The Addition of N-Butanol in Ethanol-Isooctane Mixture to Reduce Vapor Pressure of Oxygenated-Gasoline Blend

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ABSTRACT

In this work, vapor pressure of binary systems for isooctane + ethanol, isooctane + n-butanol and ethanol + n-butanol and ternary system for isooctane + ethanol + n-butanol were measured in the temperature range from 313.15 to 318.15 K using the inclined ebulliometer. The experimental results showed that the existence of n-butanol in isooctane decreases the vapor pressure of mixture, while increasing n-butanol fraction in ternary isooctane-ethanol-n-butanol mixture decreased vapor pressure of mixture. Experimental data for binary systems studied were correlated with Wilson, NRTL and UNIQUAC models with average relative deviation (ARD) of 3.5%. The optimized binary parameter pairs obtained in this work were used to estimate the ternary system. The Wilson model gave the best performance for estimation of ternary system with ARD of 5.4%. All systems studied showed non-ideal solution with positive deviation from Raoult's law.

***Keywords**: vapor pressure; n-butanol; ethanol; isooctane; gasoline*

ABSTRAK

Pada penelitian ini, tekanan uap sistem biner isooktana + etanol, isooktana + n-butanol dan etanol + n-butanol serta tekanan uap sistem terner isooktana-etanol-n-butanol telah diukur pada suhu 313,15–318,15 K menggunakan inclined ebulliometer. Hasil eksperimen menunjukkan bahwa adanya n-butanol dalam isooktana dapat menurunkan tekanan uap campuran. Peningkatan fraksi n-butanol pada campuran terner isooktana-etanol-n-butanol dapat menurunkan tekanan uap pada campuran tersebut. Data eksperimen sistem biner telah dikorelasi dengan model Wilson, NRTL dan UNIQUAC dengan average relative deviation (ARD) 3,5%. Parameter biner yang telah dioptimasi digunakan untuk memperkirakan tekanan uap sistem terner. Model Wilson memberikan perkiraan tekanan uap terbaik pada sistem terner dengan ARD 5,4%. Semua sistem yang dipelajari menunjukkan larutan non-ideal dengan penyimpangan positif dari Hukum Raoult.

***Kata Kunci**: tekanan uap; n-butanol; etanol; isooktana; bensin*

INTRODUCTION

The usage of base gasoline as fuel gives some environmental issues because of its high carbon monoxide (CO) and unburned hydrocarbons content in exhaust gas emission [1]. Oxygenated compounds are required as fuel additive to reduce exhaust gas emission. Alcohols have been known as oxygenated compound and octane booster in gasoline [2]. One of the alcohols widely used as gasoline additive is ethanol because of its availability from biomass[3]. Ethanol has high octane number [2] and low toxicity [4] that may increase the quality of gasoline. However, ethanol has low heating value and it increases the vapor pressure significantly when mixed with gasoline [5]. The vapor pressure for mixtures is one of important physical property of materials that must be concerned for designing gasoline blend. The increasing of vapor pressure gives several problems such as rising evaporative emission and tendency to vapor lock in automobile engine [6].

To cover these problems, ethanol may be mixed with longer carbon chain alcohol i.e. *n*-butanol. N-butanol is known as green fuel because it could be made from fermentation process [7,8]. Because of its long carbon chain, *n*-butanol has lower water solubility and high distribution with hydrocarbons. In addition, *n*-butanol has higher heating value and lower vapor pressure than ethanol. However, some effort has been done to decrease vapor pressure of gasoline blends i.e. by the addition of diethyl carbonate as gasoline [9] but this compound is far more expensive than *n*-butanol. With these properties, ethanol-butanol mixture may be considered as promising oxygenated gasoline

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Table 1. Material description and properties of pure components

Component	Source	Mass fraction purity^a	MW^a (g.mol^{-1})
isooctane	Merck, Germany	0.995	114.23
ethanol	Merck, Germany	0.999	46.07
n-butanol	Merck, Germany	0.995	74.12

