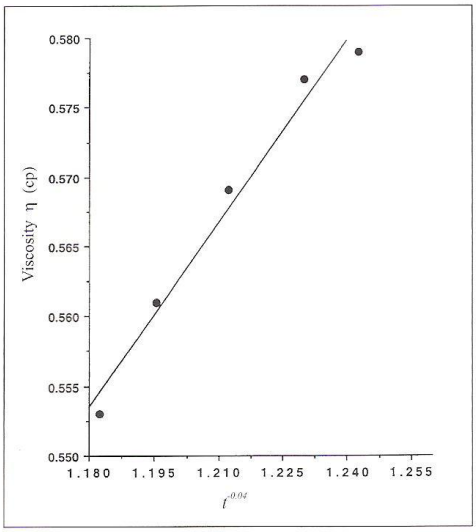

Figure 16. The measured values of the shear viscosity η versus $t^{-0.04}$ for the critical mixture of hexane–chlorex.

$210 \cdot 10^{-17}$ cm^{-1}s^2. In general, the plots of our measurements of the two binary mixtures for the extended frequencies 5, 7, 10, 15, 21, and 25 MHz yield straight lines as predicted by FB theory.

The correlation length ξ_0 and diffusion coefficient D_0 were calculated using the value of ω_0 and the measured shear viscosity coefficient η_0. The measured data of the viscosity for benzene-coconut oil were fit to the power law [15] $\eta = \eta_0 t^{-0.04}$. A least-square fit of Figure 15 yields a value of $\eta_0 = 2.76 \pm 0.02$ cp. The values of, ξ_0 and D_0 are 1.02 Å and $0.84 \cdot 10^{-5}$ cm^2/s, respectively. Figure 16 gives the shear viscosity $\eta_0 = 0.44 \pm 0.01$ cp for hexane-β,β'-dichloroethyl ether. Using the measured η_0, the correlation length ξ_0 and diffusion coefficient D_0 are calculated to be 2.6 Å and $1.8 \cdot 10^{-5}$ cm^2/s. The error in the viscosity measurements was less than 0.5%. Some measured and calcu-

Table II. Some measured and calculated quantities for the benzene-coconut oil and hexane-β,β'-dichloroethyl ether critical mixtures.
T (K), x_c, α_c/f^2 (10^{-17} s^2/cm), η_0 (cp), ω_0 ($\cdot 10^{10}$ Hz), ξ_0 (Å), D_0 (10^{-5} cm^2/s), u_0 (m/s) 5 MHz, S (10^{-9} cm^{-1}s$^{0.94}$), c_{pc} (10^6 erg/cm^{3}K).

	Measured	Calculated	from reference	
	Benzene-coconut oil $T_c = 25.4\,^\circ$C			
T			298.56	[1]
x_c			60.37% Benz	[1]
α_c/f^2	1550		1560	[1]
η_0	2.76			
ω_0		14.9		
ξ_0		1.02		
D_0		0.84		
u_0	1330.3		1334	[1]
S	220		295	
c_{pc}		2.42		
	hexane-chlorex $T_c = 12.7\,^\circ$C			
T			285.86	[2]
x_c	46.08% Hex.			
α_c/f^2	543		546	[2]
η_0	0.44			
ω_0		4.52		
ξ_0		2.6		
D_0		1.8		
u_0	1161.8		1160	[2]
S	32		61	[2]
c_{pc}		0.155		

lated quantities are given in Table II. Some measured and calculated values are compared with some values in the literatures and show a good agreement. In addition, determining the quantities (in Table II) using the binary mixtures for the first time support the dynamic scaling theory of Ferrel and Bhattacharjee (FB).

4. Conclusion

The ultrasonic absorption of the two binary mixtures of benzene-coconut oil and hexane-β,β'-dichloroethyl ether exhibit strong temperature and frequency dependence near T_c. The ultrasonic velocity behaves as a linearly decreasing function of temperature above the critical temperature satisfying linear relations. The critical mixtures of benzene-coconut oil and hexane-β,β'-dichloroethyl ether exhibit a very small dispersion. This dispersion appears to be unreal and may be due to the large scatter in velocity data measurements. The experimental results have been analysed in terms of dynamic scaling theory (FB). The data of α/α_c for the critical binary mixtures at different frequencies are compared to the scaling function $F(\omega^*)$ and show a good agreement with the theory. The experimental values of α/α_c for some frequencies of benzene-coconut oil are displaced relative to the FB theory perhaps of ω_0 which depends on the correlation length ξ_0, shear viscosity η_0, and T_c. The values of α_c/f^2 vs $f^{-1.06}$ yield a straight line and show a good agreement with the FB theory. The previous systems that have been evaluated using the dynamic scaling theory (FB) and the present work, would provide a large experimental information physical parameters on binary liquid mixtures and their behaviour in the critical region.

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