Results in Physics 11 (2018) 394–399

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.elsevier.com/locate/rinp

Modification of the Fixmen's, Kawasaki's, Hornowski's, Mistura's, and Chapan's analytic function in binary liquid mixtures

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ARTICLE INFO

Keywords:
Ultrasonic absorption
Critical binary mixture
Mode coupling theory
Analytic function

ABSTRACT

The mode-coupling theory of Fixmen, Kawasaki, Hornowski, Mistura, and Chapan failed to explain the experimental behavior of the binary liquid mixtures at critical composition and above critical temperature. The analytic function of Fixmen's, Kawasaki's, Hornowski's, Mistura's, and Chapan's theories was modified in order to get an agreement with the experimental behavior of the binary liquid mixtures at critical concentration and above the critical temperature.

Introduction

The behavior of sound propagation through absorption or dispersion in binary liquid mixture at critical composition and above critical temperature has been studied by scientists and researchers [1–31], the most important models that predict the behavior of sound waves propagation are the renormalization group theory [32], the dynamic scaling theory [33] and a package of mode-coupling theory by Fixmen [34], Kawasaki and Shiwa [35,36], Mistura [37], and Chapan [38], these theories failed to describe the experimental data over a wide range of the reduced frequency ω^*.The aim of the present study is to modify the mode-coupling theory, to have an agreement between the theoretical and experimental data for the absorption coefficient in binary liquid mixtures, so it is necessary to predict a new suitable formula for the analytic function K(y).

Experimental technique

Measurements of absorption and velocity are made using Matec pulse – echo technique. The following Matec equipment was used: a radio frequency gated amplifier model 515, which is a plug-in unit of Matec gating modulator model 5100, a doubled turned – amplifier model 252, a broadband receiver model 605 and model 666 pulse-comparator, which generates a pulse of radio frequency energy, which can be precisely controlled in amplitude in steps of 0.1 dB up to a total of 61 dB. The viscosity was measured using the Brookfield Digital Viscometer, which rotates and measures the torque necessary to overcome the viscous resistance to the induced movement. The sample was prepared by the weight using a Ohaus 16oo series balance and Sartorius scale which has a precision of 0.05 mg. The purification of chemicals was 99.9%. The absorption and velocity of sound in binary liquid mixture were measured above the critical temperature and shaken several times to insure complete mixing. The test cell was also heated above the critical temperature before the sample was added. The pulse comparator was used to measure the height of the echoes viewed on the oscilloscope. For a given number of interference peaks, the displacement of the reflector, which leads to the wavelength, is measured and used with the frequency to determine the velocity of sound in liquid.

Theoretical considerations

The mode-coupling theories by Fixmen, Mistura, Chapan, Kawasaki and Shiwa lead to the same general expression for the attenuation absorption coefficient per wavelength α_λ at critical concentration [34–38]:

$$\frac{\alpha_\lambda}{u^2(\omega)} = \pi A(T) F(\omega*) \tag{1}$$

where A(T) is the critical amplitude which is different for each theory, $u(\omega)$ is the velocity of sound, and F(ω*) is the scaling function. The general form of the scaling function is given by Eq. (2) [34–38]:

$$F(\omega*) = \int \{ \left[\frac{y^2 dy}{(1+y^2)^2} \right] \left[\frac{\omega* K(y)}{K(y)^2 + \omega*^2} \right] \} \tag{2}$$

where y = qξ, q is the wave number and$\xi = \xi_0 t^{-\nu}$ is the correlation length, while ξ_0 is the critical amplitude of the correlation length, the reduced temperature t is represented by: $t = \frac{T - T_c}{T_c}$, T_c is the critical temperature of the binary mixture, the reduced frequency is

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https://doi.org/10.1016/j.rinp.2018.09.025
Received 13 August 2018; Accepted 10 September 2018
Available online 19 September 2018

