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Phase equilibrium data on binary and ternary mixtures of methyl palmitate, hydrogen and propane

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Abstract

The hydrogenolysis of fatty acid methyl esters (FAME) to obtain fatty alcohols, is a catalytic, multiphase reaction. Reaction rates can be greatly increased by using a supercritical solvent to bring the reactive mixture into a single homogeneous phase. Knowledge of the corresponding phase equilibria is a prerequisite in order to find the homogeneous region and to determine the most favorable conditions for the hydrogenation process. This paper reports experimental phase equilibrium data on binary and ternary mixtures of methyl palmitate, hydrogen and propane. A temperature region between 360 and 450 K, and pressures up to 15 MPa were covered.
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Keywords: Supercritical hydrogenolysis; Phase equilibria; Methyl palmitate; Hydrogen; Propane

1. Introduction

Fatty alcohols (FOH) are normally produced by hydrogenolysis of fatty acid methyl esters (FAME), in a catalytic multiphase reaction. The yield of FOH and the reaction rate depend of the hydrogen pressure in the reactor [1]. On an industrial scale, the high-pressure catalytic hydrogenation is carried out in a trickle-bed or a slurry-phase reactor. A copper-based catalyst is commonly used and temperatures ranging from 523 up to 573 K and pressures between 20 and 30 MPa are the normal operating conditions [2]. A large excess of hydrogen (20–100 mol H_2 per mol of ester, according to Buchold [3] and Kreutzer [4]) and high pressures are required to overcome the problem of low hydrogen (H_2) solubility in the reaction mixture. This low solubility introduces mass transport resistance in the liquid phase and limits the availability of H_2 at the catalyst surface; i.e., the reaction rate is diffusion controlled. The low concentration of H_2 at the catalyst surface does not only have the consequence of producing relatively low reaction rates, but it also favors partial isomerization of the *cis* double-bonds into the *trans* configuration [5,6].

A way to increase the concentration of H_2 at the catalyst surface is to introduce a supercritical solvent into the reaction mixture. The role of the supercritical fluid is to bring both, the gas and the

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Table 1
Vapor–liquid isopleths of the system methyl palmitate (1)–hydrogen (2)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
$x_2 = 0.0495$		$x_2 = 0.0794$		$x_2 = 0.1028$		$x_2 = 0.1284$	
L + V → L		L + V → L		L + V → L		L + V → L	
450.26	3.506	449.84	5.830	450.33	7.773	450.08	10.010
445.32	3.576	445.13	5.935	445.40	7.899	444.73	10.190
440.36	3.636	439.91	6.035	440.43	8.044	440.33	10.365
435.44	3.706	435.37	6.155	435.47	8.194	435.29	10.540
430.51	3.781	430.23	6.265	430.54	8.349	430.40	10.740
425.54	3.851	425.37	6.380	425.56	8.514	425.44	10.920
420.61	3.926	420.20	6.515	420.60	8.679	420.45	11.130
413.19	4.041	412.96	6.705	413.19	8.934	412.95	11.456
405.73	4.166	406.05	6.905	405.76	9.229	405.86	11.801
398.31	4.301	398.40	7.130	398.33	9.524	398.24	12.181
390.92	4.441	390.84	7.360	390.88	9.839	390.89	12.581
383.44	4.596	383.44	7.595	383.44	10.164	383.42	12.991
376.06	4.751	376.08	7.850	376.02	10.514	376.12	13.431
368.59	4.911	368.72	8.140	368.69	10.884	368.71	13.916
361.23	5.086	361.36	8.430	361.27	11.274	363.77	14.261

x_2, represents mole fraction of hydrogen.

substrate, into a homogeneous single phase in which the transport resistance for hydrogen is minimized. As a consequence, the effective concentration of H_2 at the catalyst surface is significantly enhanced, leading to extremely high reaction rates compared to the traditional two-phase gas–liquid approach [6,7].