^{a}from supplier

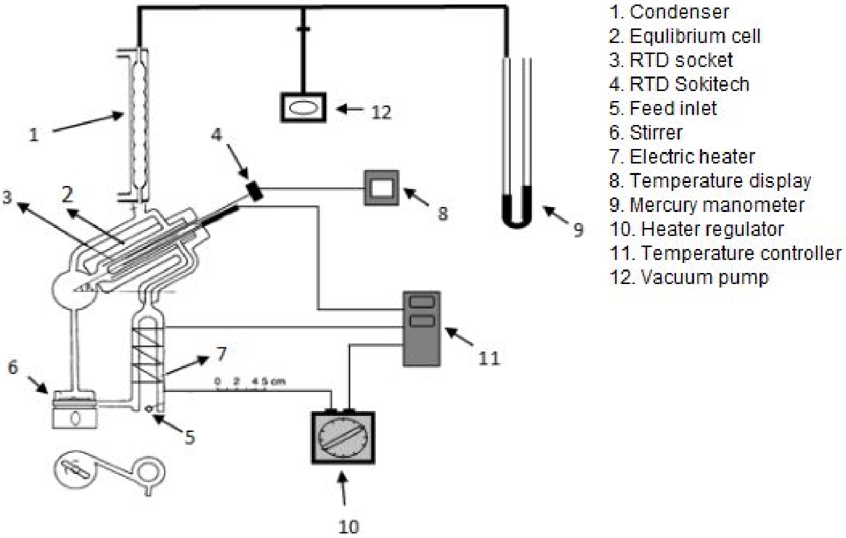

Fig 1. The schematic diagram of inclined ebulliometer used in this work

additive to get its advantage with suppress the deficiency and keep the economic value.

In order to design applicable oxygenated (*n*-butanol/ethanol) - gasoline blend, the knowledge of vapor pressure values for the systems containing ethanol, *n*-butanol and isooctane is necessary. Several studies have been reported for vapor pressure measurement of the binary systems containing isooctane, ethanol and *n*-butanol [10-12]. There was only one study reporting for vapor pressure of the ternary system containing isooctane, *n*-butanol and 1-hexene (without ethanol) [13]. Based on our knowledge, there are no published data in DIPPR (The Design Institute for Physical Properties), Dechema, NIST (National Institute of Standards and Technology) or related journals for vapor pressure measurement of the ternary system containing isooctane, ethanol and n-butanol.

Therefore in this study, vapor pressure of binary systems for isooctane + ethanol, isooctane + *n*-butanol and ethanol + *n*-butanol and ternary system for isooctane + ethanol + *n*-butanol were measured in the temperature ranging from 313.15 to 318.15 K using the inclined ebulliometer. The experimental data of binary systems were correlated using Wilson [14], NRTL [15] and UNIQUAC [16] models to obtain binary interaction parameters. These binary parameters were used to predict the vapor pressure of ternary system. The predicted vapor pressure values were compared with the experimental ternary system data.

EXPERIMENTAL SECTION

Materials

Ethanol, *n*-butanol and isooctane were used as materials in this work. These materials were purchased from Merck, Germany with materials properties and descriptions are shown in Table 1 and no additional purification was done with the materials.

Instrumentation

The apparatus used in this work was inclined ebulliometer which was designed by Li et al. [17]. There are three segments of the apparatus: ebulliometer cell, temperature control system and vacuum system. The ebulliometer cell consists of heater flask, equilibrium cell, condenser and stirrer. The temperature control system consists of electric heater, temperature controller (Shimaden SR64) and RTD temperature sensor (Sokitech) with accuracy of 0.1 K. For the vacuum system, there are a vacuum pump which can remove air and impurities from ebulliometer and U-shape mercury manometer to measure the pressure. The schematic diagram of the apparatus is shown in Fig. 1.

Table 2. Parameter for the Wagner equation of pure compounds*.