$\omega^* = \frac{\omega}{\omega_D} = \frac{2\pi f}{\omega_0 t^{z\gamma}}$ where ω is the angular frequency $\omega = 2\pi f$, $z\gamma = 1.9$ is the critical exponent, and ω_D is a characteristic temperature-dependent relaxation rate which is given by: $\omega_D = \frac{K_B T}{3\pi\mu\xi^3} = \frac{K_B T_C}{3\pi\mu_0\xi_0^3} t^{z\gamma} = \omega_0 t^{z\gamma}$,and $\omega_0 = 2D_0\xi_0^{-2}$, where D_0 is the diffusion coefficient, K_B is the Boltzmann's constant and γ is the critical exponent ($\gamma = 0.64$). K(y) is the analytic function. In Fixman theory the analytic function K(y) is given by [34]:

$$K(y) = y^2[1 + y^2] \tag{3}$$

While Kawasaki's analytic function is given by [35,36]:

$$K(y) = \frac{3}{4}\left[1 + y^2 + (y^3 - \frac{1}{y})tan^{-1}(y)\right][1.055^a + \{0.93 + 0.29\log_{10}(y)\}^a]^{\frac{1}{a}} \tag{4}$$

With fitting exponent a = 13.

The Mistura's and Chapan's analytic function is [37,38].

$$K(y) = \frac{3}{4}\left[1 + y^2 + (y^3 - \frac{1}{y})tan^{-1}(y)\right] \tag{5}$$

The critical amplitudes A(T) of the binary mixture in the mode-coupling theories are given as follows:

Fixman's and Kawasaki's critical amplitudes $A_F(T)$ and $A_K(T)$ are given by [34]:

$$A_F(T) = A_K(T) = [\frac{K_B(\gamma_0 - 1)\nu^2)}{\pi\rho u C_{Pb}\xi_0^3}]t^{-\alpha} \tag{6}$$

where K_B is Boltzman's constant, ρ is the density of the mixture, C_{Pb} is background heat capacity at constant pressure, $\gamma_0 = \frac{C_P}{C_V}$ is the ratio of heat capacities, ξ_0 is the correlation length. The form of Mistura's critical amplitude is given by [37]:

$$A_M(T) = A_F(T)[\gamma_0 - 1]^{-2}[1 - 0.5\eta]^2 \tag{7}$$

Chaban's form $A_C(T)$ is given by [38]:

$$A_C(T) = A_F(T)\left[1 - \left(\frac{\rho C_{Pb}}{\alpha_{Pb}}\right)\left(\frac{dT_c}{dP}\right)\right]^2 \tag{8}$$

$\alpha_{Pb}$is the background amplitude of the thermal expansion, $\frac{dT_c}{dP}$ is the slope line of the critical line of consolute points as a function of pressure, and η, ν, α are critical exponents which equal to 0.04, 0.62, 0.11, respectively [39].

Hornowski and Labowski [40] applied the modified version of the mode coupling theory for the acoustic anomaly based on the critical behavior of a relaxing complex heat capacity. The general expression for the critical amplitude $A_H(T)$ is given by:

$$A_H(T) = A_K(T)[1 - 0.5\eta]^2 f(d) \tag{9}$$

where η is a critical exponent = 0.04, d is a dimensionless parameter $d = \left[\frac{\rho C_{Vb}}{T\alpha_{Pb}}\right]\left[\frac{dT_c}{dP}\right]$ and the function $f(d) = |(\gamma_0 d - 1)^2 - 2d\gamma_0[d - (\gamma_0 - 1)^{-1}]|$, C_{Vb} is background heat capacity coefficient and α_{Pb} is background thermal expansion coefficient.

Results and analysis

The absorption attenuation coefficient α_λ at different temperatures and at 5, 7, 10, 15, 21 and 25 MHz frequencies for the critical binary mixture of Nitroethane and Isooctane were measured using the Matec pulse-echo technique that generates pulses through the temperature-controlled test cell. The critical temperature of the binary mixture Nitroethane and Isooctane is Tc = 31.5 °C at critical concentration of 0.50356 by weight of isooctane.

From Eq. (1) we can expect that the experimental values of F(ω*) depend on the amplitude A(T) and other same constants for specific binary mixture, from comparison between Eqs. (10), (11), and (12) we conclude that the experimental values for Fixmen and Kawasaki are the same so the modification will be the same for the two models.