For apparent reasons, the fluid mostly considered for supercritical processes is carbon dioxide (CO_2), since it is non-toxic, inert, cheap, etc. However, CO_2 is not a good solvent for heavy compounds. For methyl palmitate and components with similar chain lengths attached to it (i.e., alkanes, fatty acids, fatty acids ethyl esters, etc.), CO_2 gives a phase split up to very high pressures, whereas propane shows complete miscibility, even for heavier compounds like triglycerides, under similar conditions [8–14].

In addition to that, hydrogenolysis process conditions are likely to induce the hydrogenation of carbon dioxide towards the formation of methanol, which is considered to be an undesirable side product. Furthermore, the methanol synthesis reaction is highly exothermic, posing a potential runaway hazard [15]. Therefore, investigations for substitutes of CO_2, with emphasis on lower alkanes (ethane to hexane), are currently underway [16,17].

The use of propane as a supercritical solvent for the homogeneous hydrogenation of fatty acid methyl esters has been investigated by Van den Hark and co-workers [18–20], Macher et al. [21] and Andersson et al. [22]. Reaction rates 500 times higher than those obtained in the traditional two-phase process have been reported [21].

Supercritical hydrogenation is demonstrably effective and is potentially attractive, with significant advantages over conventional methods. However, a major question is whether at the conditions of the hydrogenation process a single phase is really present. Therefore, it is essential to know the phase behavior of the reactive system in the region of interest in terms of pressure, temperature and composition; especially

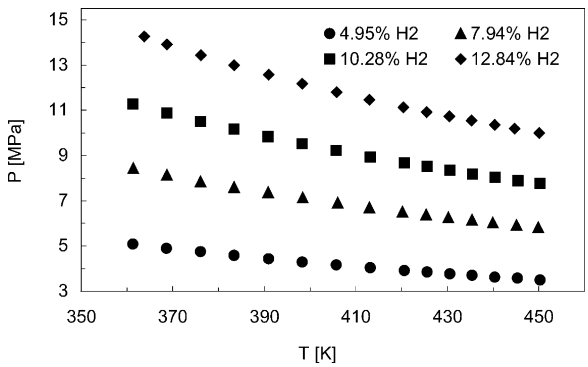

Fig. 1. Solubility of hydrogen in methyl palmitate, given as a pressure–temperature diagram at constant H_2 composition (mole %).

Table 2
Vapor–liquid isopleths of the system methyl palmitate (1)–propane (2)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
$x_2 = 0.9655$		$x_2 = 0.9219$		$x_2 = 0.8011$	
L + V → V		L + V → V		L + V → V	
450.22	9.541	450.55	9.778	450.63	8.355
447.77	9.436	440.66	9.218	440.68	7.755
445.30	9.321	430.76	8.578	430.76	7.135
442.81	9.201	420.81	7.868	420.83	6.485
440.26	9.076	410.90	7.088	410.92	5.850
437.84	8.946	401.00	6.278	401.03	5.215
435.39	8.811	391.07	5.468	391.12	4.595
432.92	8.671	381.18	4.708	381.24	4.015
430.47	8.526	371.27	4.018	371.42	3.475
425.49	8.221	361.40	3.413	361.50	2.985
420.53	7.891	351.56	2.883	351.66	2.540
415.61	7.546	341.72	2.413	341.80	2.145
410.66	7.186	331.86	2.003	331.92	1.795
408.16	6.996	321.99	1.648	322.03	1.490
405.70	6.811	312.11	1.358	312.17	1.225
403.24	6.621				
403.00	6.601				
402.84	6.586				
L = V					
402.71	6.580				
L + V → L					
402.52	6.566	$x_1 = 0.6537$			
		L + V → L			
402.25	6.541	450.46	6.227		
401.72	6.501	440.51	5.827		
400.75	6.426	430.60	5.432		
398.24	6.231	420.69	5.007		
395.80	6.036	410.76	4.592		
393.32	5.841	400.84	4.192		
390.85	5.646	391.01	3.722		
385.88	5.247	381.10	3.367		
380.90	4.862	371.18	3.002		
376.00	4.502	361.33	2.662		
371.11	4.162	351.55	2.342		
366.19	3.837	341.69	2.057		
361.27	3.527	331.83	1.797		
356.36	3.242				
351.46	2.972				
346.53	2.717				
341.59	2.482				
336.65	2.257				
331.72	2.047				
326.79	1.857				
321.87	1.702				

