

*Article*

## Density Determination of Ethyl Acetate-Palm Oil Mixture in Supercritical Condition

Cholada Komintarachat<sup>1,a</sup>, Ruengwit Sawangkeaw<sup>2,b</sup>, and Somkiat Ngamprasertsith<sup>3,4,c,\*</sup>

<sup>1</sup> Program in Petrochemistry, Faculty of Science, Chulalongkorn University, Phayathai Road, Pathumwan, Bangkok 10330, Thailand

<sup>2</sup> The Institute of Biotechnology and Genetic Engineering, Chulalongkorn University, Institute Bldg. 3, 254 Phayathai Rd., Pathumwan, Bangkok 10330, Thailand

<sup>3</sup> Fuels Research Center, Department of Chemical Technology, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand

<sup>4</sup> Center of Excellence on Petrochemical and Materials Technology, Chulalongkorn University, 254 Phayathai Road, Pathumwan, Bangkok 10330, Thailand

E-mail: <sup>a</sup>dachonla@gmail.com, <sup>b</sup>ruengwit.s@chula.ac.th, <sup>c</sup>somkiat.n@chula.ac.th (Corresponding author)

**Abstract.** The non-catalytic transesterification of palm oil in supercritical ethyl acetate in continuous system to produce biofuel is complex to understand thermodynamic properties during extreme condition, particularly density of two reactants. In this study, an indirect procedure for density measurement of palm oil and ethyl acetate mixture in a batch system at high temperature and pressure was experimentally investigated using isochoric method. Its apparatus comprise a constant volume reactor which was individually loaded with a mixture of ethyl acetate and palm oil in different molar ratios (10:1 to 30:1) and global densities (0.26 to 0.53 g/cm<sup>3</sup>). During temperature increment, the changing of pressure was recorded in real-time to obtain the pressure-temperature relationship. After measuring the change of pressure at various global densities, the pressure-temperature related specific global density diagram was successfully constructed. At high global densities and high molar ratios of ethyl acetate to palm oil, the transition point took place closing to the estimated phase boundaries separating the region of vapor-liquid and homogenous phase of the mixture. The results will be further employed as database for accurate residence time calculation in continuous reactor, especially for biofuel production from palm oil in supercritical ethyl acetate.

**Keywords:** Biofuel, density, ethyl acetate, isochoric method, palm oil, supercritical condition.

ENGINEERING JOURNAL Volume 19 Issue 2

Received 15 June 2014

Accepted 5 November 2014

Published 30 April 2015

Online at <http://www.engj.org/>

DOI:10.4186/ej.2015.19.2.29

## 1. Introduction

Biodiesel is a renewable fuel which produced from triglyceride derived from vegetable oils and animal grease. The main reaction of biodiesel production is transesterification reaction between triglyceride and alcohol as illustrated in Fig 1. The alcohols commonly used in transesterification reaction are methanol and ethanol. However, methanol, widely employed as the reacting reagent due to its reasonable price and high reactivity, is commercially synthesized from natural gas. On the other hand, ethanol is alternatively produced by fermentation of agricultural biomass from renewable resources.

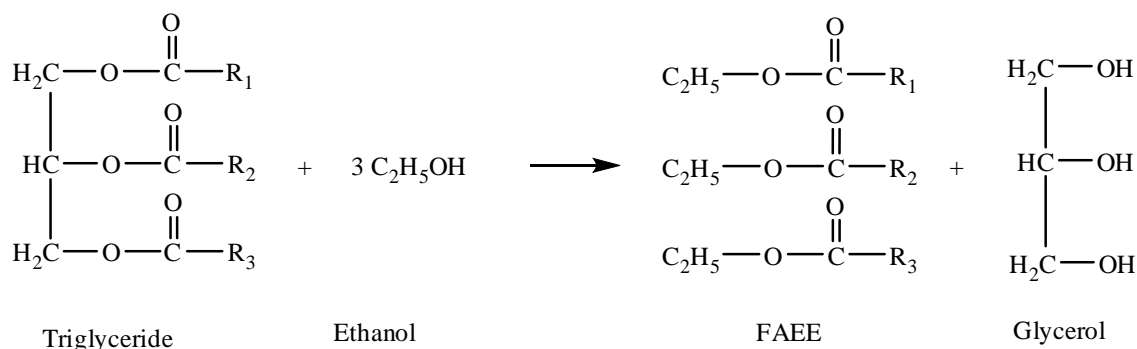


Fig. 1. The transesterification reactions of palm oil (triglycerides) with ethanol.

A non-catalytic transesterification in the continuous system is beneficial in terms of high production efficiency, environmentally friendly process and feedstock flexibility over that of homogeneous catalytic process [1]. It has been developed to produce biodiesel from vegetable oils or animal fats in supercritical condition [2]. As the supercritical conditions provide solubility [3] between heterogeneous phases under the conditions that a liquid and its vapor become identical [4–6], reducing the mass transport limitation between phases and increasing the rate of reactions are simply removed [7]. However, the by-product glycerol may be excessive that poses an additional step in separation and purification.

