J. Chem. Eng. Data **2003,** *48*, 571−575

Fluid Phase Equilibria of Binary *n*-Alkane + Squalane Systems

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In this study, the phase behavior of binary systems of squalane ($C_{30}H_{62}$, 2,6,10,15,19,23-hexamethyltetracosane) with ethane, and propane, is investigated. For both systems, vapor−liquid equilibrium data were measured using the synthetic method in the temperature range (263 to 473) K and pressure range (0.2 to 16) MPa. Also a liquid−liquid-vapor curve was determined for the ethane + squalane system. The experiments were carried out for various mixtures covering almost the whole composition range. Additionally, the experimental data have been modeled using the Soave−Redlich−Kwong equation of state using two adjustable parameters in the classical mixing rules.

Introduction

Phase behavior measurements for binary systems of short and long paraffins are very important both for practical process design and for developing theoretical models. For example, these systems can be considered as models for mixtures of oligomers and polymers in low-molar-mass solvents that are of interest for the design of dewaxers in a polymer plant. Such data can also be used to obtain the parameters for different types of thermodynamic models such as equations of state, group contribution methods and molecular simulation calculations, models that can be used in further predictions.

Although systems of short and long chain normal alkanes have been extensively studied, very little information has been published for systems with branched long-chain alkanes. Paas et al.[1] have reported some work on binary systems of squalane with methane and ethane. However, their work focused only on mixtures with high concentration of ethane (more than 0.9 in mole fraction) and low and very low temperatures. Recently, Aalto et al.[2] have reported some bubble point pressures and densities for the binary propane + squalane in the vicinity of the critical point of propane. In this paper, we present fluid phase equilibrium data for the two binary systems, ethane + squalane and propane + squalane, over wide ranges of temperature, composition, and pressure.

Experimental Section

Method. The experiments were carried out using the so-called Cailletet apparatus according to the synthetic method. In a binary mixture with constant overall composition, a two-phase equilibrium is bivariant; therefore, by setting the temperature to a certain constant value, only the pressure could be varied until a phase change is observed visually. Bubble- and dew- points are the points at which the last bubble of vapor, or the last droplet of liquid, disappears, respectively ($L + V \rightarrow L$ or $L + V \rightarrow V$). The bubble- and dew-point pressures are measured visually as a function of temperature for mixtures with known composition.

A three-phase equilibrium is monovariant in a binary mixture with a given composition, so if the temperature is fixed, the volume can be adjusted to observe three phases and the recorded pressure is the equilibrium pressure.

A detailed description of the apparatus and the procedure is given elsewhere.[3] A sample of the mixture with known composition is confined in a narrow glass tube, mounted in a thermostatic bath. During the experiments, the temperature is maintained constant to within 0.03 K. The temperature is measured with a Pt100 resistance thermometer, which was calibrated against a standard thermometer with an accuracy of ±0.01 K. The uncertainty in the measured temperature was approximately 0.02 K. The pressure is applied hydraulically and is measured with a dead weight pressure gauge (Budenberg) with an accuracy of 0.005 MPa. The experiments were done for various mixtures covering almost the whole composition range in a temperature range of (263 to 473) K and a pressure range of (0.2 to 16) MPa.

Materials. The mole fraction purity of the ethane (Messer Griesheim) and the propane (Messer Griesheim) used was greater than 0.9995. The squalane was purchased from Merk Schuchardt as “squalane for synthesis” with a minimum purity of at least 99% (GC) and was used without further purification.

Results. The measured *p*-*T* equilibrium data for the binary system ethane + squalane at 12 different compositions are summarized in Table 1. It has been found that this binary exhibits partial miscibility in the liquid phase; that is, in this binary, a three-phase equilibrium liquid−liquid-vapor with lower (LCEP) and upper (UCEP) critical endpoints exists. In the LCEP, the heavy liquid and the light liquid become critical in the presence of a noncritical vapor phase ($L_2 = L_1 + V$). In the UCEP, the light liquid and the vapor become critical in the presence of a noncritical heavy liquid phase ($L_2 + L_1 = V$). The liquid−liquid split occurs in mixtures with composition rich in ethane. For such compositions the transitions from the two-phase $L_1 + V$ to the homogeneous region L_1, and from the two-phase region $L_1 + L_2$ to the homogeneous region L_1, and liquid−liquid-vapor (L_1L_2V) equilibrium data were mea-