x_2, represents mole fraction of propane.

the region where the single phase occurs should be accurately known.

To the best of our knowledge, no experimental data are available in the literature on the phase behavior of mixtures of interest for the supercritical hydrogenation of fatty acid methyl esters. To fill this gap, experimental phase equilibrium data have been measured on binary and ternary mixtures composed of supercritical propane, hydrogen and methyl palmitate. This paper reports the experimental results.

2. Experimental

All measurements were carried out in a Cailletet apparatus, which operates according to the synthetic method. A sample of known overall composition is brought under vacuum conditions into the closed top of a thick-walled Pyrex glass tube. After dosing the chemicals, the open end of the high-pressure tube is sealed with mercury, again under vacuum conditions. Then, the open bottom-end of the tube is immersed in mercury, contained in an autoclave. The autoclave is connected to a high-pressure hydraulic system applying oil as the pressure medium. Mercury is also used as the pressure-transmitting fluid between the hydraulic oil and the sample. The pressure inside the tube can be modified by means of a hand screw pump, while the pressure is measured with a dead weight pressure balance. Pressure measurements are as accurate as 0.03% of the reading. The Cailletet tube is jacketed,

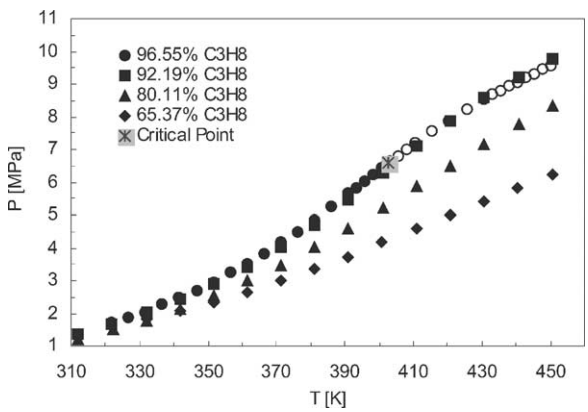

Fig. 2. Vapor–liquid equilibria of the binary system MP + C_3H_8 (filled symbols: bubble points; open symbols: dew points). Concentrations are given in mole % of propane.

and silicon oil is used as the thermostatic fluid. A platinum resistance thermometer (Pt 100), located as close as possible to the mixture under investigation, measures the temperature, which is controlled within ± 0.02 K by a thermostat. The sample inside the tube is stirred by a stainless steel ball, which is moved up and down by two button magnets activated by a rotating disc mounted on a stirring motor.

A range of temperatures from 312 to 450 K (maximum operating temperature of the apparatus) and pressures between 2.5 up to 15 MPa were covered. The latter pressure is the upper limit allowed for the Pyrex glass Cailletet tube.

Table 3
Vapor–liquid isopleths of the ternary system methyl palmitate (1) + hydrogen (2) + propane (3)