Phase characteristics in the supercritical reactor during the homogeneous reaction depends on the reactants and reaction conditions [8]; many solvents can be chosen to operate the system in a single fluid phase. A systematic procedure to find solvents to achieve homogeneous conditions has been developed to produce biofuel [9–11]. One among other solvents, ethyl acetate is a carboxylate ester that can be used as supercritical solvent instead of alcohols in transesterification reaction [12], to obtain fatty acid ethyl ester (FAEE) or biofuel and triacetin following the reaction shown in Fig. 2. The ethyl acetate is synthesized from esterification of ethanol and acetic acid produced from renewable resources [13]. Typically, the triacetin is an additive to improve cold flow properties and pour point of biofuel [14].

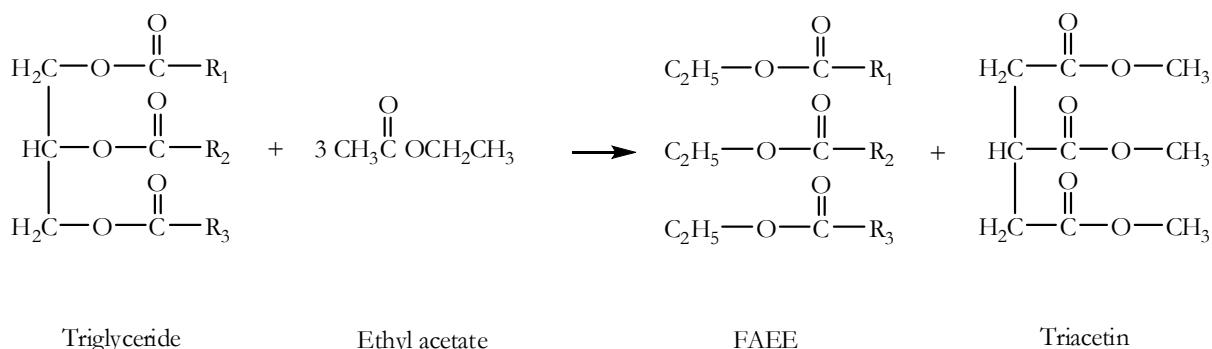


Fig. 2. The transesterification reaction of palm oil (triglycerides) with ethyl acetate.

Due to the complex theory of fluid mixtures, the studies of Economou [15] and You et al. [16] experimentally explored an understanding of the thermodynamic behavior of pure fluids, leading to the development of quantitative prediction methods [17]. Among thermodynamic properties describing variable state of condition such as pressure (p), temperature (T) and density (ρ), the accurate measurement of fluid mixture density is the most difficult to measurement. Recently, Velez et al. [18, 19] measured the

density of reacting mixtures between sunflower oil and supercritical alcohols by isochoric technique in the ranges of 553–618 K temperatures, 100–400 bar pressures and 25–40 alcohol to oil molar ratios. This technique is an attractive procedure for obtaining accurate densities and phase transitions using a close constant volume reactor with temperature and pressure monitoring. Palavra et al. [20] described the principles of the isochoric methods in the research review and determined the amount of fluid inside in the high pressure cell as well as fluid mixtures density measurement. The information obtained from these experiments are not only provided the variation of density with pressure and temperature for each reacting system composition, but also give the conditions in which the system transforms from heterogeneous phase to homogeneous phase [21].

To understand the relationship of temperature and pressure at a given global density, the indirect isochoric method has been proposed for this study. This is due to the fact that, at this condition, the combination of two substances in homogeneity is difficult to directly observe. Additionally, the temperature and pressure determination at constant global density by isochoric method is readily and convenient for mixture at supercritical conditions. In the system of this study, the vegetable oil is completely dissolved in ethyl acetate at atmospheric condition [22] that reduces mass transfer and solubility limitations [23]. The experimental results will show the variation of pressure versus temperature at constant composition of reacting mixture. The relationship of temperature, pressure and global density will be investigated and it can generate a diagram of temperature-pressure-global density individual reaction. It is thus possible to locate the boundaries between the vapor-liquid phase and homogeneous regions for the reacting systems.

## 2. Experimental Section

### 2.1. Materials

The compounds used in this experiment were ethyl acetate (99.97% from Fisher Scientific) and palm olein oil (with a major fatty acid composition w/w of 37% palmitic, 46% oleic and 11% linoleic acids). The palm olein oil was obtained from Morakot Industries Co., Ltd.

### 2.2. Apparatus and Procedures

The measurement cell configuration is shown in Fig. 3 [18]. The global density of ethyl acetate and palm oil mixture was calculated from the known mixture weight divided by the cell volume. The dimension of the measurement cell made from SUS316 stainless steel are 0.95 cm outside diameter and 50 cm length with a working volume of 23 cm<sup>3</sup> (by water substitution) at room temperature. A tubular furnace from Lenton, model 2416CG, was used as a heating source. This cell was insulated with ceramic fiber and aluminum foil to stabilize the inside temperature during the measurement. The pressure in the cell was measured by a high-temperature, tube and socket pressure gauge, model EN 837-1 from Fantinelli Srl with a 10 cm panel diameter.