Component	A	B	C	D	Tc (K)	Pc (bar)
isooctane[a]	-7.389	1.2529	-3.1661	-2.2200	544	25.7
ethanol[a]	-8.518	0.3416	-5.7368	8.3258	513.9	61.4
n-butanol[a]	-8.007	0.5378	-9.3424	6.6869	563.1	44.2

[a]Ref [19]

* $\ln P_r^{sat} = \left\{A(1-Tr)+B(1-Tr)^{1.5}+C(1-Tr)^{2.5}+D(1-Tr)^{5}\right\}/Tr$, where $Tr = T/Tc$ and $P_r^{sat} = P^{sat}/Pc$

Table 3. Vapor pressure data of isooctane (1) + ethanol (2) binary system

T (K)	P (kPa)	T (K)	P (kPa)	T (K)	P (kPa)
Ethanol content 10 wt%		*Ethanol content 20 wt%*		*Ethanol content 30 wt%*	
303.3	13.06	303.2	14.93	302.5	16.66
305.6	16.13	305.4	17.19	305.2	19.73
308.4	17.86	308.2	19.33	308.2	21.19
310.3	20.79	310.5	22.93	310.1	23.73
313.2	22.79	312.9	25.19	312.9	25.86
315.5	25.86	315.4	27.19	315.3	29.06
318.2	27.86	318.2	29.46	317.5	31.73
Ethanol content 40 wt%		*Ethanol content 50 wt%*			
302.8	16.66	302.6	16.79		
305.0	19.86	305.0	18.79		
308.2	21.33	308.2	20.79		
309.9	23.59	309.7	22.39		
312.6	25.99	312.8	25.59		
315.2	29.59	315.2	28.13		
317.7	32.39	318.0	31.73		

Table 4. Vapor pressure data of isooctane (1) + *n*-butanol (3) binary system

T (K)	P (kPa)	T (K)	P (kPa)	T (K)	P (kPa)
n-butanol content 10 wt%		*n-butanol content 20 wt%*		*n-butanol content 30 wt%*	
303.4	8.13	303.2	8.13	303.3	7.86
305.7	9.19	305.4	9.06	305.6	8.79
308.2	10.39	308.2	10.26	308.2	9.59
311.6	12.53	310.5	11.06	310.5	10.93
313.1	13.06	313.2	11.86	312.9	11.59
315.1	13.99	315.7	13.46	315.7	12.79
317.6	15.33	318.3	14.26	318.3	14.26
n-butanol content 40 wt%		*n-butanol content 50 wt%*			
303.3	7.06	303.2	6.26		
305.2	8.39	305.1	7.86		
308.2	9.19	308.2	8.79		
310.3	10.93	311.3	10.53		
312.7	11.73	313.6	11.59		
315.5	13.19	315.9	12.39		
318.2	15.46	318.3	13.19		

Procedure

The solution of certain composition was prepared gravimetrically and introduced into ebulliometer cell. After that, the vacuum pump was used to remove air and impurities from the apparatus. The temperature controller was set at desired temperature by heating the heater flask. When the temperature was reached and the pressure in equilibrium cell gave constant value, the system was in equilibrium state. The temperature and pressure at this condition was recorded as equilibrium value. The same procedure was repeated for other composition of solution (binary and ternary system) and other temperature variables as listed in Tables 3–6.

Table 5. Vapor pressure data of ethanol (2) + *n*-butanol (3) binary system

T (K)	P (kPa)	T (K)	P (kPa)	T (K)	P (kPa)
n-butanol content 10 wt%		*n-butanol content 20 wt%*		*n-butanol content 30 wt%*	
303.2	9.19	303.4	9.06	303.2	8.53
305.4	11.19	305.4	10.53	306.2	11.06
308.2	12.66	308.2	12.39	308.2	12.13
310.3	14.26	310.4	14.13	310.2	13.73
313.6	16.93	313.3	15.19	313.0	15.86
315.3	18.13	315.7	17.73	315.7	17.86
318.2	21.99	318.4	20.93	318.5	21.19
n-butanol content 40 wt%		*n-butanol content 50 wt%*			
303.6	8.53	302.9	6.79		
305.4	10.26	305.5	9.59		
308.2	11.86	308.2	10.79		
310.4	13.19	310.5	12.26		
313.3	15.06	313.3	13.73		
315.7	16.53	316.8	16.79		
318.2	19.99	318.4	18.79		