T (K)	p (MPa)	T (K)	p (MPa)
$x_2 = 0.0311$, $x_3 = 0.9356$		$x_2 = 0.0614$, $x_3 = 0.9062$	
L + V → V		L + V → V	
450.26	10.586	450.21	11.640
447.82	10.496	447.76	11.560
445.35	10.401	445.27	11.475
442.83	10.291	442.85	11.385
440.37	10.176	440.38	11.290
437.89	10.056	437.92	11.185
435.43	9.937	435.42	11.080
432.94	9.807	432.98	10.970
430.47	9.677	430.50	10.855
428.02	9.537	428.00	10.735
425.54	9.397	425.55	10.610
423.02	9.247	423.09	10.480
420.57	9.097	420.58	10.345
418.10	8.942	418.12	10.210
415.61	8.782	415.68	10.070
413.13	8.617	413.15	9.920
410.63	8.452	410.70	9.780
408.17	8.287	408.22	9.625
405.68	8.117	405.76	9.485
403.89	7.991	404.42	9.400
403.57	7.969	403.30	9.335
L = V		L = V	
403.22	7.95	402.99	9.317
L + V → L		L + V → L	
402.85	7.921	402.77	9.310
402.51	7.899	402.28	9.280
400.73	7.781	400.82	9.200
398.28	7.621	398.34	9.065
395.79	7.466	395.85	8.940
393.35	7.306	393.36	8.820
390.87	7.156	390.92	8.705

Effect of hydrogen addition. Compositions x are mole fractions.

Samples were prepared by dosing gravimetrically the solid component (methyl palmitate) into the Cailletet tube. Then, gases (hydrogen and/or propane) were dosed volumetrically into the tube via a gas-dosing device. For further details on the experimental facility one is referred to Raeissi and Peters [23].

For each measurement the temperature was fixed and the pressure was varied (increased or decreased stepwise) until a phase change was observed visually. Bubble and dew points are obtained as the points at which the last bubble of vapor or the last droplet of liquid disappears, respectively. Critical points are determined visually using a number of criteria, including critical opalescence and the equality of volumes of the two phases at the critical point. For a given sample, the whole procedure is repeated at several temperatures to obtain the equilibrium curve of that particular isopleth.

Aldrich supplied methyl palmitate (purity better than 99%). Propane (purity better than 99.95% molar) was obtained from Scott Specialty Gases and hydrogen (purity of 99.999% molar) from Hoek Loos. All chemicals were used without any further purification.

3. Results and discussion

3.1. Binary systems

In this work, the solubility of hydrogen (H_2) in methyl palmitate (MP) was measured. Table 1 reports

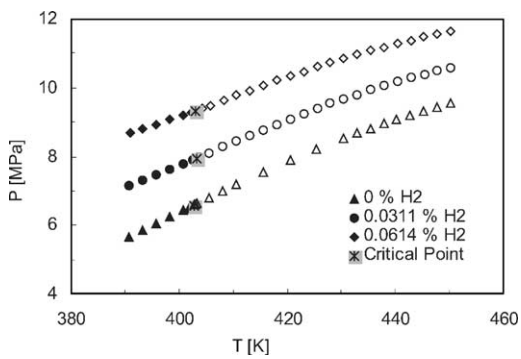

Fig. 3. Effect of hydrogen on the phase behaviour of two ternary isopleths with a fixed MP/C_3H_8 ratio of 0.036. For comparison, the isopleth of the binary system C_3H_8 + MP with the same MP/C_3H_8 ratio has been included (filled symbols: bubble points; open symbols: dew points). Concentrations are given in mole % of H_2.

Table 4
Vapor–liquid isopleths of the ternary system methyl palmitate (1) + hydrogen (2) + propane (3)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
$x_2 = 0.0734$, $x_3 = 0.2850$		$x_2 = 0.0506$, $x_3 = 0.5065$		$x_2 = 0.0230$, $x_3 = 0.7719$	
L + V → L		L + V → L		L + V → L	
450.25	8.053	450.28	8.654	450.23	9.798
445.34	8.083	445.32	8.589	445.27	9.543
440.38	8.128	440.38	8.524	440.33	9.283
435.44	8.173	435.41	8.454	435.39	9.023
430.51	8.228	430.51	8.389	430.45	8.753
425.52	8.278	425.54	8.329	425.48	8.488
420.59	8.333	420.58	8.260	420.55	8.213
413.14	8.433	413.15	8.180	413.15	7.808
405.67	8.538	405.72	8.105	405.68	7.403
398.28	8.653	398.31	8.035	398.27	7.013
390.84	8.788	390.86	7.990	390.85	6.633
383.45	8.921	383.45	7.945	383.42	6.273
376.06	9.086	376.05	7.920	376.05	5.943
368.68	9.271	368.65	7.915	368.61	5.638
361.31	9.466	361.27	7.930	361.25	5.358

