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Two Binary Liquid Critical Mixtures Belong to Class of Universality

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Abstract

The dynamic shear viscosity of a binary liquid mixture phenol–water has been measured at different temperatures (50.0°C ≤ T ≤ 75.0°C) and different concentrations (0.00% up to 100.00% by weight of phenol). The critical temperature T_c and critical concentration x_c are found to be 67.0°C and 33.90% by weight of phenol respectively, the critical mass density ρ_c is measured to be 0.8952 g/cm^3. The critical and background amplitudes of specific heat at constant pressure are calculated to be 78.12 J/kg.K and 85.29 J/kg.K respectively. The pressure derivative of the critical temperature along the critical line T_c' is calculated to be 9.722 ×10^{-6} K/Pa.

In addition, dynamic shear viscosity of binary liquid mixture phenol–cyclohexane has been measured at different temperatures (14.0°C ≤ T ≤ 30.0°C) and different concentrations (2.00% up to 39.70% by weight of phenol). The critical temperature T_c and critical concentration x_c are found to be 17.0°C and 2.70% by weight of phenol respectively; the critical mass density ρ_c is measured to be 0.7627 g/cm^3. The critical and background amplitudes of isobaric thermal expansion coefficient α_{pc} and α_{pb} are calculated to be 8×10^{-6} K^{-1}, 6×10^{-4} K^{-1} respectively. The pressure derivative of the critical temperature T_c' for the binary is calculated to be 2.8572 × 10^{-8} K/Pa. The universal quantity R^+_ξ for the binary liquid critical mixture phenol–water is calculated to be 0.2716 ± 0.0005. In addition, the universal quantity R^+_ξ for the binary liquid critical mixture phenol–cyclohexane is calculated to be 0.2699 ± 0.0001. The calculated values of the universal quantity R^+_ξ are in a good agreement with the theoretical value of R^+_ζ which is equal 0.2710. The two binary liquid critical mixture belong to the class of universality "Two–Scale–Factor Universality".

Keywords: Binary liquid; Critical mixture; Homogeneous

Introduction

Binary liquid mixtures and critical point

Mixtures are the product of a mechanical blending or mixing of chemical substances like elements and compounds, without chemical bonding or other chemical changes, so that each ingredient substance retains its own chemical properties [1]. Mixtures can be either homogeneous or heterogeneous. A homogeneous mixture is a type of mixture in which the composition is uniform and every part of the solution has the same properties. A heterogeneous mixture is a type of mixture in which the components can be seen, as there are two or more phases present.

Binary liquid mixtures are combination of two pure liquid substances, which have a limited solubility of each one in the other [2]. Critical point is the point at which phase transition occurs at certain temperature called critical temperature and concentration [3]. The critical point represents the boundary between regions of homogeneous and heterogeneous behavior in phase diagrams for mixtures [4]. Hypothesis of universality greatly reduces the variety of different types of critical behavior by classifying all systems into a small number of equivalence classes [5].

The phenomenological theory of scaling has been extremely useful in understanding critical phenomena in model systems and in real materials [5]. The first characteristic of a universality class is that all the systems have the same critical exponents. In addition, the equation of state, the correlation functions and other quantities become identical near criticality, provided one matches the scales of the order parameter, the ordering field, the correlation length and the correlation time [5].

A property of hyper scaling or hyper universality (Two–Scale–Factor Universality) applies to systems in the universality classes of fluctuation-dominated (i.e., non-mean-field) critical behavior. These ideas were first developed phenomenologically and later confirmed by explicit renormalization group (RG) calculations [5]. The RG theory of critical phenomena has elucidated the mathematical mechanism for scaling and universality, and has provided a number of calculational tools for estimating universal properties [5].

Theory

Viscosity

The viscosity of a fluid is a measure of its resistance to gradual deformation by shear stress or tensile stress [6]. Viscosity is affected by different factors such as temperature, shear rate, catalyst, pressure, molecular weight concentration and storage age [7].

Mode coupling theory and shear viscosity of binary mixtures

The mode coupling theory explains the behavior of the binary mixtures at the critical temperature and concentration. The mode coupling approach of Kawasaki and Perl and Ferrell predicts a critical anomaly of the dynamic shear viscosity coefficient according to the law [8,9].

$$\frac{\eta - \eta 0}{\eta} = \frac{\Delta \eta}{\eta} = A \ln \zeta + A \ln q_D \quad (2.1)$$

Where η_0 the noncritical part of the measured shear viscosity.