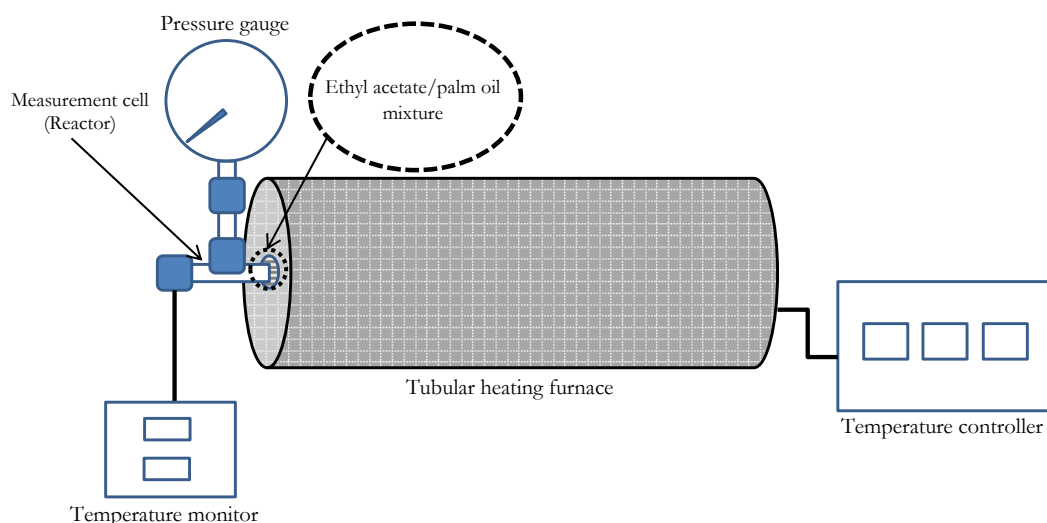


Fig. 3. Schematic diagram of experimental set-up.

At the beginning, the predetermined masses of ethyl acetate and palm oil were added to the cell. The temperature was increased slowly at the rate of 5 K/min from room temperature to 673 K. During the temperature increment, both pressure and temperature were stabilized and recorded. In this way, the pressure variations with temperature of mixtures of a given mass charged into the cell were measured; the global density (mass charged divided by cell volume) was known for each run. This information provided the variation of one value of density with temperature and pressure over the range of 303 K to 673 K and 30 bar to 150 bar. By repeating this procedure for different densities and reacting mixture compositions, the values of density over the range of conditions required were subsequently attained. To obtain precise transition points, all conditions were repeated for three times; repeatability and reproducibility can be achieved. In each condition, the results shown in later sections represent average values with error bars at 95% confidence.

In order to accurately correlate the pressure measurement of the ethyl acetate-palm oil system, the experimental measurement values were compared with the data calculation from the Peng-Robinson equation of state (PR EOS) [24] for predicting the vapor pressure and phase behavior near the critical temperature of single-component system. The values of critical pressure and temperature as well as other key properties of the pure substances (palm oil and ethyl acetate) used in this study are listed in Table 1 [25, 26].

Table 1. Characteristics of pure compounds.

Substance	$T_c$ (K)	$P_c$ (bar)	$M_w$ (g/mol)	$\omega$
Palm oil [25]	954.1	3.6	850.0	1.800
Ethyl acetate [26]	523	38.8	88.11	0.362

### 3. Results and Discussion

#### 3.1. The p-V-T Relationship of Ethyl Acetate

The pressures corresponding to the temperature of ethyl acetate obtained by experiment and calculation by the PR EOS in comparison are shown in Fig. 4 at the constant molar volume of  $9.86 \times 10^{-4} \text{ m}^3/\text{mol}$ . It has apparently shown that the measured data are in good agreement with those from the PR EOS, indicating that there is a potential for the further experimental results being acceptable.

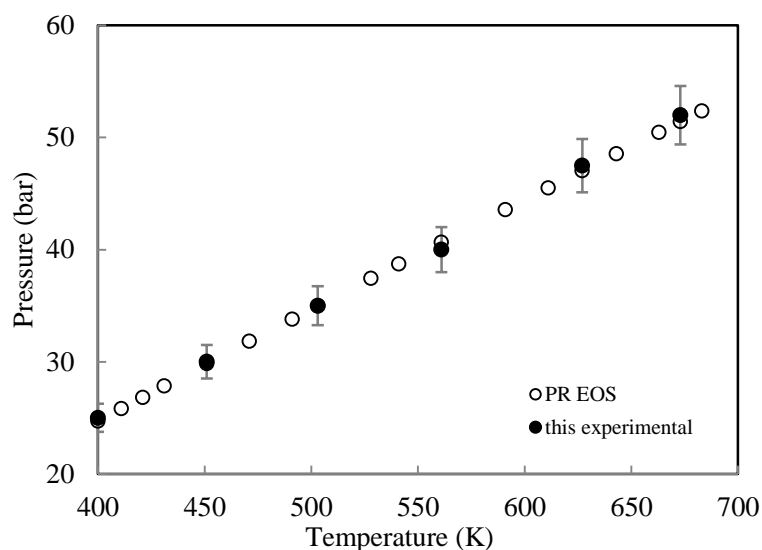


Fig. 4. Comparison of experimental data (○) for the ethyl acetate with predictions using the Peng-Robinson equation of state (PR EOS) and experimental (●).