Effect of propane addition. Compositions x are mole fractions.

the vapor–liquid equilibrium conditions obtained as bubble point (L + V → L) transitions, for four different isopleths of low hydrogen concentration. The data show the typical behavior of hydrogen solubility; i.e. the solubility increases with temperature (see Fig. 1). The low solubility of H_2 in the liquid phase produces a large increase in the bubble pressure of the mixture, after a small increase in the mole fraction of hydrogen.

The binary system propane (C_3H_8) + methyl palmitate (MP) was also studied. Table 2 contains the results obtained for four different isopleths and Fig. 2 shows the results graphically. For the isopleth with 96.55 mol % of propane, not only bubble points (L + V → L), but also dew (L + V → V) and critical points (L = V) were determined.

3.2. Ternary systems

Table 3 contains the bubble, dew and critical points measured for two ternary isopleths having a fixed

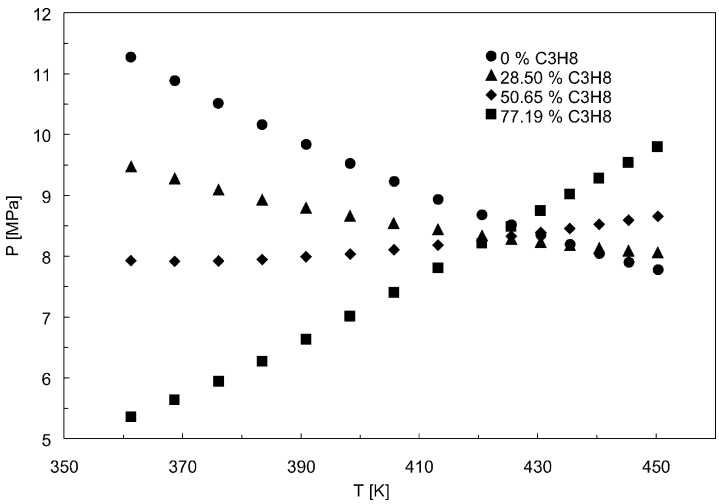

Fig. 4. Effect of propane on the phase behavior of the ternary system $H_2 + C_3H_8$ + MP. Concentrations are given in mole % of propane.