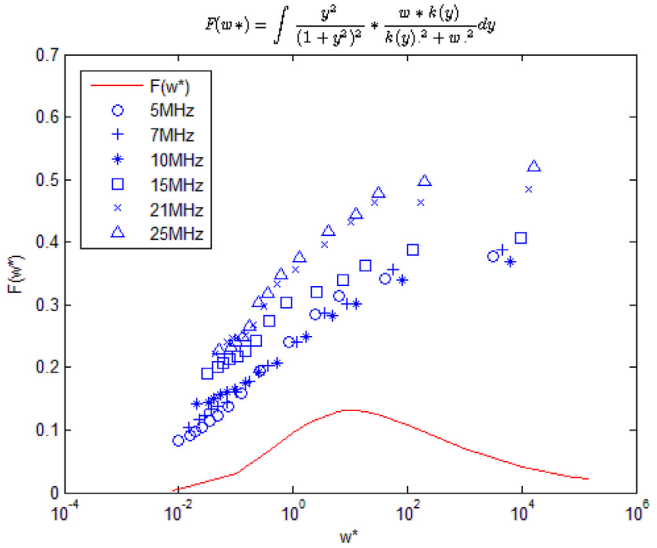

Fig. 1. The experimental absorption values of $\frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to Fixmen theory along with the theoretical scaling integral F(ω*).

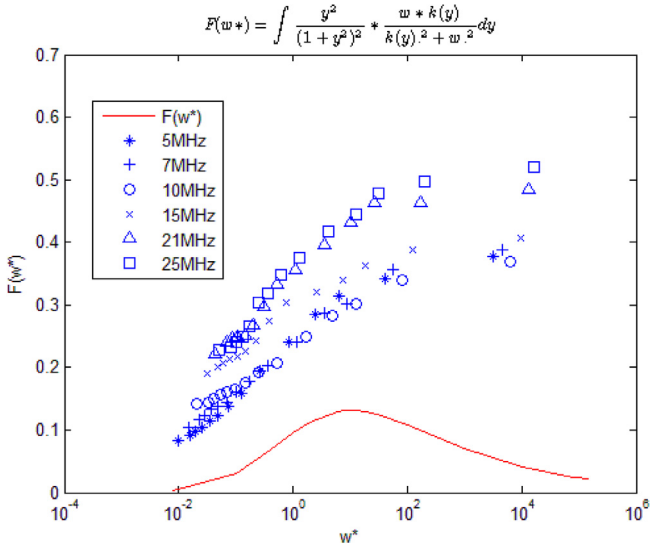

Fig. 2. The experimental absorption values of $F(\omega*) = \frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to Kawasaki and Shiwa theory along with the theoretical scaling integral F(ω*).

Figs. 1–5 represent plots of experimental absorption attenuation values of F(ω*) versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies of Fixmen's, Kawasaki's, Hornowskie's, Mistura's, and Chapan's expressions respectively, along with the theoretical scaling integral F(ω*).

The provided Figures illustrate that there is a disagreement between the theoretical and experimental data at different frequencies. Because of this disagreement, we focus our efforts in this study to modify the analytic function for the mode-coupling theory in order to have an agreement between theory and the experimental data for F(ω*) at different frequencies and temperatures for the five mode coupling theories. The analytic function K(y) affects in the form of the scaling function F(ω*) as shown in Eq. (2). The final modified form of analytic function $k_m(y, \omega^*)$ is given by the relation:

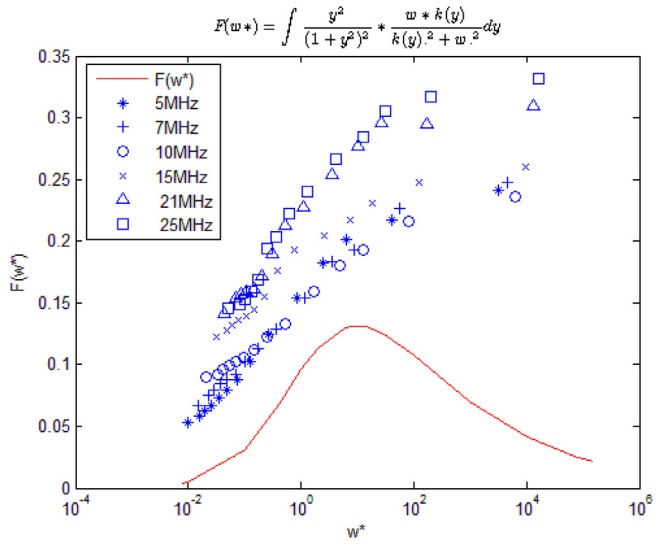

Fig. 3. The experimental absorption values of $F(\omega*) = \frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to Hornowski and Labowski theory along with the theoretical scaling integral F(ω*).

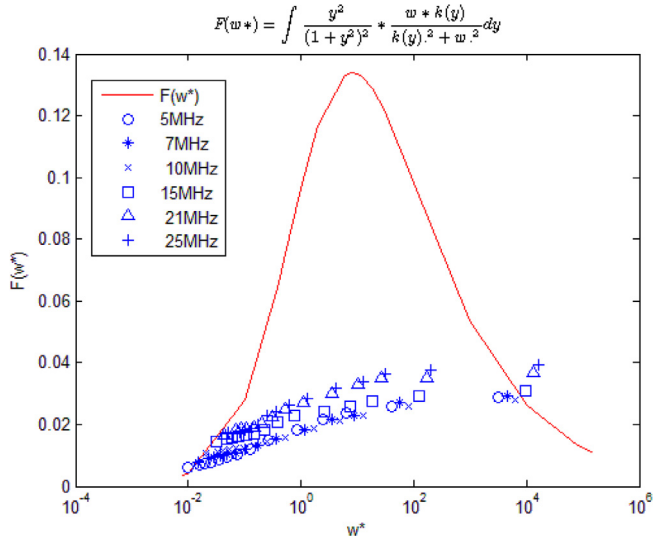

Fig. 4. The experimental absorption values of $F(\omega*) = \frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to Mistura theory along with the theoretical scaling integral F(ω*).

$$k_m(y,\omega^*) = \frac{\omega^* + \omega^* \times \sqrt{1-4\left[\left(\frac{y^2+y}{1+y^2}\right)\times[-0.1762\times\omega^{*-0.225}+0.5428]\right]^2}}{2\left[\left[\frac{y^2+y}{1+y^2}\right]\times[-0.1762\times\omega^{*-0.225}+0.5428]\right]} \quad (10)$$

Fixmen, Kawasaki, Hornowski, Mistura, and Chapan modification analytic function is applied to the binary mixture Nitroethane-Isooctane. The modified analytic function depends on the correlation length ($y = q\xi$) and the reduced frequency $\omega^* = \frac{2\pi f}{\frac{k_B T}{3\pi\mu_0\xi_0^3}t^{z\nu}}$ which depends on the correlation length (ξ) and depends also on the shear viscosity μ.

Figures from 6 to 10 predict plots of Fixmen, Kawasaki, Hornowski, Mistura and Chapan experimental absorption attenuation values of F (ω*) versus the reduced frequency ω* for Nitroethane and Isooctane

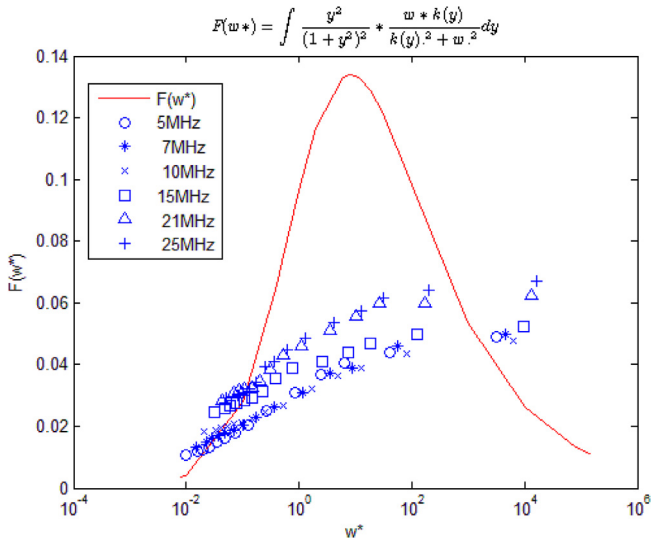

Fig. 5. The experimental absorption values of $F(\omega*) = \frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to Chapan theory along with the theoretical scaling integral F(ω*).