### 3.2. The p-V-T Relationship of Ethyl Acetate-Palm Oil System

In the constant volume reactor, the relationship of pressure and temperature at constant global density or molar volume was investigated. The information during the process generates P-T diagram as shown in Fig. 5. The discontinuity of slope of the isochoric line indicates phase transition point [27].

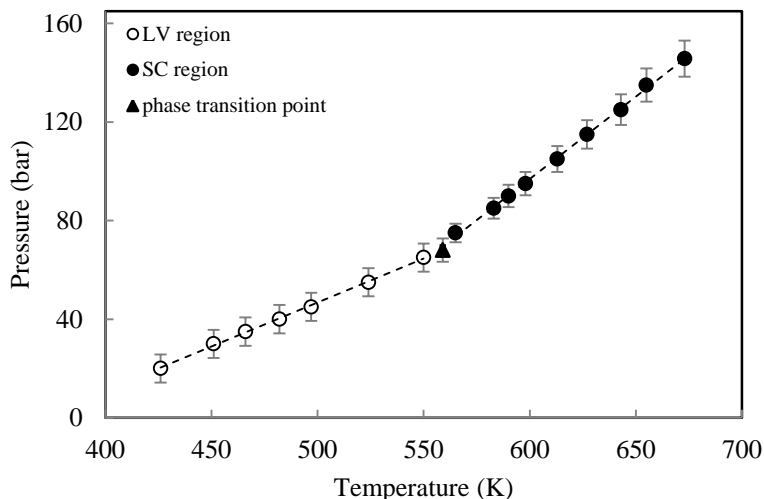


Fig. 5. Pressure vs. temperature for the reactive mixture of ethyl acetate:palm oil molar ratio of 20:1, global density = 0.44 g/cm<sup>3</sup>.

The palm oil is completely miscible in ethyl acetate at ambient condition. After temperature increased the vapor-liquid (LV) region of ethyl acetate-palm oil mixture changes to supercritical (SC) fluid. The pressure-temperature curve consists of two sections with separated linear isochoric lines. The lower pressure-temperature line corresponds to the LV region of the mixture. In this range, the liquid phase of ethyl acetate-palm oil mixture forms mutual solubility system while the vapor phase is ethyl acetate rich phase. It is to note that the isochoric line of the mixture has a lower slope comparing to the vapor pressure line of pure ethyl acetate (Fig. 4) due to the low volatility of palm oil [28]. In Fig. 5, when the phase transition point was reached, the LV phase was transformed into vapor and the system performs as a single-phase supercritical fluid. It could be deduced that the mixture in the single phase region was a monophasic dense gas; the appearance of the phase boundaries must be, however, confirmed by conducting experiments in a high temperature and pressure view-cell [29] but this technique has both strength and limitation. Visual observation at the boundary between phases may sometimes be obscured or may be poor or absent [30]. In Fig. 5, phase transition point of the mixture was observed at 553 K and 66 bar where it is above the critical point of pure ethyl acetate. The phase transition points of ethyl acetate-palm oil systems at different compositions and global densities were tabulated in Table 2.

The phase transition points in Table 2 were assumingly observed as critical points of the mixture at different compositions. The mixtures have lower critical temperature than that of palm oil and slightly higher than that of ethyl acetate (see Table 1). As the mixtures contain ethyl acetate in greater proportion than palm oil, the critical temperature of mixture is close to that of ethyl acetate rather than palm oil.

### 3.3. The Effect of Global Densities and Molar Ratio of Ethyl Acetate to Palm Oil

The P-T diagram of molar ratio of ethyl acetate: palm oil and global densities varied from 0.26 to 0.53 g/cm<sup>3</sup> in this study are shown in Fig. 6. The slope of P-T diagram slightly changes at the individual phase transition point corresponding to its global density, except at the global density of 0.26 g/cm<sup>3</sup>. In lower pressure (30-70 bar) and temperature (400-560 K) region, the mixture with a higher density generated a higher pressure. Within the SC region, the mixture that has higher global density simultaneously shifts the transition point to the lowered temperatures and pressures. The obtained results are in agreement with

Hegel et al. [6] who reported that the phase transitions in a dense liquid phase depend mainly on the reactant concentration and global density.

Table 2. Phase transition points of ethyl acetate-palm oil systems at various conditions.