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10.1021/je020127z CCC: $25.00

Table 1. Experimental Vapor−Liquid and Liquid−Liquid Equilibrium Data for the System [*x* Ethane + (1 − *x*) Squalane] at Fixed Molar Composition

x		T/K	p/MPa	T/K	p/MPa	T/K	p/MPa
0.9799	$L_1 + V \rightarrow L_1$	248.78	1.265	263.57	1.885	273.59	2.405
		283.57	3.035	293.60	3.790		
	$L_1 + L_2 \rightarrow L_1$	297.54	4.380	298.79	4.665	300.50	5.040
		303.56	5.705	313.57	7.765	323.56	9.660
		328.61	10.566				
0.9752	$L_1 + V \rightarrow L_1$	263.53	1.885	273.52	2.405	283.46	3.030
		293.55	3.785	294.44	3.860		
	$L_1 + L_2 \rightarrow L_1$	297.40	4.380	298.46	4.620	299.37	4.820
		300.34	5.035	301.35	5.260	302.38	5.485
		303.43	5.715	303.46	5.720	304.42	5.925
		305.32	6.120	306.30	6.280	307.07	6.490
		308.40	6.765	313.36	7.775	318.40	8.760
		323.42	9.695	328.47	10.595	332.95	11.330
		342.93	12.925	352.97	14.380		
0.9504	$L_1 + V \rightarrow L_1$	263.59	1.865	273.58	2.385	283.52	3.015
		293.57	3.785				
	$L_1 + L_2 \rightarrow L_1$	298.48	4.410	303.43	5.515	308.48	6.600
		313.41	7.615	318.40	8.605	323.41	9.560
		332.57	11.225	342.50	12.865	352.47	14.365
0.8992	$L + V \rightarrow L$	263.49	1.815	273.58	2.320	283.49	2.925
		293.52	3.665	303.49	4.575	313.49	5.870
		323.05	7.520	333.06	9.235	343.03	10.850
		353.00	12.360	362.94	13.760	372.89	15.050
		382.81	16.240				
0.8498	$L + V \rightarrow L$	263.55	1.655	273.59	2.115	283.69	2.665
		293.48	3.300	303.51	4.065	313.55	4.965
		323.50	6.015	332.92	7.230	342.92	8.525
		353.00	9.825	363.03	11.085	373.03	12.280
		382.96	13.395	392.85	14.435		
0.8007	$L + V \rightarrow L$	263.45	1.500	273.42	1.900	283.49	2.380
		293.46	2.935	303.51	3.580	313.49	4.300
		323.45	5.110	332.95	5.945	342.82	6.885
		352.79	7.860	362.87	8.860	372.91	9.850
		382.87	10.790	392.80	11.695	402.79	12.550
		412.83	13.365	422.85	14.120	432.86	14.825
0.7012	$L + V \rightarrow L$	263.48	1.205	273.39	1.505	283.48	1.870
		293.59	2.270	299.35	2.540	313.46	3.210
		323.45	3.750	332.97	4.300	342.94	4.905
		352.85	5.525	362.83	6.165	372.56	6.805
		382.78	7.460	392.76	8.095	402.72	8.715
		412.76	9.315	422.77	9.850	432.79	10.435
0.7012	$L + V \rightarrow L$	442.79	10.950	452.81	11.425	462.81	11.875
		472.79	12.290				
0.5975	$L + V \rightarrow L$	263.78	0.885	273.62	1.195	283.59	1.450
		293.51	1.735	303.50	2.050	313.58	2.400
		322.98	2.825	323.61	2.770	332.79	3.145
		342.67	3.555	352.91	3.980	362.99	4.425
		373.01	4.830	382.43	5.270	382.43	5.270
		392.31	5.700	402.04	6.140	412.06	6.545
		422.70	6.950	432.69	7.340	442.68	7.715
		452.71	8.060	462.68	8.405	472.67	8.715
0.5031	$L + V \rightarrow L$	273.52	0.865	283.69	1.070	293.52	1.265
		303.50	1.495	313.57	1.745	323.03	1.990
		332.98	2.270	342.90	2.570	352.77	2.855
		362.36	3.165	372.43	3.480	382.68	3.780
		392.25	4.100	402.38	4.405	412.46	4.705
		422.71	4.995	432.51	5.290	442.71	5.560
		452.73	5.825	462.70	6.070	472.72	6.310
0.3989	$L + V \rightarrow L$	273.64	0.610	283.53	0.745	293.59	0.895
		303.53	1.050	313.60	1.225	323.48	1.405
		332.67	1.580	333.66	1.600	342.60	1.775
		352.73	1.990	362.89	2.195	372.89	2.410
		382.84	2.615	392.74	2.835	402.71	3.035
		412.63	3.240	422.64	3.440	432.65	3.640
		442.52	3.830	452.66	4.015	462.66	4.190
		472.68	4.360				
0.2994	$L + V \rightarrow L$	273.57	0.430	283.58	0.530	293.68	0.630
		303.50	0.720	313.48	0.830	323.62	0.955
		332.92	1.075	333.66	1.080	342.75	1.200
		352.72	1.330	362.79	1.470	372.75	1.605
		382.70	1.745	392.82	1.890	402.76	2.025
		412.35	2.160	422.39	2.295	432.49	2.425
		442.52	2.560	452.57	2.685	462.59	2.805
		472.60	2.925				
0.2046	$L + V \rightarrow L$	283.60	0.330	293.53	0.385	303.60	0.450
		313.56	0.520	323.48	0.600	332.99	0.670
		342.92	0.745	352.87	0.825	362.85	0.915
		372.79	1.000	382.75	1.090	392.77	1.170
		402.82	1.260	412.52	1.345	422.85	1.425
		422.85	1.425	432.82	1.520	442.84	1.600
		452.91	1.655	462.75	1.740	472.76	1.815