Table 5
Vapor–liquid isopleths of the ternary system methyl palmitate (1) + hydrogen (2) + propane (3)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
$x_1 = 0.0745$, $x_3 = 0.8515$		$x_1 = 0.0668$, $x_3 = 0.8696$		$x_1 = 0.0551$, $x_3 = 0.8918$	
L + V → L		L + V → L		L + V → L	
455.29	13.531	455.29	12.891	450.23	11.987
452.80	13.471	454.33	12.851	447.82	11.897
450.18	13.421	453.80	12.826	445.31	11.797
447.78	13.351			442.87	11.692
445.39	13.281	L = V		440.38	11.587
442.89	13.206	453.52	12.816	437.95	11.482
440.40	13.126			436.90	11.432
437.92	13.046	L + V → L		436.44	11.412
435.47	12.961	453.26	12.811		
433.00	12.871	452.80	12.796	L = V	
430.54	12.776	452.28	12.781	436.26	11.402
428.06	12.681	451.85	12.761		
425.57	12.576	450.26	12.726	L + V →L	
423.08	12.476	447.83	12.641	435.92	11.387
420.60	12.366	448.05	12.636	435.46	11.367
418.12	12.256	445.29	12.546	432.94	11.252
415.66	12.146	442.88	12.456	430.48	11.137
413.20	12.031	440.43	12.366	427.96	11.012
410.73	11.911	437.94	12.276	425.56	10.897
408.22	11.791	435.47	12.181	423.12	10.772
405.75	11.666	433.01	12.081	420.56	10.642
403.25	11.546	430.50	11.976	415.59	10.382
400.81	11.421	428.04	11.871	410.66	10.117
398.33	11.296	425.59	11.761	405.73	9.837
395.81	11.176	423.08	11.646	400.79	9.547
393.38	11.056	420.60	11.531	395.78	9.257
390.91	10.936	415.66	11.286	390.89	8.977
		410.64	11.031		
		405.72	10.771		
		400.76	10.501		
		395.81	10.236		
		390.87	9.976		
$x_1 = 0.0471$, $x_3 = 0.9084$		$x_1 = 0.0334$, $x_3 = 0.9356$			
L + V → V		L + V → V			
450.26	11.365	450.26	10.586		
445.32	11.155	447.82	10.496		
440.31	10.930	445.35	10.401		
435.41	10.695	442.83	10.291		
432.95	10.570	440.37	10.176		
430.55	10.445	437.89	10.056		
428.01	10.305	435.43	9.937		
425.47	10.170	432.94	9.807		
424.24	10.100	430.47	9.677		
423.03	10.035	428.02	9.537		
422.55	10.005	425.54	9.397		
422.08	9.975	423.02	9.247		
421.56	9.945	420.57	9.097		
421.08	9.917	418.10	8.942		
420.77	9.897	415.61	8.782		
420.57	9.887	413.13	8.617		
		410.63	8.452		

Table 5 (*Continued*)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
L = V		408.17	8.287		
420.46	9.881	405.68	8.117		
		403.89	7.991		
L + V → L		403.57	7.969		
420.36	9.874				
420.26	9.869	L = V			
420.06	9.860	403.22	7.944		
419.57	9.830				
419.05	9.805	L + V → L			
418.58	9.775	402.85	7.921		
418.10	9.764	402.51	7.899		
415.63	9.604	400.73	7.781		
413.10	9.459	398.28	7.621		
410.65	9.314	395.79	7.466		
405.70	9.019	393.35	7.306		
400.76	8.719	390.87	7.156		
395.81	8.419				
390.87	8.119				

Ratio H_2:MP = 1. Compositions x are mole fractions.

Table 6
Vapor–liquid isopleths of the ternary system methyl palmitate (1) + hydrogen (2) + propane (3)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
$x_1 = 0.0342$, $x_3 = 0.8269$		$x_1 = 0.0287$, $x_3 = 0.8516$		$x_1 = 0.0247$, $x_3 = 0.8760$	
L + V → V		L + V → V		L + V → V	
425.60	14.343	450.60	13.935	450.61	12.669
420.70	14.183	445.69	13.785	445.67	12.564
415.73	14.013	440.73	13.665	440.68	12.439
410.75	13.818	435.74	13.535	435.74	12.294
405.84	13.638	430.72	13.375	430.79	12.124
400.88	13.378	425.82	13.205	425.81	11.969
400.36	13.353	420.85	13.020	420.86	11.750
399.97	13.343	415.91	12.815	415.90	11.525
		410.94	12.600	410.93	11.280
L = V		405.99	12.380	405.94	11.025
399.45	13.323	401.04	12.160	401.03	10.765
		396.18	11.965	396.06	10.510
L + V → L		396.08	11.960	393.63	10.385
398.90	13.313			392.59	10.350
395.93	13.288	L = V			
391.00	13.263	396.00	11.960	L = V	
386.05	13.243			392.41	10.340
381.07	13.258	L + V → L			
376.19	13.318	395.88	11.955	L + V → L	
371.23	13.413	395.62	11.945	392.29	10.340
366.29	13.553	393.63	11.875	392.05	10.335
361.37	13.718	391.14	11.835	391.62	10.315
356.41	13.913	388.65	11.800	386.17	10.180
351.45	14.143	386.19	11.775	381.25	10.130
346.55	14.393	381.24	11.770	376.28	10.120