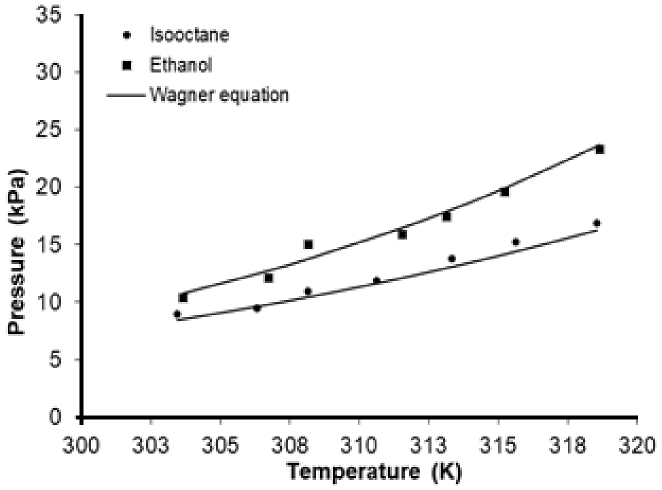

Fig 2. The comparison between pure vapor pressure experimental data and calculated values

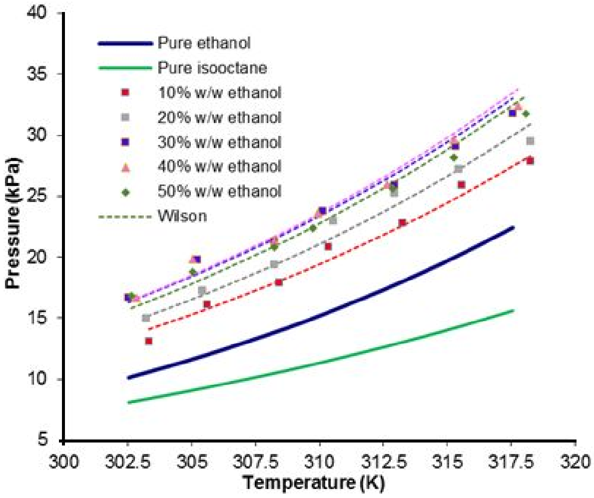

Fig 3. The comparison between experimental data and calculated vapor pressure by the Wilson model for isooctane + ethanol binary system

RESULT AND DISCUSSION

Vapor Pressure of Pure Components

The vapor pressures of pure components have been measured for isooctane and ethanol. These pure vapor pressure experimental data were compared with literature values which were calculated using Wagner equation with constants which were listed in Table 2. The comparison between pure vapor pressure experimental data and calculated values are shown in Fig. 2. From this comparison, the experimental data agreed well with the literature values with average relative deviation (ARD) value of 3.5% for isooctane and 3.0% for ethanol.

Vapor Pressure of Binary Systems

The vapor pressure measurement of binary systems is required to understand how the physical behavior between each pair of chemical used in these systems. The experimental vapor pressure data for isooctane (1) + ethanol (2), isooctane (1) + *n*-butanol (3) and ethanol (2) + *n*-butanol (3) binary systems are presented in Tables 3–5. The vapor pressure of isooctane – ethanol mixtures were found higher than pure isooctane and pure ethanol as shown in Fig. 3. The vapor pressures were increasing during increasing of ethanol fraction in isooctane. The isooctane – ethanol mixtures showed strongly non-ideal solution and azeotropic phenomenon because of the difference