Ethyl acetate:Oil Molar ratio	Global density (g/cm <sup>3</sup> )	Phase transition point	
		Temperature (K)	Pressure (bar)
10:1	0.26	–	–
	0.35	572	74
	0.44	570	72
	0.53	559	67
20:1	0.26	–	–
	0.35	558	68
	0.44	553	66
	0.53	549	61
30:1	0.26	–	–
	0.35	541	61
	0.44	537	59
	0.53	533	57

In the LV region in Fig. 6a – 6c (left hand side of the phase transition point), the pressure-temperature slopes for all densities are nearly identical. Konynenburg and Scott [31] described the phase behavior of a binary mixture that the critical point of the mixture was located at where the density and composition of the two phases are assembling. Therefore, the critical points of ethyl acetate-palm oil mixture at different molar ratios in this study were predicted as phase transition points. It can also be seen in Fig. 6a – 6c that at the density of 0.26 g/cm<sup>3</sup> of all the molar ratios of ethyl acetate to palm oil, there is no transition point, due to the density of the mixtures lower than the critical density of ethyl acetate (0.308 g/cm<sup>3</sup>).

Velez et al. [19] determined the phase transition of sunflower oil and ethanol mixture in supercritical condition using isochoric method. Comparing to the palm oil/ethyl acetate mixture, the phase transition points of the two systems at constant global density of 0.44 g/cm<sup>3</sup> are shown in Table 3. The phase transition point of the sunflower oil/ethanol mixture was significantly higher for temperature and pressure than this work. It can be presumed that the sunflower oil/ethanol mixture requires high temperature and pressure to transform heterogeneity towards homogeneity. It is to note that the higher molar ratio of ethanol to sunflower oil generates a rich vapour phase of ethanol, increasing the system temperature and pressure. In addition, the polarity of vegetable oil and reactant at room temperature affects the solubility of mixture and phase transition points in supercritical condition [32].

Table 3. Pressure and temperature at phase transition of a reacting system in supercritical reactor.

System	Molar ratio	Phase transition point		
		global density (g/cm <sup>3</sup> )	Temperature (K)	Pressure (bar)
Sunflower oil/ethanol [19]	1:40	0.44	588	110
Palm oil/ethyl acetate (this work)	1:30	0.44	553	66