Table 2. Experimental Data of the Three-Phase Equilibrium L_1L_2V in the Ethane-Squalane System

T/K	p/MPa	T/K	p/MPa	T/K	p/MPa
295.57	3.930[a]	298.79	4.231	302.53	4.581
295.71	3.961	299.33	4.287	303.43	4.685
296.00	3.979	300.34	4.374	304.42	4.775
296.53	4.028	300.94	4.436	305.32	4.865
297.38	4.105	301.35	4.474	306.51	4.996
297.89	4.166	302.37	4.570	306.69	5.013[b]
298.30	4.196				

[a] LCEP ($L_2 = L_1 + V$). [b] UCEP ($L_2 + L_1 = V$).

sured (see Figure 1). As depicted in Figure 1, which present an isopleth for $x_{\text{ethane}} = 0.9504$, the intersection of the $L_1 + V \rightarrow L_1$ and $L_2 + L_1 \rightarrow L_1$ curves gives a point on the three-phase equilibrium line. The three-phase equilibrium data and both the critical end points are presented in Table 2. The two critical end points measured in this work are slightly different from the ones reported by Paas et al.,[1] i.e., $T_{\text{LCEP}} = 295.7$ K, $p_{\text{LCEP}} = 3.87$ MPa, and $T_{\text{UCEP}} = 306.8$ K, $p_{\text{UCEP}} = 4.92$ MPa. In accordance with ref 1, an additional liquid−liquid immiscibility should occur in this system at very low temperatures. However, this could not be measured with the experimental setup used in this work and can be observed only in strongly undercooled liquid mixtures. Therefore, the present work suggests that the fluid phase behavior of this system most likely belongs to type V in the classification of Van Konynenburg and Scott.[4]

In the system propane + squalane, no liquid−liquid immiscibility is found in the range of temperature and pressure covered by this work. The measured bubble-point ($L + V \rightarrow L$), dew-point ($L + V \rightarrow V$), and critical-point ($L = V$) temperatures and pressures for this binary, at 13 different compositions, are given in Table 3. Therefore, according to the present, work the fluid phase behavior in