A is a constant which was calculated by D'Arrigo and given by A ~ 0.054 [10], and q_D is Debye momentum cutoff. The dynamic shear viscosity is temperature dependent at the critical concentration which

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is given by the power law [11,12].

$$\eta = \eta_0 t^{-x_\eta \nu} \quad (2.2)$$

Where t is the reduced temperature $t = \frac{T - T_c}{T_c}$, x_η and ν are critical exponents where x_η ν = 0.04 [10,11].

Two–scale–factor universality

The Two–Scale–Factor Universality has been used in modern theories to explain the critical phenomena of binary liquid mixtures by predicting R_ζ [13]. The Two–Scale–Factor Universality hypothesis states that the critical amplitudes do not depend on three different scales of parameter (length, temperature, external field) but only on two scales of parameter [14]. Most of the observed quantities depend only on the dimensionalities of the space (d) and of the order parameter (n) [15].

The fluid and binary mixtures transitions belong to the same class of universality d=3, n=1[16]. All binary liquid mixtures with critical mixing points belong to the same universality class. The universality concept offers the possibility to relate critical amplitudes of these systems. The exponents are universal and related by the so-called scaling laws [16].

The amplitudes of the correlation length, thermal expansion and specific heat can be deduced using the universal amplitude combinations [15,17-19]. Correlation length is a measure of the distances over which the spin–spin (or density–density) correlations in the system extend [20]. The correlation length of a binary mixture at critical composition exhibits an anomalous behavior conforming to the following exponential law:

$$\xi = \xi^+_0 t^{-\nu}, T > T_c \quad (2.3)$$

Where ν is a critical exponent which accepted to be 0.630 ± 0.001,¶ ξ^+_0 is the critical amplitude and t is the reduced temperature $t\frac{T - T_c}{T_c}$, = where T_c is the critical temperature [21].

The specific heat at constant pressure c_p in zero field is singular and is given by:

$$c_p = c^+_{pc} t^{-\alpha} + c^+_{pb}, T > T_c \quad (2.4)$$

Where c_{pc} and c_{pb} are the critical and background amplitudes of the specific heat and α = 0.11 is the critical exponent [6, 22, 23, 24].

The asymptotic behavior of the thermal expansion α_p can be represented by power law of the form,

$$\alpha_p = \alpha^+_{pc} t^{-\alpha} + \alpha^+_{pb}, T > T_c \quad (2.5)$$

Where α_{pc} and α_{pb} are the critical and the background amplitudes of the thermal expansion [16]. With these three amplitudes $\xi^+_{0,}$ $c^+_{pc,}$ and $a^+_{pc,}$ it is possible to construct a quantity, denoted R^+_ζ, which is universal in the same sense as critical indices are universal. This quantity is defined as:

$$R^+_\xi = \xi_0 \left(\frac{á\, c_{pc} \rho_c}{K_B}\right)^{1/d} = \xi_0 \left(\frac{á\, T_c \alpha_{pc}}{T_c' K_B}\right)^{1/d} \quad (2.6)$$

Where d=3 is the dimension of the space, K_B is Boltzman's constant, ρ_c is the density of the critical mixture at critical temperature T_c and concentration $T_c' = \frac{dT_c}{dp}$ is the pressure derivative of the critical temperature along the critical line [16]. The theoretical value of the universal constant $R^+_\xi = \nu\left(\frac{n}{4\pi}\right)^{1/d}$ in three dimensions for n = 1, d = 3 and ν = 0.64 which equal 0.2710 [16].

Experimental

Methodology

In this work two binary mixtures were used the phenol–water binary mixture and phenol–cyclohexane binary mixture. The viscosities were measured for both at different temperatures and concentrations. The critical temperature, concentration, heat capacity at constant pressure and density were measured of each mixture.

Experimental Apparatus

Viscosity Apparatus:

- **Capillary Viscometer:** is a device used to measure the viscosity of the liquid with a known density by measuring the time for a known volume of the liquid to flow through the capillary under the influence of gravity [25].

- **Brookfield Digital Viscometer Model DV-I+:** It measures the viscosity of a liquid in centipoises with accuracy ± 1%. It is used to measure the dynamic viscosity from 1 up to$1.33 \times 10_7$ cP.