Table 6 (*Continued*)

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
		376.31	11.800	371.33	10.140
		371.37	11.870	366.39	10.175
		366.42	11.970	361.44	10.235
		361.47	12.100	351.56	10.415
		351.58	12.420	341.74	10.660
		341.78	12.825	331.9	10.980
		331.93	13.320	322.01	11.385
		322.04	13.910		
		317.11	14.245		
$x_1 = 0.0199$, $x_3 = 0.9006$		$x_1 = 0.0149$, $x_3 = 0.9251$			
L + V → V		L + V → V			
450.56	11.421	450.61	10.376		
445.62	11.286	445.65	10.296		
440.66	11.156	440.65	10.151		
435.70	11.001	435.67	9.986		
430.76	10.826	430.70	9.821		
425.79	10.626	425.73	9.631		
420.84	10.411	420.82	9.416		
415.88	10.171	415.86	9.180		
410.91	9.911	410.91	8.920		
405.97	9.636	405.50	8.635		
400.99	9.341	401.02	8.340		
396.05	9.050	396.06	8.030		
391.13	8.775	391.13	7.720		
389.63	8.700	386.19	7.450		
389.46	8.690	386.01	7.440		
389.15	8.680				
L = V		L = V			
389.05	8.675	385.87	7.435		
L + V → L		L + V → L			
388.93	8.670	385.77	7.430		
388.16	8.640	385.68	7.425		
386.17	8.580	381.25	7.310		
385.19	8.560	376.29	7.235		
384.20	8.540	371.36	7.165		
383.22	8.520	366.40	7.095		
382.22	8.505	361.47	7.040		
381.24	8.495	351.64	6.945		
380.75	8.490	341.78	6.895		
379.75	8.475	331.90	6.910		
378.76	8.465	322.03	6.990		
376.30	8.450	312.17	7.130		
373.83	8.430				
371.33	8.420				
368.90	8.410				
366.40	8.400				
361.44	8.425				
351.61	8.460				
341.78	8.550				
331.88	8.710				
322.00	8.940				
312.15	9.255				

Ratio H_2:MP = 4. Compositions x are mole fractions.

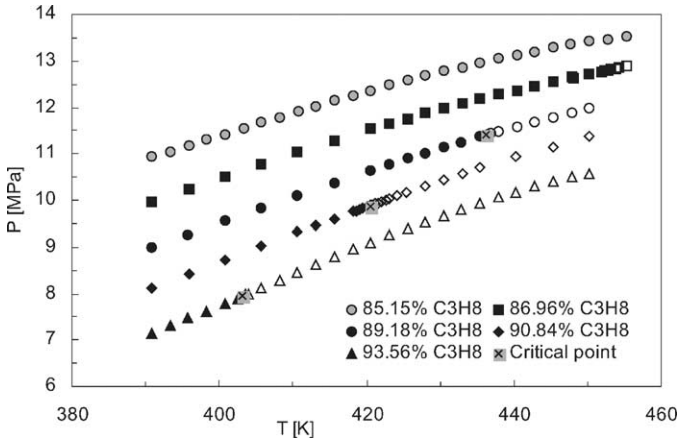

Fig. 5. Experimental phase equilibria of the ternary system $H_2 + C_3H_8 + MP$ at a H_2:MP ratio = 1 (filled dots: bubble points; open dots: dew points). Concentrations are given in mole % of propane.

MP/C_3H_8 ratio equal to 0.036. Both ternary isopleths, along with the binary isopleth of the system $MP + C_3H_8$ with the same mixing ratio, are shown in Fig. 3. This figure shows the increase in pressure produced by the addition of hydrogen to the mixture.