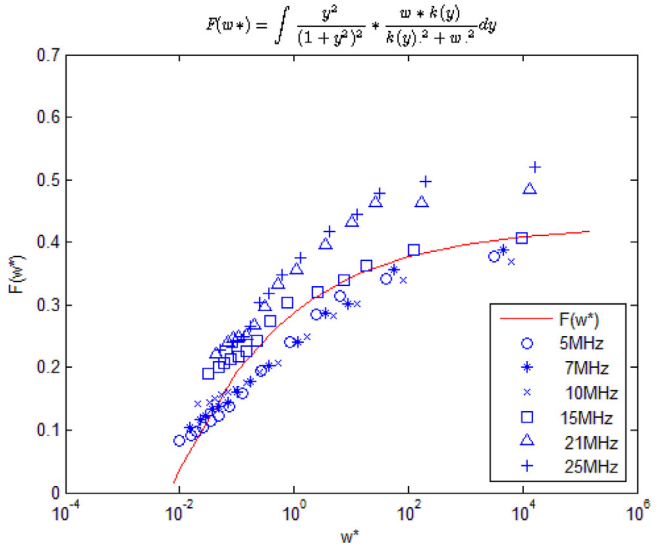

Fig. 6. Kawasaki's experimental data of the absorption attenuation coefficient per wavelength α_λ at critical concentration at different frequencies 5, 7, 10, 15, 21 and 25 MHz for the binary mixture Nitroethane-Isooctane versus the reduced frequency ω^* frequencies according to the general modification for the analytic function K(y, ω*) [Eq. (10)].

binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies. The general modification for the analytic function is given in Eq. (10) along with the theoretical scaling integral F(ω*).

Figs. 6 and 7 show an excellent agreement for large reduced frequency ω^* between experimental values and theoretical predictions for the general modified analytic function K(y, ω*), while Figs. 8, 9, and 10 represent an agreement between theoretical and experimental value of F(ω*) in its general shape but different in the amplitude of F(ω*). So we made another correction to have more suitable diagram. The new correction of K(y, ω*) according to Hornowski theory $k_{mH}(y,\omega^*)$ is given in Eq. (11):

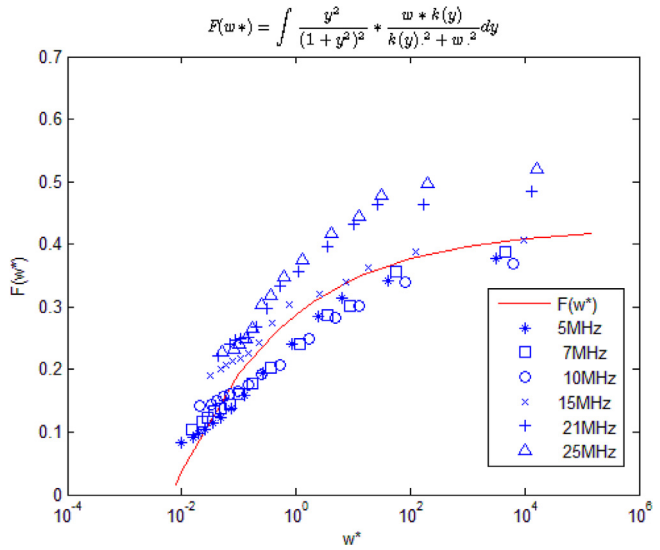

Fig. 7. Fixmen's experimental data of the absorption attenuation coefficient per wavelength α_λ at critical concentration at different frequencies 5, 7, 10, 15, 21 and 25 MHz for the binary mixture Nitroethane-Isooctane versus the reduced frequency ω^* according to the general modification for the analytic function K (y, ω*) [Eq. (10)].