Density Apparatus: A pycnometer is used to measure the density of the mixtures and pure substances. The Analytical Balance HR-200: It is used to measure the mass in gm unit with accuracy of ± 0.00005%.

Temperature Apparatus: Digital Prima long Thermometer it is used to measure the temperature of the samples with accuracy ± 1%, the range of the temperature from -20 up to 100°C. Julabo F25-MV Refrigerated and Heating Circulator is used to control the temperature of the samples with accuracy 1%.

The calorimeter: It is an instrument used to measure the heat of chemical reactions or physical changes as well as heat capacity. A calorimeter has been constructed with glass pyrex beaker instead of the aluminum, and a nichrome resistance wire covered by a U- tube glass. This calorimeter has been constructed to avoid the reaction of phenol with metals.

Results and Discussion

Viscosity measurements

Phenol–Water Binary Mixture: The results of the dynamic viscosity η as a function of temperature for different concentrations of phenol–water binary mixture are evaluated. The dynamic shear viscosities of phenol–water binary mixture are plotted as a function of temperature at different concentrations of phenol in appendix B. The critical temperature occurs when the two phases of the binary mixture become one phase which appears as anomaly at 67.0°C for the concentration 33.90% by weight of phenol, as shown in Figure 1. In addition, the mixture was visually observed as one phase at the critical temperature and concentration.

Phenol–cyclohexane binary mixture: The results of the dynamic viscosity η as a function of temperature for different concentrations of phenol–cyclohexane binary mixture are evaluated. The dynamic shear viscosities of phenol–cyclohexane binary mixture are plotted as a function of temperature at different concentrations of phenol in appendix B.

The critical temperature occurs when the two phases of the binary

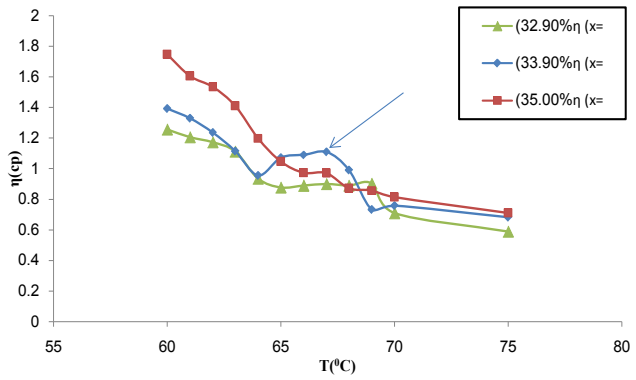

Figure 1: The dynamic shear viscosity of phenol–water mixture as a function of temperature at concentrations 32.90%, 33.90% and 35.00% by weight of phenol.

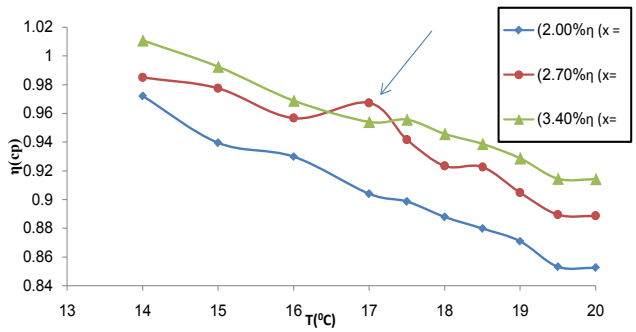

Figure 2: The dynamic shear viscosity of phenol–cyclohexane mixture as a function of temperature at concentrations 2.00%, 2.70% and 3.40% by weight of phenol.

mixture become one phase which appears as anomaly at 17.0°C for the concentration 2.70 % by weight of phenol, as shown in Figure 2. In addition, the mixture was visually observed as one phase at the critical temperature and concentration.

Mass density measurements

Phenol–Water Binary Mixture: The critical mass density ρ_c = 0.8952 g/cm^3 ± 0.0001 at the critical concentration x_c = 33.90% by weight of phenol and the critical temperature T_c = 67.0°C for the binary mixture phenol–water.

Phenol–Cyclohexane Binary Mixture: The critical mass density ρ_c = 0.7627 g/cm^3 ± 0.0001 at the critical concentration x_c = 2.70% by weight of phenol and the critical temperature T_c = 17.0°C for the binary mixture phenol–cyclohexane.