In order to see the effect of propane on the phase behavior of the reactive mixture, ternary isopleths on the $MP + H_2 + C_3H_8$ system were measured, increasing the amount of propane in the mixture from 0 to 77 mol %, while keeping the MP/H_2 ratio at a fixed value of 8.7. The measured bubble pressures of different isopleths are reported in Table 4 and shown in Fig. 4. From this figure it becomes clear that isopleths with low concentration of C_3H_8 in the liquid phase have a negative slope in a pressure versus temperature diagram (hydrogen-like behavior); i.e. they show the typical solubility behavior of gases at highly reduced temperatures. On the other hand, at higher C_3H_8 concentrations the slope of the isopleths change sign and become positive (propane-like behavior). From Fig. 4, it can also be observed that at propane concentrations close to 50% molar, the influence of the temperature on the equilibrium pressure becomes almost negligible.

Also ternary systems with high propane concentrations ($\geq$75 mol %) were measured. Previous research by Van den Hark and Härröd [20] and Macher et al. [21], along with predictions by Pereda et al. [24], suggests that high propane concentrations are needed in the supercritical hydrogenation process, to achieve single-phase conditions at the required ratio of the reaction components.

When the H_2 concentration in the ternary mixture is increased, it acts as an anti-solvent, rising the equilibrium pressures of the entire system. This phenomenon may reduce the solubility of methyl palmitate in the supercritical media. To quantify this effect, measurements have been carried out in ternary mixtures of $H_2 + MP + C_3H_8$ having fixed ratios of H_2 to MP and

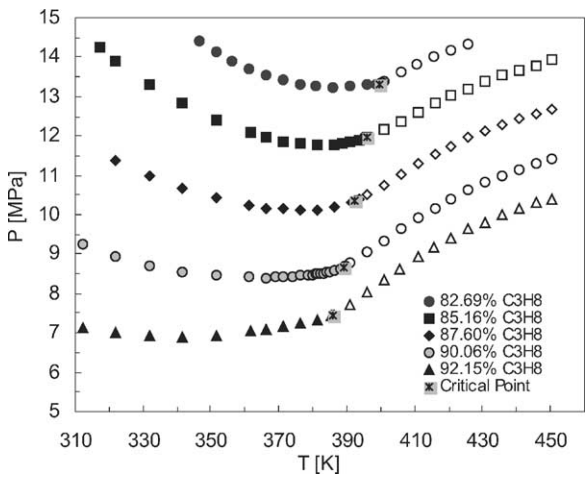

Fig. 6. Experimental phase equilibria of the ternary system $H_2 + C_3H_8 + MP$ at a H_2:MP ratio = 4 (filled dots: bubble points; open dots: dew points). Concentrations are given in mole % of propane.

variable propane concentrations, which makes a systematic comparison of the results possible.

The results are given in Tables 5 and 6 and depicted in Figs. 5 and 6. It becomes apparent that not only with increasing propane concentration the solubility of the reaction mixture (H_2 + MP) increases. Fig. 6 also shows that for higher concentrations of hydrogen (i.e. a higher H_2:MP ratio), the slopes of the isopleths change sign at lower temperatures; i.e. an anti-solvent effect occurs towards lower propane concentrations. In both ternary systems mixture critical points could be established, as represented in Figs. 5 and 6. Both critical loci proceed towards the critical point of pure propane.

4. Conclusions

Experimental phase equilibrium data were obtained for the binary systems H_2 + MP and C_3H_8 + MP and the ternary H_2 + C_3H_8 + MP. Not only bubble points but also dew and critical points could be determined for some of the isopleths. In general, it can be concluded that increasing propane concentration causes increasing solubilities of the reaction mixture H_2 + MP, which means that propane could be an adequate solvent for the homogeneous hydrogenation of fatty acid methyl esters.

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