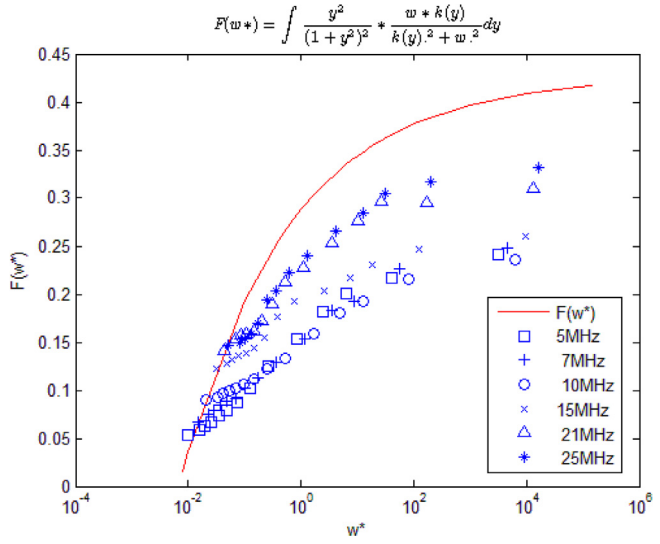

Fig. 8. Hornowski's experimental data of absorption values of F $(\omega^*) = \frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the general modification for the analytic function K(y, ω*) [Eq. (10)].

$$k_{mH}(y, \omega^*) = \frac{\omega^* + \omega^* \times \sqrt{4 - 4\left[\left(\frac{y^2+y}{1+y^2}\right) \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]^2}}{2\left[\left[\frac{y^2+y}{1+y^2}\right] \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]} \quad (11)$$

Fig. 11 represents plot of Hornowski experimental absorption attenuation values of F(ω*) versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies, the new general modification for the analytic function is given in Eq. (11) along with the theoretical scaling integral F(ω*).

For Mistura theory, the new correction for the modified analytic function $k_{mM}(y, \omega^*)$is given by Eq. (12):

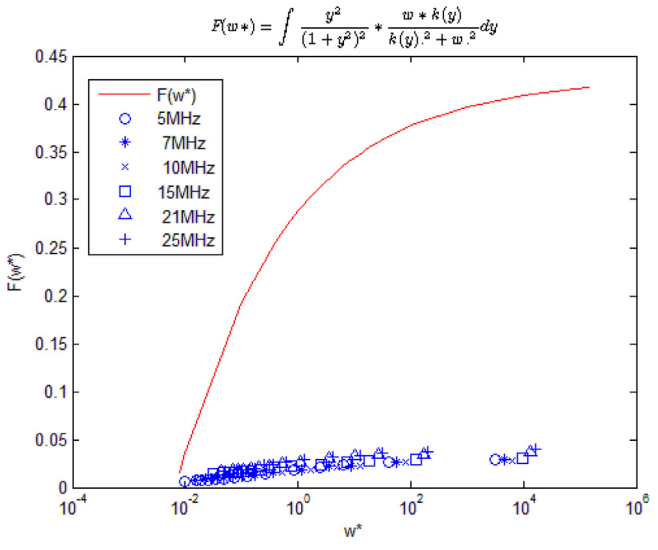

Fig. 9. Mistura's experimental data of absorption values of F(ω*) = $\frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the general modification for the analytic function K(y, ω*) [Eq. (10)].