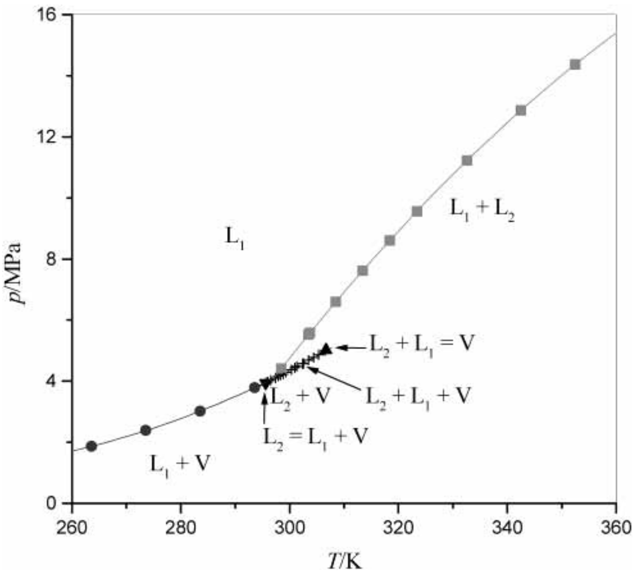

Figure 1. *p-T* diagram showing two-phase transition curves and the three-phase equilibrium line for the binary system [*x* ethane + (1 − *x*) squalane] with *x* = 0.9504: ●, $L_1 + V \rightarrow L_1$; ■, $L_2 + L_1 \rightarrow L_1$; +, $L_2 + L_1 + V$; ▼, LCEP ($L_2 = L_1 + V$); ▲, UCEP ($L_2 + L_1 = V$).

Table 3. Experimental Vapor−Liquid Equilibrium Data for the System [x Propane + (1 − x) Squalane] at Fixed Molar Composition

x		T/K	p/MPa	T/K	p/MPa	T/K	p/MPa
0.9803	L + V → L	263.49	0.349	273.49	0.476	283.46	0.635
		293.45	0.831	303.40	1.069	313.41	1.354
		323.40	1.690	333.43	2.084	343.42	2.540
		353.36	3.062	363.35	3.669	373.35	4.374
		383.36	5.227				
	L = V	393.46	6.123				
	L + V → V	394.39	6.303	395.41	6.403	403.39	7.163
		413.40	8.063	423.41	8.888	433.48	9.638
0.9751	L + V → L	263.56	0.347	273.62	0.474	283.59	0.634
		293.56	0.832	303.50	1.069	303.56	1.071
		313.50	1.357	322.97	1.680	323.58	1.697
		332.93	2.075	342.85	2.525	352.74	3.040
		362.76	3.655	367.78	4.000	372.78	4.360
		382.70	5.200	387.68	5.685	392.66	6.170
		397.66	6.660				
	L = V	398.67	6.755				
	L + V → V	399.16	6.805	400.70	6.945	402.65	7.140
		407.65	7.600	412.68	8.045	422.66	8.885
		432.65	9.650	442.65	10.335	452.68	10.940
		462.68	11.460	472.73	11.905		
0.9701	L + V → L	263.46	0.333	273.50	0.459	283.47	0.617
		293.45	0.814	303.39	1.047	313.38	1.327
		323.35	1.656	333.33	2.044	343.35	2.512
		353.35	3.032	363.36	3.636	373.51	4.349
		383.42	5.192	393.48	6.176	403.48	7.141
		404.45	7.236				
	L = V	406.44	7.416				
	L + V → V	408.46	7.601	413.48	8.051	423.45	8.891
		428.48	9.291	433.61	9.672	443.51	10.357
		453.51	10.977	463.52	11.512	473.51	11.967
0.9600	L + V → L	263.45	0.340	273.41	0.464	283.44	0.621
		293.42	0.813	303.40	1.047	313.38	1.327
		323.35	1.655	333.35	2.040	343.30	2.489
		353.23	3.003	363.20	3.599	373.17	4.296
		383.19	5.134	393.39	6.142	403.34	7.137
		413.34	8.072	423.31	8.937	428.31	9.342
		430.30	9.497	432.29	9.647	438.27	10.092
		442.30	10.372				
	L = V	442.77	10.407				
	L + V → V	443.30	10.437	448.30	10.772		
0.9499	L + V → L	263.43	0.338	273.48	0.463	283.46	0.618
		293.44	0.809	303.44	1.042	313.42	1.320
		323.41	1.651	333.41	2.041	343.46	2.502
		353.33	3.016	363.29	3.612	373.20	4.304
		373.20	4.304	383.14	5.130	393.08	6.092
		402.94	7.082	412.70	8.017	422.58	8.882
		432.30	9.662	442.13	10.382	447.18	10.723
		447.21	10.717	452.16	11.038	457.12	11.333
		461.09	11.558				
0.9000	L + V → L	273.52	0.441	283.50	0.587	293.43	0.765
		303.40	0.987	263.49	0.323	313.42	1.245
		323.40	1.558	333.41	1.912	333.53	1.918
		343.52	2.330	353.51	2.808	363.47	3.358
0.9000	L + V → L	373.45	3.979	383.42	4.689	393.33	5.479
		403.39	6.336	403.28	6.342	413.11	7.212
		422.66	8.052	432.79	8.887	442.59	9.652
		452.20	10.357	461.96	11.012	471.86	11.607
0.7999	L + V → L	263.44	0.274	273.47	0.372	283.44	0.494
		293.44	0.644	303.40	0.823	313.38	1.037
		323.39	1.286	333.32	1.570	343.27	1.893
		353.23	2.260	363.22	2.669	373.21	3.121
		383.21	3.614	393.22	4.148	403.24	4.713
		413.25	5.299	423.26	5.908	433.42	6.519
		443.41	7.119	453.39	7.704	463.34	8.269
		473.34	8.809				
0.7005	L + V → L	263.48	0.225	273.52	0.305	283.45	0.403
		293.47	0.524	303.42	0.667	313.40	0.833
		323.41	1.025	333.41	1.246	343.29	1.492
		353.28	1.766	363.27	2.069	373.27	2.399
		383.25	2.752	393.23	3.134	403.23	3.526
		413.25	3.937	423.22	4.360	433.18	4.788
		443.28	5.219	453.22	5.648	463.22	6.069
		473.28	6.480				
0.6012	L + V → L	263.47	0.180	273.49	0.242	283.46	0.320
		293.48	0.416	303.40	0.530	313.39	0.656
		323.37	0.802	333.36	0.961	343.34	1.152
		353.29	1.357	363.28	1.581	373.26	1.824
		383.29	2.084	393.26	2.354	403.27	2.637
		413.30	2.932	423.28	3.234	433.30	3.541
		443.30	3.852	453.29	4.154	463.28	4.462
		473.28	4.763				
0.5012	L + V → L	298.89	0.360	303.81	0.407	313.44	0.499
		323.39	0.609	333.30	0.738	343.31	0.872
		353.50	1.024	363.50	1.186	373.49	1.360
		383.49	1.549	393.52	1.741	403.53	1.947
		413.56	2.157	423.63	2.372	433.68	2.598
		443.36	2.807	453.32	3.032	463.27	3.249
0.3980	L + V → L	297.92	0.259	304.56	0.300	313.42	0.362
		323.35	0.440	333.30	0.529	343.27	0.624
		353.24	0.731	363.35	0.846	373.31	0.967
		383.28	1.097	393.26	1.231	403.25	1.373
		413.22	1.528	423.20	1.665	433.16	1.814
		443.14	1.970	453.08	2.119	463.07	2.272
		473.06	2.426				
0.3018	L + V → L	315.97	0.273	325.31	0.325	332.42	0.366
		341.83	0.427	347.72	0.467	352.71	0.503
		362.61	0.581	372.72	0.666	382.71	0.753
		392.69	0.844	402.67	0.939	412.58	1.038
		422.66	1.140	432.68	1.241	442.65	1.344
		452.68	1.449	462.66	1.554	473.82	1.669
0.1525	L + V → L	332.87	0.182	342.79	0.205	352.77	0.237
		362.74	0.271	372.76	0.307	382.71	0.346
		392.72	0.388	402.71	0.430	412.70	0.474
		422.70	0.519	432.73	0.565	442.72	0.610
		453.22	0.658	463.21	0.705	473.39	0.751