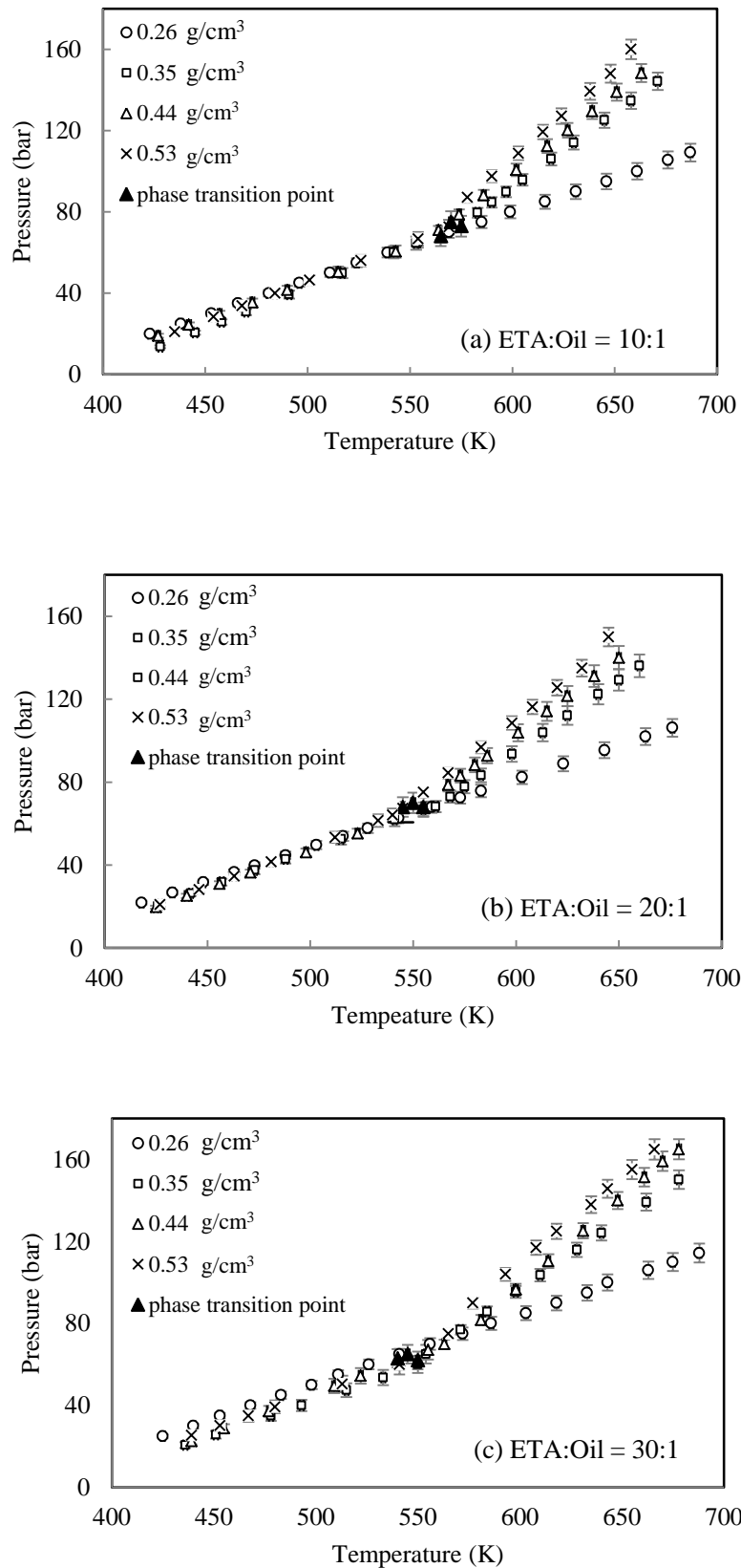


Fig. 6. Pressure vs. temperature of ethyl acetate and palm oil mixtures in molar ratio of (a) 10:1 (b) 20:1 and (c) 30:1 in different global densities (see Appendix for data).

#### 4. Conclusions

The density determination of ethyl acetate and palm oil system in supercritical condition has been accomplished in this work by indirect isochoric method. The molar ratio and global density of the mixture affected the LV region transformation to single phase of the mixture. The phase transition point from the LV region to SC region was also approximately located. The transition points were lowered by increasing global density and ethyl acetate: oil molar ratio. The subsequent data can be applied to find conditions of temperature and pressure that a homogeneous mixture of ethyl acetate and palm oil is optimized for producing biofuel in supercritical conditions.

#### Acknowledgment

The authors would like to thank the Program in Petrochemistry and Polymer Science of the Faculty of Science at Chulalongkorn University and the Ratchadaphiseksomphot Endowment Fund of Chulalongkorn University (RES560530071-EN) for financial support.

#### References

- [1] R. Sawangkeaw, K. Bunyakiat, and S. Ngamprasertsith, "A review of laboratory-scale research on lipid conversion to biodiesel with supercritical methanol (2001–2009)," *The Journal of Supercritical Fluids*, vol. 55, pp. 1–13, 2010.
- [2] K. Bunyakiat, S. Makmee, R. Sawangkeaw, and S. Ngamprasertsith, "Continuous Production of Biodiesel via Transesterification from Vegetable Oils in Supercritical Methanol," *Energy & Fuels*, vol. 20, pp. 812–817, 2006.
- [3] H. Sim Yeoh, G. Hean Chong, N. Mohd Azahan, R. Abdul Rahman, and T. S. Yaw Choong, "Solubility Measurement Method and Mathematical Modeling in Supercritical Fluids," *Engineering Journal*, vol. 17, pp. 67–78, 2013.
- [4] T. Fang, Wahyudiono, B. Al-Duri, Y. Shimoyama, Y. Iwai, M. Sasaki, *et al.*, "Supercritical Methanol Process of Modifying Oil Byproduct for Concentrating Natural Tocopherols," *Industrial & Engineering Chemistry Research*, vol. 46, pp. 5325–5332, 2007.
- [5] S. Glisic, O. Montoya, A. Orlovic, and D. Skala, "Vapor-liquid equilibria of triglycerides-methanol mixtures and their influence on the biodiesel synthesis under supercritical conditions of methanol," *Journal of the Serbian Chemical Society*, vol. 72, pp. 13–27, 2007.
- [6] P. Hegel, G. Mabe, S. Pereda, and E. A. Brignole, "Phase Transitions in a Biodiesel Reactor Using Supercritical Methanol," *Industrial & Engineering Chemistry Research*, vol. 46, pp. 6360–6365, 2007.
- [7] S. Saka and D. Kusdiana, "Biodiesel fuel from rapeseed oil as prepared in supercritical methanol," *Fuel*, vol. 80, pp. 225–231, 2001.
- [8] T. Pinnarat and P. E. Savage, "Assessment of Noncatalytic Biodiesel Synthesis Using Supercritical Reaction Conditions," *Industrial & Engineering Chemistry Research*, vol. 47, pp. 6801–6808, 2008.
- [9] S. Pereda, S. B. Bottini, and E. A. Brignole, "Supercritical fluids and phase behavior in heterogeneous gas-liquid catalytic reactions," *Applied Catalysis A: General*, vol. 281, pp. 129–137, 2005.
- [10] R. Sawangkeaw, K. Bunyakiat, and S. Ngamprasertsith, "Effect of co-solvents on production of biodiesel via transesterification in supercritical methanol," *Green Chemistry*, vol. 9, p. 679, 2007.
- [11] N. Jomtib, C. Prommuak, M. Goto, M. Sasaki, and A. Shotipruk, "Effect of Co-Solvents on Transesterification of Refined Palm Oil in Supercritical Methanol," *Engineering Journal*, vol. 15, pp. 49–58, 2011.
- [12] F. Goembira, K. Matsuura, and S. Saka, "Biodiesel production from rapeseed oil by various supercritical carboxylate esters," *Fuel*, vol. 97, pp. 373–378, 2012.
- [13] B. Jorgensen, S. Egholmchristiansen, M. Dahlthomsen, and C. Christensen, "Aerobic oxidation of aqueous ethanol using heterogeneous gold catalysts: Efficient routes to acetic acid and ethyl acetate," *Journal of Catalysis*, vol. 251, pp. 332–337, 2007.
- [14] A. Casas, J. R. n. Ruiz, M. a. J. s. Ramos, and A. n. Pérez, "Effects of Triacetin on Biodiesel Quality," *Energy & Fuels*, vol. 24, pp. 4481–4489, 2010.