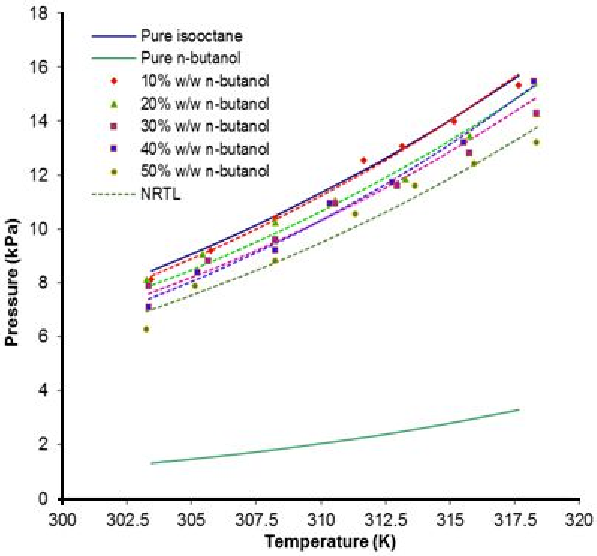

Fig 4. The comparison between experimental data and calculated vapor pressure by the NRTL model for isooctane + *n*-butanol binary system

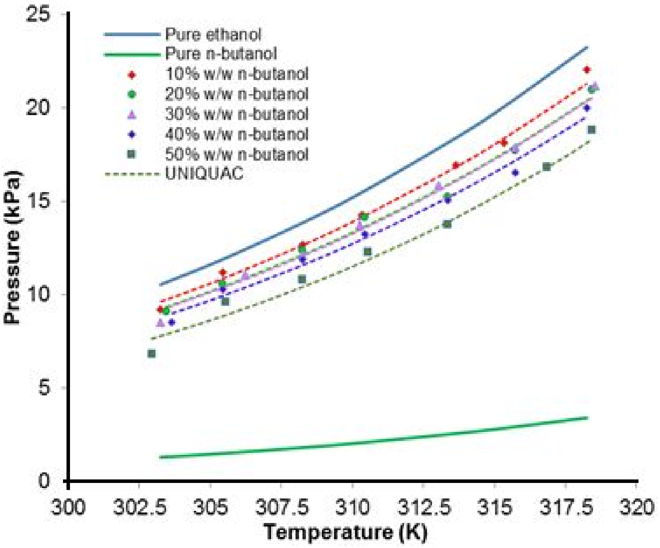

Fig 5. The comparison between experimental data and calculated vapor pressure by the UNIQUAC model for ethanol + *n*-butanol binary system

Table 6. Vapor pressure data of isooctane (1) + ethanol (2) + *n*-butanol (3) ternary system

T (K)	P (kPa)	T (K)	P (kPa)	T (K)	P (kPa)
Isooctane 92.5%, ethanol 5%, n-butanol 2.5% (wt%)		*Isooctane 90% ethanol 5%, n-butanol 5% (wt%)*		*Isooctane 87.5%, ethanol 5%, n-butanol 7.5% (wt%)*	
302.9	11.06	302.9	11.06	302.9	11.46
305.5	12.66	305.4	12.93	305.5	12.66
308.2	14.53	308.2	14.79	308.3	14.93
310.3	16.39	310.4	16.66	310.5	16.39
313.2	17.59	313.3	17.99	313.4	18.39
315.4	19.33	315.6	19.19	315.7	19.73
318.0	21.73	318.2	21.99	318.4	22.26

Table 7. Physical properties and parameters of pure components used in the activity coefficients correlation[a]

component	V^L/cm^3.mol^{-1}	*r*	*q*
isooctane	166.62	5.8463	5.008
ethanol	58.68	2.5755	2.588
n-butanol	91.96	3.9243	3.668

[a] Ref [19]

of chemical structure and polarity between isooctane and ethanol. Meanwhile, the n-butanol addition in isooctane caused lower vapor pressure than pure isooctane vapor pressure as shown in Fig. 4. The isooctane – *n*-butanol mixtures showed non-ideal solution because after the addition of n-butanol up to 50% w/w, the vapor pressure were slightly changing from isooctane pure vapor pressure. This happens because isooctane and butanol have different chemical groups (hydrocarbon and alcohol) even though *n*-butanol has longer chain carbon. Fig. 5 showed the *n*-butanol addition in ethanol which caused lower vapor pressure than the pure ethanol vapor pressure. The ethanol – *n*-butanol mixtures showed non-ideal solution even these compounds have the same groups. This happens because *n*-butanol has longer chain carbon compared with ethanol.