the system propane + squalane is of type I in the before mentioned classification,[4] even though at lower temperatures a liquid−liquid immiscibility with an upper critical solution temperature as for a type II behavior might occur.

Isothermal p, x sections were obtained by interpolation from polynomials fitted to the p, T data.

Modeling

The bubble-point VLE data of both binary systems were modeled with the Soave−Redlich−Kwong (SRK) equation of state[5] using the VLE95 program[6] for fluid phase equilibrium calculations. The classical one-fluid van der Waals mixing rules were applied as follows:

$$a = \sum_{i=1}^{2}\sum_{j=1}^{2} x_i x_j (a_i a_j)^{0.5}(1 - k_{ij}) \qquad (1)$$

$$b = \sum_{i=1}^{2}\sum_{j=1}^{2} x_i x_j \frac{(b_i + b_j)}{2}(1 - l_{ij}) \qquad (2)$$

Figures 2 and 3 show that the calculated VLE curves in the two investigated systems are in agreement with the experimental isothermal p,x data. The optimized values of the interaction parameters, k_{ij} and l_{ij}, for each isotherm, and the average absolute deviation in estimating the saturation pressures, AADP, eq 3 are summarized in Table 4.

$$\mathrm{AADP} = \frac{1}{\mathrm{NEXP}} \sum_{i=1}^{\mathrm{NEXP}} \left| \frac{p_i^{\exp} - p_i^{\mathrm{calc}}}{p_i^{\exp}} \right| 100 \qquad (3)$$

The prediction of three-phase region for the binary mixture ethane + squalane was performed with the binary interaction parameters set to zero and with the binary interaction parameters fitted to vapor−liquid equilibrium data at the temperature of LCEP. The calculated critical curve and the predicted the three-phase curve are shown in Figures 4 and 5. The results obtained are in qualitative agreement with the experimental data.