- [15] I. G. Economou, "Statistical Associating Fluid Theory: A Successful Model for the Calculation of Thermodynamic and Phase Equilibrium Properties of Complex Fluid Mixtures," *Industrial & Engineering Chemistry Research*, vol. 41, pp. 953–962, 2002.
- [16] S.-S. You, K.-P. Yoo, and C. S. Lee, "An approximate nonrandom lattice theory of fluids," *Fluid Phase Equilibria*, vol. 93, pp. 193–213, 1994.
- [17] A. Fazlali, H. Modarress, and G. A. Monsoori, "Phase behavior prediction of complex petroleum fluids," *Fluid Phase Equilibria*, vol. 179, pp. 297–317, 2001.
- [18] A. Velez, P. Hegel, G. Mabe, and E. A. Brignole, "Density and Conversion in Biodiesel Production with Supercritical Methanol," *Industrial & Engineering Chemistry Research*, vol. 49, pp. 7666–7670, 2010.
- [19] A. Velez, S. Pereda, and E. A. Brignole, "Isochoric lines and determination of phase transitions in supercritical reactors," *The Journal of Supercritical Fluids*, vol. 55, pp. 643–647, 2010.
- [20] A. M. F. Palavra, M. A. Tavares Cardoso, J. A. P. Coelho, and M. F. B. Mourato, "Density Measurements of Fluids and Their Mixtures at High Pressure," *Chemical Engineering & Technology*, vol. 30, pp. 689–694, 2007.
- [21] M. Küster and B. Stijckhert, "Density changes of fluid inclusions in high-pressure low-temperature metamorphic rocks from crete: a thermobarometric approach based on the creep strength of the host minerals," *Lithos*, vol. 42, p. 17, 1997.
- [22] M. K. Modi, J. R. Reddy, B. V. Rao, and R. B. Prasad, "Lipase-mediated conversion of vegetable oils into biodiesel using ethyl acetate as acyl acceptor," *Bioresour Technol*, vol. 98, pp. 1260–4, Apr 2007.
- [23] I. De Marco, O. Knauer, F. Cice, A. Braeuer, and E. Reverchon, "Interactions of phase equilibria, jet fluid dynamics and mass transfer during supercritical antisolvent micronization: The influence of solvents," *Chemical Engineering Journal*, vol. 203, pp. 71–80, 2012.
- [24] D. Y. Peng and D. B. Robinson, "A new two-constant equation of state," *Industrial Engineering Chemistry Fundamentals*, vol. 15, pp. 59–64, 1976.
- [25] S.-A. Hong, J.-D. Kim, J. Kim, J. W. Kang, and I.-J. Kang, "Phase equilibria of palm oil, palm kernel oil, and oleic acid+supercritical carbon dioxide and modeling using Peng–Robinson EOS," *Journal of Industrial and Engineering Chemistry*, vol. 16, pp. 859–865, 2010.
- [26] T. Hu, Z. Qin, G. Wang, X. Hou, and J. Wang, "Critical Properties of the Reacting Mixture in the Esterification of Acetic Acid with Ethanol," *Journal of Chemical & Engineering Data*, vol. 49, pp. 1809–1814, 2004.
- [27] M. Atilhan, J. Zhou, S. Ejaz, D. Cristancho, J. Holste, and K. R. Hall, "Phase Behavior Concerns for Multicomponent Natural Gas-Like Mixtures," in *Proceedings of the 1st Annual Gas Processing Symposium*, 2009.
- [28] L. J. Florusse, T. Fornari, S. B. Bottini, and C. J. Peters, "Phase behavior of carbon dioxide—low-molecular weight triglycerides binary systems: measurements and thermodynamic modeling," *The Journal of Supercritical Fluids*, vol. 31, pp. 123–132, 2004.
- [29] R. M. Oag, P. J. King, C. J. Mellor, M. W. George, J. Ke, M. Poliakoff, V. K. Popov, and V. N. Bagratashvili, "Determining phase boundaries and vapour/liquid critical points in supercritical fluids: a multi-technique approach," *The Journal of Supercritical Fluids*, vol. 30, pp. 259–272, 2004.
- [30] W. Mayer, S. Hoffmann, G. Meier, and I. Alig, "Critical fluctuations in a binary mixture of polyethylene glycol and polypropylene glycol studied by ultrasonic and light scattering experiments," *Physical Review E*, vol. 55, pp. 3102–3110, 1997.
- [31] P. H. V. Konynenburg and R. L. Scott, "Critical Lines and Phase Equilibria in Binary Van Der Waals Mixtures," *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 298, pp. 495–540, 1980.
- [32] J. M. Bernal, P. Lozano, E. Garcia-Verdugo, M. I. Burguete, G. Sanchez-Gomez, G. Lopez-Lopez, M. Pucheault, M. Vaultier, and S. V. Luis, "Supercritical synthesis of biodiesel," *Molecules*, vol. 17, pp. 8696–719, 2012.