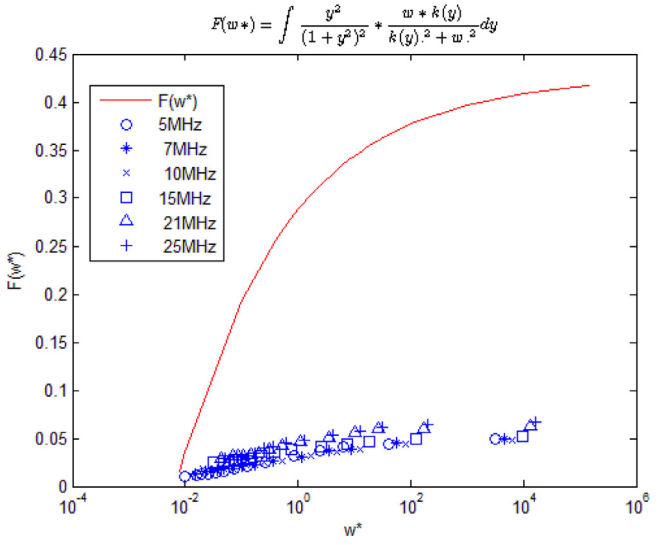

Fig. 10. Chapan's experimental data of absorption values of F(ω*) = $\frac{\alpha_\lambda}{\pi u^2(\omega)A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the general modification for the analytic function K(y, ω*) [Eq. (10)].

$$k_{mM}(y, \omega^*) = \frac{\omega^* + \omega^* \times \sqrt{559 - 4\left[\left(\frac{y^2+y}{1+y^2}\right) \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]^2}}{2\left[\left[\frac{y^2+y}{1+y^2}\right] \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]} \quad (12)$$

Fig. 12 predicts plot of Mistura experimental absorption attenuation values of F(ω*) versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies, the new general modification for the analytic function $k_{mM}(y, \omega^*)$is given in Eq. (12) along with the theoretical scaling integral F(ω*).

Finally, for Chapan theory the new correction for the modified analytic function $k_{mC}(y, \omega^*)$is given by Eq. (13):

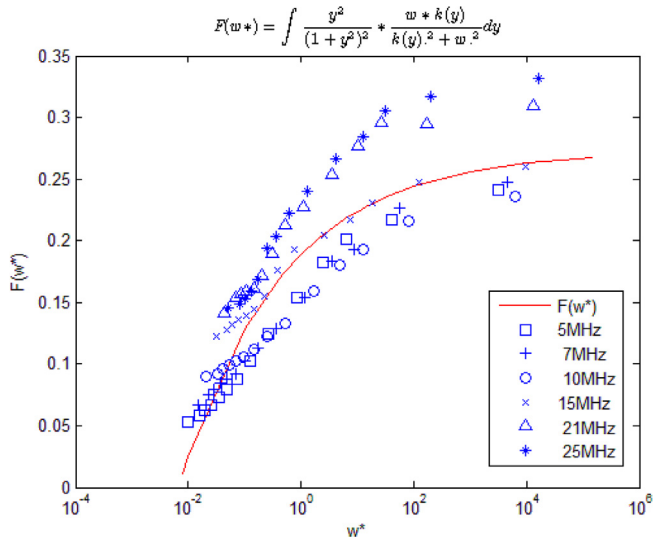

Fig. 11. The experimental data of absorption values of $F(\omega^*) = \frac{\alpha_\lambda}{\pi u^2(\omega) A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the new modified Hornowski model of $k_{mH}(y, \omega^*)$.

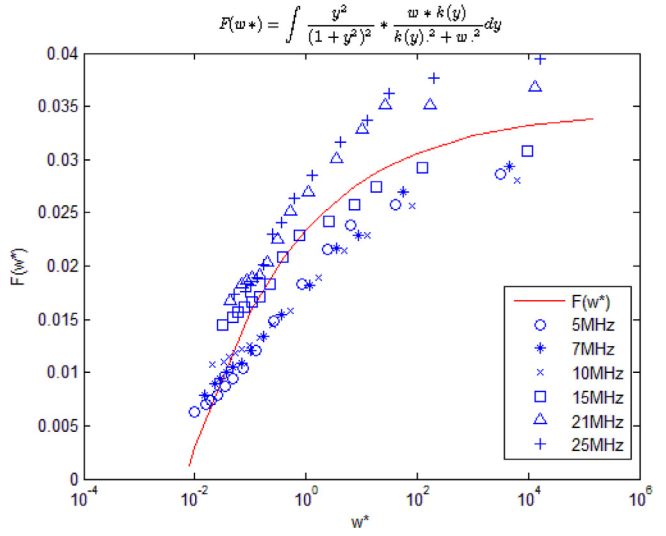

Fig. 12. The experimental data of absorption values of $F(\omega^*) = \frac{\alpha_\lambda}{\pi u^2(\omega) A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the new modified Mistura model of $k_{mM}(y, \omega^*)$.