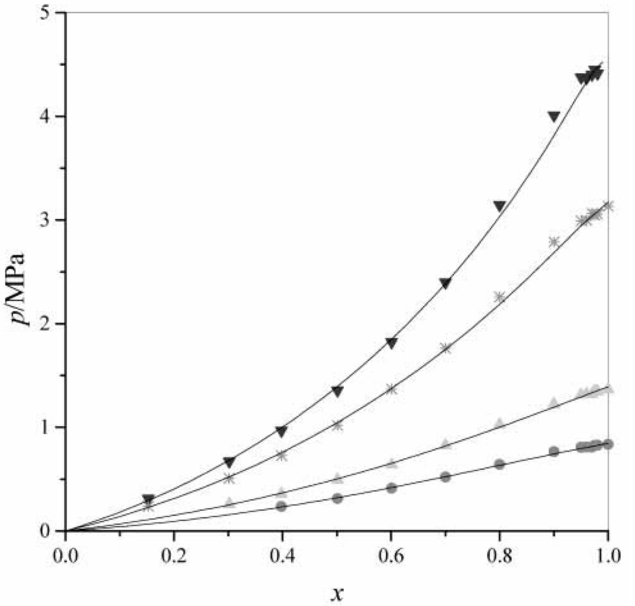

Figure 2. Vapor − liquid equilibria for [x propane + (1 − x) squalane] depicted as isothermal p, x sections. Experimental data: ●, 293.15 K; ▲, 313.15 K; *, 353.15 K; ▼, 373.15 K; solid lines, calculated with SRK-EoS and k_{ij} and l_{ij} from Table 4.

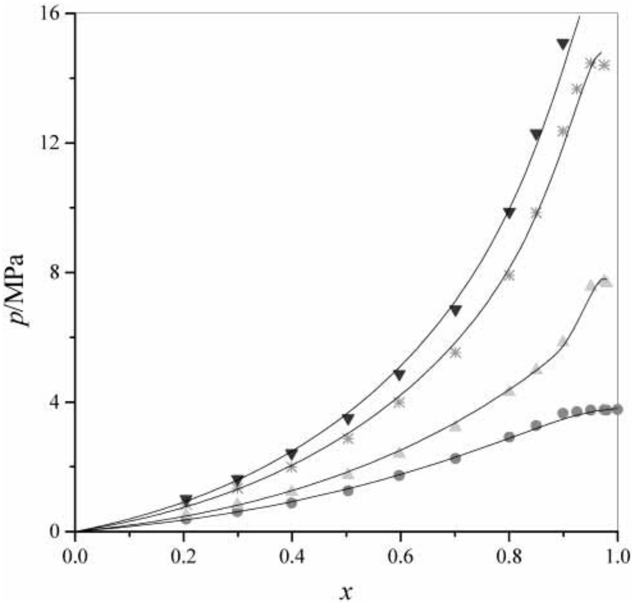

Figure 3. Vapor−liquid equilibria for [x ethane + (1 − x) squalane] depicted as isothermal p, x sections. Experimental data; ●, 293.15 K; ▲, 313.15 K; *, 353.15 K; ▼, 373.15 K; solid lines, calculated with SRK-EoS and k_{ij} and l_{ij} from Table 4.