Specific heat measurements

Phenol–water binary mixture: The specific heat at constant pressure c_p for the binary mixture phenol–water is calculated using the relation:

$$W = IVt = (m_r c_r + m_b c_b + m_g c_g + m_p c_p)\Delta T$$

The specific heat at constant pressure is given by relation (2.4) which is

$$c_p = c^+_{pc}\tau^{-\alpha} + c^+_{pb} \quad (2.4)$$

Where t is the reduced temperature and α is the critical exponent which equals 0.11. The specific heat at constant pressure and $\tau^{-\alpha}$ at different temperatures are presented in Table 1.

The specific heat at constant pressure is plotted with $\tau^{-\alpha}$ in Figure 3, using the relation

$$c_p = c^+_{pc}\tau^{-\alpha} + c^+_{pb}$$

The slope of the line is c_{pc} = 78.12 ± 0.04 J/kg.K,, which is the critical amplitude of specific heat at constant pressure of the binary mixture phenol–water, and the intercept is c_{pb} = 85.29 ± 0.02 J/kg.K which is the background amplitude of specific heat at constant pressure.

Calculation of the universal quantity R^+_ξ

The Two–Scale–Factor Universality relation (2.6),

$$R^+_\xi = \xi_0 \left(\frac{\alpha c_{pc}\rho_c}{K_B}\right)^{1/d} = \xi_0 \left(\frac{\alpha T_c \alpha_{pc}}{T_c' K_B}\right)^{1/d} \quad (2.6)$$

The universal quantity R^+_ξ can be calculated using the first term

$$R^+_\xi = \xi_0 \left(\frac{\alpha c_{pc}\rho_c}{K_B}\right)^{1/d}$$

Where the critical exponent α= 0.11, Boltzmann's constant K_B = 1.38×10^{-23} J/K, and the dimensionality d = 3.

Phenol–water binary mixture: The universal quantity R^+_ξ can be calculated for the binary mixture phenol–water to be 0.2716 ± 0.0125 by substituting the values of c_{pc} = 78.12 J/kg.K, ρ_c = 0.8952 gm/cm^3,

Temperature (°C)	$\frac{T - T_c}{T_c}$	$\tau^{-\alpha}$	c_p (J/kg.K)
68.0	0.014925	1.588065	30.4
68.5	0.022388	1.518792	29.9
69.0	0.029851	1.471482	29.4
69.5	0.037313	1.435803	28.6
70.0	0.044776	1.407294	27.2
71.0	0.059701	1.363458	23.5
72.0	0.074627	1.330398	19.5
73.0	0.089552	1.303982	16.4
74.0	0.104478	1.282057	13.7
75.0	0.119403	1.263364	10.9

Table 1: Specific heat data of binary mixture phenol - water.

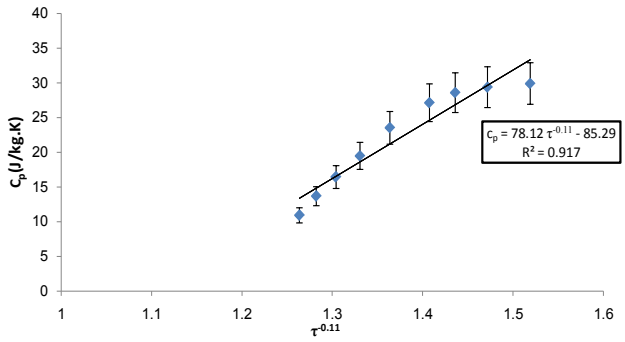

Figure 3: The specific heat at constant pressure c_p versus $\tau^{-0.11}$ for binary mixture phenol–water.

and ξ_0 = 3.3Å [26]. The theoretical value of $R^+_\zeta = v(\frac{n}{4\pi})^{\frac{1}{d}}$ in three dimensions for n = 1, d = 3 and ν = 0.64 , equal 0.2710.

Phenol–cyclohexane binary mixture: The universal quantity R^+_ξ can be calculated for the binary mixture phenol–cyclohexane to be 0.2699 ± 0.0125 by substituting the values of c_{pc} = 106.60 J/kg.K [27], ρ_c = 0.7627 gm/cm³, and ξ_0 = 3.12Å. The theoretical value of R^+_ζ = 0.2710. This indicates that the two binary liquid critical mixtures phenol–water and phenol–cyclohexane belong to the same class of universality "Two–Scale–Factor Universality".