$$k_{mC}(y, \omega^*) = \frac{\omega^* + \omega^* \times \sqrt{197 - 4\left[\left(\frac{y^2+y}{1+y^2}\right) \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]^2}}{2\left[\left[\frac{y^2+y}{1+y^2}\right] \times [-0.1762 \times \omega^{*-0.225} + 0.5428]\right]} \quad (13)$$

Fig. 13 predicts plot of Chapan experimental absorption attenuation values of F(ω*) versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies, the new general modification for the analytic function $k_{mC}(y, \omega^*)$is given in Eq. (13) along with the theoretical scaling integral F(ω*).

At the end of this section we conclude from the results we have in all previous five theories of mode-coupling theories that we can unite trend for modification of these theories. We modify the analytic function K(y) to K(y, ω*), the experimental data for the five theories have the same

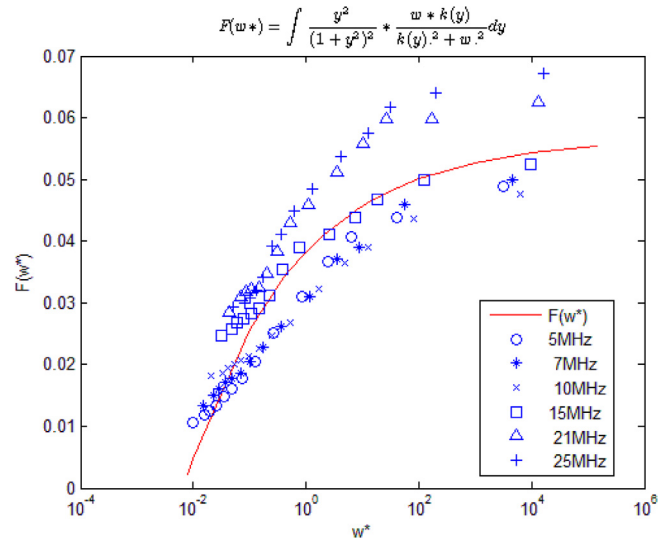

Fig. 13. The experimental data of absorption values of $F(\omega^*) = \frac{\alpha_\lambda}{\pi u^2(\omega) A(T)}$ versus the reduced frequency ω* for Nitroethane and Isooctane binary mixture at 5, 7, 10, 15, 21 and 25 MHz frequencies according to the new modified Chapan model of $k_{mC}(y, \omega^*)$.

shape but differ in the vertical amplitude because the experimental data for F(ω*) depends on the amplitude A(t) as shown in Eq. (1), and the amplitude A(T) for Fixmen theory equals with Kawasaki theory and also, the amplitude for Mistura, Chapan and Hornowski theories depends on Fixmen and Kawasaki theories as shown in Eqs. (6), (7), and Eq. (8) in addition to other constant parameters for the same binary liquid mixture, so the result we have for the modification analytic function for Mistura, Hornowski and Chapan have the same general formula but it must differ in vertical amplitude.

Conclusion

Mode-coupling theories was used to describe the critical behavior of binary liquid mixtures and above critical point. The analytic function of Fixmen, Kawasaki, Mistura, Hornowski and Chapan theories are not suitable to yield proper scaling function in order to get an agreement with experimental data. The analytic function of Mode-coupling theories K(y) relates to the correlation length y = qξ. The disagreement between theoretical and experimental data for Fixmen, Kawasaki, Mistura, Hornowski and Chapan theories for sound propagation in binary liquid mixture is provided in Figs. 1–5. The modified Kawasaki's analytic function depends on the correlation length ξ and on the shear viscosity μ. By Introducing these physical parameters, Fixmen, Kawasaki, Mistura, Hornowski and Chapan theories is improved to describe the critical behavior of binary liquid mixtures above critical temperature and at critical composition, in order to explain the trend of the experimental data, the modification of the analytic function predicts an excellent agreement between theoretical and experimental data for sound propagation in binary liquid mixture.

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