## Appendix

Table A1. The experimental average data for temperature and pressure at molar ratio of ethyl acetate to palm oil of 10:1 and various global densities.

Density of palm oil/ethyl acetate (g/cm <sup>3</sup> )							
0.26		0.35		0.44		0.53	
T	P	T	P	T	P	T	P
(K)	(bar)	(K)	(bar)	(K)	(bar)	(K)	(bar)
423	20.0	445	20.6	427	20.0	435	21.1
438	25.0	458	25.9	442	24.4	454	28.4
453	30.0	470	30.8	457	29.8	468	33.8
466	35.0	491	39.3	473	35.5	484	39.9
481	40.0	517	49.9	490	41.6	501	46.4
496	45.0	542	60.1	515	50.5	526	56.0
511	50.0	553	64.6	543	60.5	554	66.8
524	55.0	565	69.5	565	68.4	569	72.5
539	60.0	583	79.6	564	71.0	578	87.1
554	65.0	590	84.7	574	78.8	590	97.6
569	70.0	597	89.9	586	88.2	603	108.9
585	75.0	605	95.8	602	100.7	615	119.3
599	80.0	619	106.1	617	112.4	624	127.1
616	85.0	630	114.1	627	120.2	638	139.3
631	90.0	645	125.2	639	129.6	648	148.0
646	95.0	658	134.7	651	139.0	658	156.7
661	100.0	671	144.3	663	148.3	658	160.0

Table A2. The experimental average data for temperature and pressure at molar ratio of ethyl acetate to palm oil of 20:1 and various global densities.

Density of palm oil-ethyl acetate (g/cm <sup>3</sup> )							
0.26		0.35		0.44		0.53	
T	P	T	P	T	P	T	P
(K)	(bar)	(K)	(bar)	(K)	(bar)	(K)	(bar)
418	21.8	442	26.4	425	20.0	427	20.9
433	26.7	457	31.8	440	25.1	446	28.1
448	31.6	473	37.5	456	30.9	463	34.6
463	36.5	488	42.9	471	36.4	481	41.5
473	39.8	515	52.5	498	46.2	512	53.4
488	44.7	541	61.9	523	55.3	533	61.4
503	49.6	555	66.9	538	60.7	540	64.1
516	53.9	561	68.2	567	78.7	545	67.3
528	57.8	568	73.0	573	83.2	555	75.1
543	62.7	575	77.8	580	88.3	567	84.4
559	67.9	583	83.3	586	92.8	583	96.9
573	72.5	598	93.6	601	103.8	598	108.5
583	75.8	613	103.9	615	114.2	608	116.3
603	82.3	625	112.1	625	121.6	620	125.6
623	88.9	640	122.4	638	131.2	632	134.9
643	95.4	650	129.3	650	140.0	645	150.0

Table A3. The experimental average data for temperature and pressure at molar ratio of ethyl acetate to palm oil of 30:1 and various global densities.

Density of palm oil-ethyl acetate (g/cm <sup>3</sup> )							
0.26		0.35		0.44		0.53	
T	P	T	P	T	P	T	P
(K)	(bar)	(K)	(bar)	(K)	(bar)	(K)	(bar)
410	20.0	436	20.6	439	20.0	423	22.6
425	25.0	451	25.7	455	25.4	439	28.8
440	30.0	478	34.9	477	30.1	453	37.2
453	35.0	493	40.0	509	34.9	467	49.5
468	40.0	515	47.5	522	39.3	480	54.5
483	45.0	533	53.6	541	50.5	513	61.8
498	50.0	550	60.0	555	60.0	541	67.2
511	55.0	565	75.0	563	75.0	565	70.0
526	60.0	577	90.0	581	90.0	577	81.7
541	65.0	593	104.0	598	104.0	593	96.5
556	70.0	608	117.0	614	117.0	608	110.4
572	75.0	618	125.0	631	125.0	618	125.2
586	80.0	635	138.0	648	138.0	635	140.0
603	85.0	643	145.7	661	145.7	643	151.3
618	90.0	655	155.1	670	159.1	655	155.1
633	95.0	666	164.9	678	165.0	666	164.9