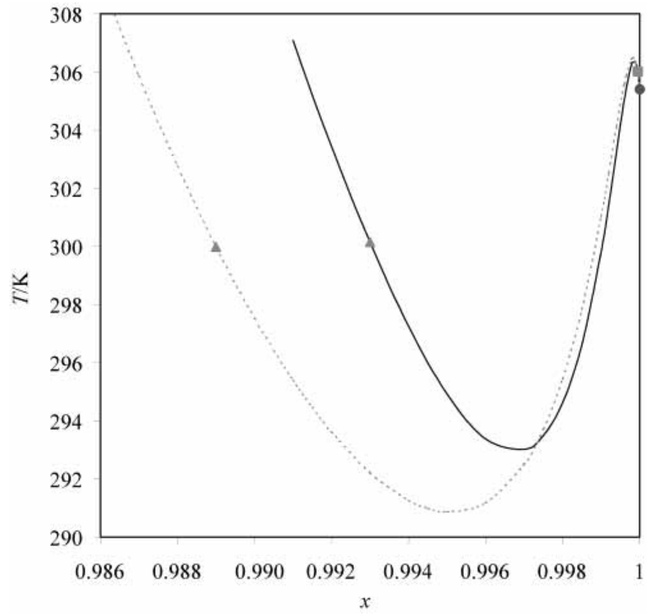

Figure 4. $T-x$ projection of the critical curve for the system [x ethane + (1 − x) squalane]. Solid line, calculated with SRK-EoS and $k_{ij} = l_{ij} = 0$; dashed line, calculated with SRK-EoS and k_{ij} and l_{ij}, from VLE at LCEP; ●, ethane CP; ■, UCEP; ▲, LCEP.

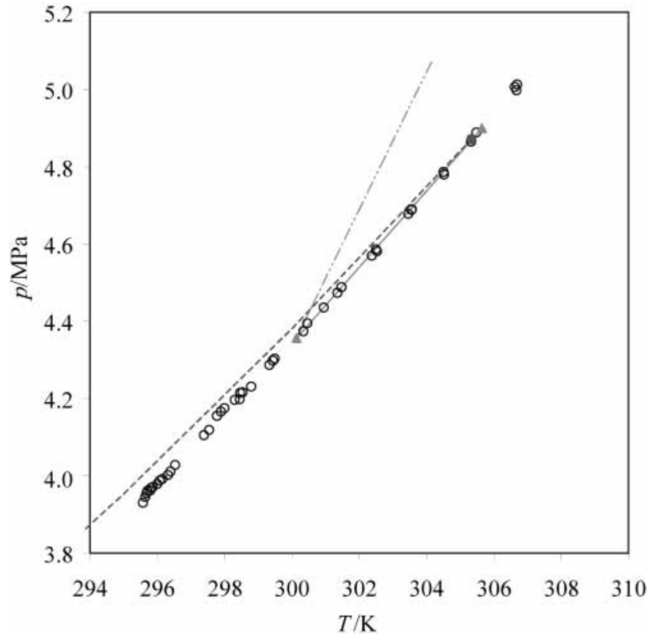

Figure 5. $p-T$ projection of the three phase curve and of the critical line for the system ethane + squalane: ○, LLVE experimental data; - - -, ethane LVE; ●, ethane CP; −▲−, LLVE calculated with SRK-EoS and $k_{ij} = l_{ij} = 0$; − - - −, critical line ($L_1 = L_2$) calculated with SRK-EoS and $k_{ij} = l_{ij} = 0$.

Table 4. Modeling Results of the Vapor − Liquid Equilibria

	ethane−squalane system			propane−squalane system		
T/K	k_{ij}	l_{ij}	AADP %	k_{ij}	l_{ij}	AADP %
293.15	0.0151	0.0229	2.38	0.0014	0.0214	1.38
313.15	0.0236	0.0289	3.18	−0.0059	0.0189	1.65
353.15	0.0239	0.0350	3.52	−0.0090	0.0203	1.93
373.15	0.0209	0.0364	3.68	−0.0080	0.0235	2.20

Discussions and Conclusions

This paper presents fluid phase equilibrium data for the binary systems ethane + squalane and propane + squalane. The results confirm the occurrence of a liquid−liquid-vapor equilibrium in the ethane + squalane system at temperatures close to the critical point of ethane. The present measurements indicate that this binary is of type V in the classification of Van Konynenburg and Scott.[4] In the temperature range covered in this work, the system propane + squalane behaves as a type I system in the before mentioned classification. However, according to some earlier work[1] the systems ethane + squalane and propane + squalane should exhibit type IV and type II behavior, respectively. Further investigations in strongly undercooled conditions should be done to confirm this hypothesis (i.e., to show the occurrence of a liquid−liquid demixing at low temperatures with an upper critical solution temperature). The VLE modeling results and the predicted LLV line are in qualitative agreement with the experimental data.

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Received for review July 10, 2002. Accepted December 20, 2002.

JE020127Z