The experimental data of binary systems were correlated by using Wilson, NRTL and UNIQUAC models to obtain the fitted binary parameters. The physical properties and parameters of each component used in the activity model correlation were shown in Table 7. Barker's method [18] was used as objective function to optimize the binary parameters for these systems studied in this work which was described by Equation (1):

$$OF = \sum_{i=1}^{n} \left(P_{i,cal} - P_{i,exp}\right)^2 \qquad (1)$$

where *n* is the number of data points, *P* is the vapor pressure, and the subscripts *cal* and *exp* refer to calculated and experimental values, respectively. Due to low pressure condition, the vapor phase is assumed

Table 8. The fitted binary parameter of activity coefficient model and average relative deviation (ARD)

Model	Parameters			*ARD* (%)
	A_{12} (cal·mol^{-1})	A_{21} (cal·mol^{-1})	α	
	isooctane (1) + ethanol (2)			
Wilson[a]	5065.53	1091.53		3.69
NRTL[b]	-272.08	2335.45	0.2	3.65
UNIQUAC[c]	46.34	410.55		3.53
	isooctane (1) + *n*-butanol (3)			
Wilson[a]	524.07	1008.69		3.33
NRTL[b]	1077.33	23.60	0.2	3.26
UNIQUAC[c]	516.65	-186.61		3.21
	ethanol (2) + *n*-butanol (3)			
Wilson[b]	517.99	108.91		3.71
NRTL[c]	257.57	254.00	0.2	3.88
UNIQUAC[d]	-447.20	7876.21		3.82

[a] $A_{ji} = \lambda_{ji} - \lambda jj$; [b] $A_{ji} = g_{ji} - g_{ii}$; [c] $A_{ji} = u_{ji} - u_{ii}$

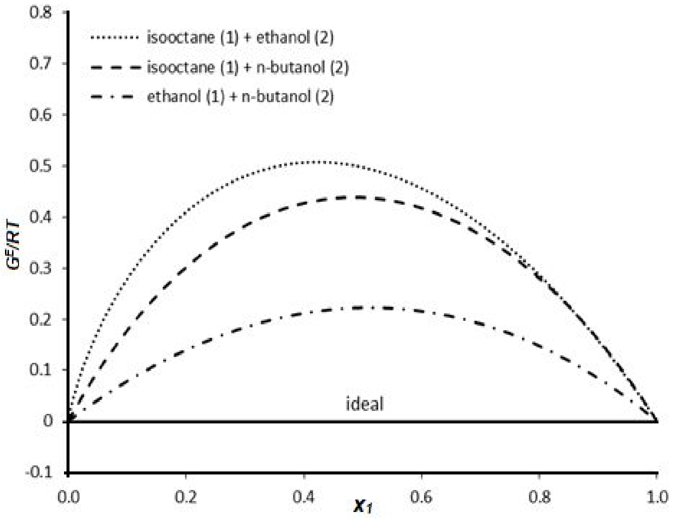

Fig 6. The excess gibbs energy values of the binary systems at 308.15 K

as ideal gas and the calculated vapor pressure can be obtained using the following equation:

$$P_{cal} = \sum_{i=1}^{m} x_i \gamma_i P_i^s \tag{2}$$

where *m* is the number of component in the mixture, x_i is the liquid mole fraction of component *i*, γ_i is the activity coefficient of component *i* and P_i^s is the pure vapor pressure of the component *i* which calculated by Wagner equation. The best fitted binary interaction parameters were listed in Table 8.

The average relative deviation (ARD) of vapor pressure between experimental data and calculated values were used to evaluate the correlation which is described by:

$$ARD = \frac{1}{n}\sum_{i-1}^{n}\left|\frac{P_{cal} - P_{exp}}{P_{exp}}.100\%\right| \tag{3}$$

For isooctane (1) + ethanol (2) system, the experimental data were well correlated using Wilson, NRTL and UNIQUAC models with ARD values of 3.7%, 3.% and 3.53% respectively. Isooctane (1) + *n*-butanol (3) system give ARD values of 3.3%, 3.3% and 3.2% respectively. Ethanol (2) + *n*-butanol (3) system give ARD values of 3.7%, 3.9% and 3.8% respectively. Fig. 3–5 showed the comparison between experimental data and calculated vapor pressure by the models. The correlation lines in the Figures showed good agreement with the experimental data.