Calculation of $T_c^{'}$

The Two–Scale–Factor Universality relation (2.6),

$$R^+_\xi = \xi_0 \left(\frac{\alpha c_{pc}\rho_c}{K_B}\right)^{1/d} = \xi_0 \left(\frac{\alpha T_c \alpha_{pc}}{T_c^{'} K_B}\right)^{1/d} \quad (2.6)$$

The pressure derivative of the critical temperature along the critical line $T_c^{'}$ can be calculated using the second term

$$R^+_\xi = \xi_0 \left(\frac{\alpha T_c \alpha_{pc}}{T_c^{'} K_B}\right)^{\frac{1}{d}}$$

Phenol–water binary mixture: The pressure derivative of the critical temperature along the critical line $T_c^{'}$ for the binary mixture phenol–water can be calculated to be 9.722 ×10⁻⁶ K/Pa. The values of T_c = 340 K, α_{pc} = 0.002 K⁻¹ [28], ξ_0 = 3.3Å [26] and R^+_ξ = 0.2716.

Phenol–cyclohexne binary mixture

The critical amplitude of isobaric thermal expansion coefficient α_{pc} is need to calculate $T_c^{'}$. The isobaric thermal expansion coefficient α_p can be calculated using the relation:

$$\alpha_p = \frac{1}{V}\left(\frac{dV}{dT}\right)_p \quad (4.1)$$

Where V: is the volume, T: is the temperature and P: is the pressure. Equation 4.1 could be expressed in another form by applying $V = \frac{m}{\tilde{n}}$.

$$\alpha_p = -\frac{1}{\rho}\left(\frac{d\rho}{dT}\right)_p = \alpha_p = \rho\left(\frac{d\rho^{-1}}{dT}\right) \quad (4.2)$$

Where, m : is the mass and ρ: is the density of the binary mixture. The values of the density and its reciprocal at different temperatures above the critical point are presented in Table 2. The reciprocal of the density is fitted with the corresponding temperatures and the slope $\left(\frac{d\rho^{-1}}{dT}\right)$ is determined from Figure 4.

T(K)	Mass density ρ(gm/cm³)	ρ^{-1}(cm³/gm)
290.5	0.7376	1.3561
291.0	0.7374	1.3565
291.5	0.7372	1.3568
292.0	0.7370	1.3572
292.5	0.7368	1.3574
293.0	0.7367	1.3579
293.5	0.7364	1.3589
294.0	0.7359	1.3592
295.0	0.7357	1.3602
298.0	0.7352	1.3616

Table 2: The mass density and its reciprocal values at different temperatures for the binary mixture phenol - cyclohexane.

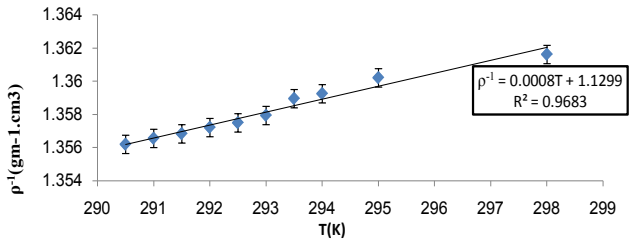

Figure 4: The reciprocal of density for the binary mixture phenol–cyclohexane as a function of temperature.

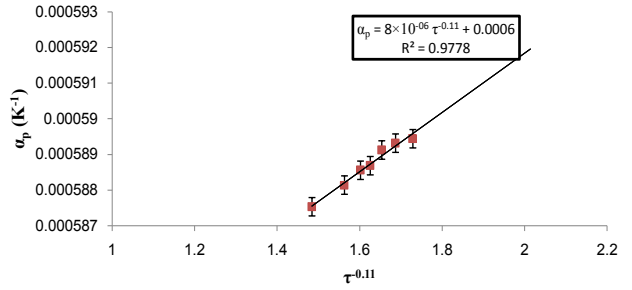

Figure 5: The isobaric thermal expansion coefficient for the binary mixture phenol–cyclohexane as a function of $\tau^{-0.11}$.

The slope from Figure 5 is $\frac{d\rho^{-1}}{dT} = 0.0008\frac{cm^3}{g.K}$

The critical and the background isobaric thermal expansion coefficients can be determined by linear fitting of the isobaric thermal expansion coefficient α_p versus $\tau^{-\alpha}$, where $\tau = \frac{T - T_c}{T_c}$ and α =0.11 depending on the relation:

$$\alpha_p = \alpha_{pc}\tau^{-\alpha} + \alpha_{pb} \quad (2.4)$$

The values of α_p are calculated using equation (4.2) and the data are presented in Table 3. The data of the isobaric thermal expansion coefficient α_p are plotted versus $\tau^{-0.11}$ as shown in Figure 5. The slope of the line represents the critical isobaric thermal expansion coefficient α_{pc} = 8×10⁻⁶ K⁻¹, and the intercept of the line represents the background isobaric thermal expansion coefficient α_{pb}= 6 × 10⁻⁴ K⁻¹.