From the activity coefficient analysis based on Wilson, NRTL, and UNIQUAC models, the physical behavior may be explained for each binary system studied. The non-ideality of solution could be expressed as excess Gibbs energy where excess Gibbs energy is the deviation between actual energy Gibbs and ideal energy Gibbs. Excess Gibbs energy were calculated from activity coefficient as described by:

$$\frac{G^E}{RT} = \sum_{i=1}^{m} x_1 \ln \gamma_i \tag{4}$$

where G^E is the excess Gibbs energy of solution, *R* is the universal gas constant and *T* is the temperature condition.

Fig. 6 showed the excess Gibbs energy values of the binary systems studied based on the fitted activity binary parameters at 308.15 K. For isooctane – ethanol system, the excess Gibbs energy has large values with magnitude in terms of G^E/RT up to 0.5 which indicates the strongly non-ideal of mixtures and gives impact for the vapor pressure of mixtures by Equation 2 with azeotropic behavior. For isooctane – *n*-butanol system, the excess Gibbs energy has large values with magnitude in terms of G^E/RT up to 0.4 which indicates the non-ideal of mixtures. However, n-butanol has very

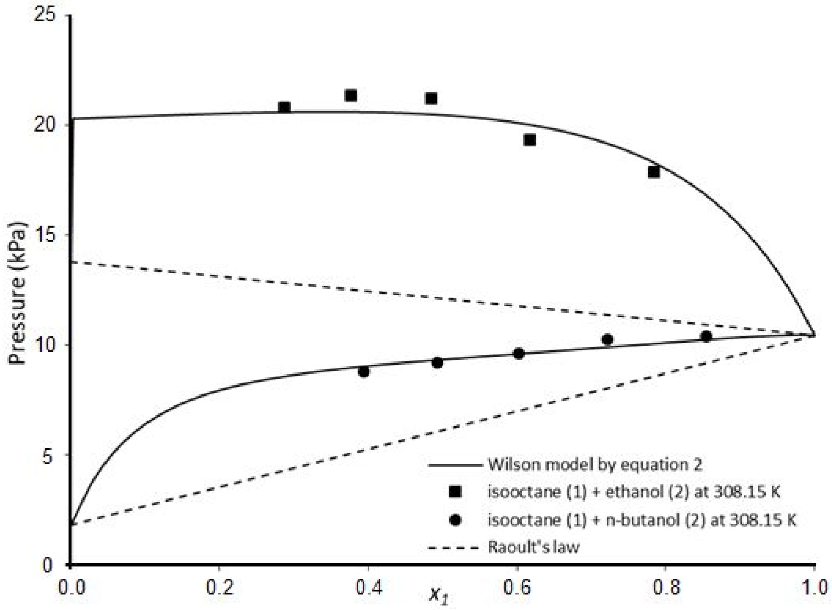

Fig 7. The comparison of the vapor pressure calculated by Equation 2 with the Raoult's law for isooctane (1) + ethanol (2) and isooctane (1) + n-butanol (2) systems at 308.15 K

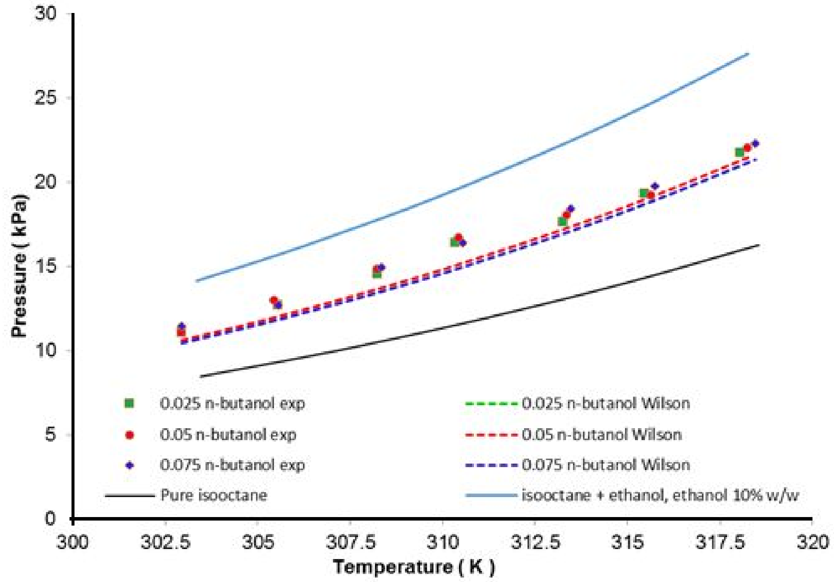

Fig 8. The comparison between experimental data and calculated vapor pressure by the Wilson model for isooctane + ethanol + n-butanol ternary system

low pure vapor pressure and the value of activity coefficient does not give quite impact compared with pure vapor pressure of *n*-butanol. For ethanol – *n*-butanol system, the excess Gibbs energy has quite large values with magnitude in terms of G^E/RT up to 0.2. This indicates the ethanol – *n*-butanol mixtures is non-ideal mixtures because *n*-butanol has long chain carbon even though the compounds have the same group.

To give more reliability for sensing the non-ideality of solution, the vapor pressure measurement data were compared with the Raoult's Law (ideal condition) when γ_i =1 and Equation 2 become as follows:

$$P_{Raoult} = \sum_{i=1}^{m} x_i P_i^s \qquad (5)$$

Fig. 7 showed the comparison of the vapor pressure calculated by Equation 2 with the Raoult's law (Equation 5). The binary systems showed non-ideal solution with positive deviation from Raoult's law because vapor pressure measurement data have higher value than the vapor pressure calculated by Raoult's law.

Vapor Pressure of Ternary System

The experimental vapor pressure data for ternary system of isooctane (1) + ethanol (2) + *n*-butanol (3) have been measured as listed in Table 6. Fig. 8 showed the addition of *n*-butanol in isooctane-ethanol mixtures which could reduce the vapor pressure significantly with small amount of *n*-butanol. The vapor pressure of ternary system with 0.025–0.075 *n*-butanol weight fractions have higher vapor pressure than pure isooctane, but lower vapor pressure with (0.9 isooctane + 0.1 ethanol) mixture. The experimental data of ternary system were compared with prediction values using Wilson, NRTL and UNIQUAC models with the binary interaction parameters from Table 8. Fig. 6 also presented the experimental data and prediction result with Wilson model for the ternary system of isooctane (1) + ethanol (2) + *n*-butanol (3). The activity coefficient models based on binary parameters gave sufficient prediction result of ternary system with ARD value of 5.4% for Wilson, 9.2% for NRTL and 8.9% for UNIQUAC models. The Wilson model gave the best performance for the estimation of ternary system. The ternary system studied showed non-ideal solution and positive deviation with the Raoult's law.

CONCLUSION

In this work, vapor pressure of binary systems for isooctane + ethanol, isooctane + *n*-butanol and ethanol + n-butanol and ternary system for isooctane + ethanol + *n*-butanol were measured in the temperature range from 313.15 to 318.15 K using the inclined ebulliometer. The experimental results showed that the existence of n-butanol in isooctane could decrease the vapor pressure of mixture. The increase of n-butanol fraction in ternary isooctane-ethanol-n-butanol mixture decreased vapor pressure of mixture. The activity coefficient models based on binary parameters gave sufficient prediction result of the ternary system with ARD values of 5.4% for Wilson, 9.2% for NRTL and 8.9% for UNIQUAC models. The Wilson model gave the best performance for the estimation of ternary system. All systems showed non-ideal solution and had positive deviation with the Raoult's law.

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