The pressure derivative of the critical temperature along the critical line $T_c^{'}$ for the binary mixture phenol–cyclohexane is calculated to be 2.8572 × 10⁻⁸ K/Pa. The values of T_c = 290 K, α_{pc} = 6 × 10⁻⁶ K⁻¹, ξ_0 = 3.12Å [26] and R^+_ξ = 0.2699.

Conclusion

The dynamic shear viscosity of two binary liquid mixtures phenol–water and phenol–cyclohexane has been measured at different temperatures and concentrations. The critical temperature and critical concentration for the binary liquid mixture phenol–water were T_c=67.0°C and x_c=33.90 by weight of phenol. The critical density ρc for the binary liquid mixture phenol–water was found to be 0.8952 g/cm3 at the critical temperature and concentration.

The specific heat at constant pressure cp of the binary liquid mixture phenol–water has been measured; the critical cpc and back ground cpb amplitudes of the specific heat at constant pressure have been calculated to be 78.11 J/kg.K and 85.29 J/kg.K respectively.

T(K)	$a_p(K^{-1})$	$T^{-0.11}$
292.0	0.000589	1.728823
292.5	0.000589	1.686904
293.0	0.000589	1.65341
293.5	0.000589	1.62561
294.0	0.000589	1.601907
295.0	0.000588	1.563065
298.0	0.000588	1.484307

Table 3: The isobaric thermal expansion coefficient at different temperatures for the critical mixture of phenol - cyclohexane.

Parameter	This work	Previous works
T_c	67.0°C	66.4°C [a] 69.0°C [b]
x_c	33.90%	34.6% [a] 34.0% [b]
ρ_c	0.8952g/cm³	-
cp_c	$78.117 \frac{J}{k_g.K}$	-
cp_b	$85.292 \frac{J}{k_g.K}$	-
T'_c	9.722×10^{-6} K/Pascal	-
R^+_ξ	0.2716	-

Table 4: Summary of the measured and calculated results in this work and previous works for phenol - water binary.

Parameter	This work	Previous work
T_c	17.0°C	-
x_c	2.70%	-
ρ_c	0.7627g/cm³	-
a_{pc}	8×10^{-6} K^{-1}	
a_{pb}	6×10^{-4} K^{-1}	
T'_c	2.8572×10^{-8} K/Pa	-
R^+_ξ	0.2699	-

Table 5: Summary of the measured and calculated results in this work for phenol - cyclohexane binary mixture.

The pressure derivative of the critical temperature along the critical line T_c^' is calculated for the binary mixture phenol–water to be 9.722×10-6 K/Pa.

The critical temperature and critical concentration for the binary liquid mixture phenol–cyclohexane have been measured, the results were T_c=17.0°C and x_c=2.70% by weight of phenol. The critical density ρc for the binary liquid mixture phenol–cyclohexane is found to be 0.7627 g/cm³ at the critical temperature and concentration. The pressure derivative of the critical temperature along the critical line T_c^' is calculated for the binary mixture phenol–cyclohexane to be 2.8572 × 10-8K/Pa. The isobaric thermal expansion coefficient αp for phenol–cyclohexane binary mixture are calculated at different temperatures, the critical αpc and back ground αpb amplitudes is determined to be 8×10^{-6} K-1, 6×10^{-4} K-1 respectively. The measured and calculated parameters for phenol–water binary mixture are summarized in Table 4. The measured and calculated parameters for phenol–cyclohexane binary mixture are summarized in Table 5. The universal quantity R+ξ for the binary liquid critical mixture phenol–water is calculated to be 0.2716 ± 0.0005. In addition, the universal quantity R+ξ for the binary liquid critical mixture phenol–cyclohexane is calculated to be 0.2699 ± 0.0001. The calculated values of the universal quantity R+ξ are in a good agreement with the theoretical value of R+ζ which is equal 0.2710. As a result the two binary liquid critical mixtures belong to the class of universality "Two–Scale–Factor Universality".

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