Bubble Point Pressures and Densities for the Binary Systems of Propane with Triacontane, Hexatriacontane, Tetracontane, Pentacontane, and Squalane at 353−373 K and 4.00−7.00 MPa

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Bubble point pressures and densities were measured with a variable volume cell for the binary systems propane + triacontane (n-$C_{30}H_{62}$), + hexatriacontane (n-$C_{36}H_{74}$), + tetracontane (n-$C_{40}H_{82}$), + pentacontane (n-$C_{50}H_{102}$), and + squalane ($C_{30}H_{62}$, 2,6,10,15,19,23-hexamethyltetracosane). The densities were measured for compressed liquids or supercritical fluids in the one-phase region. The studied temperature and pressure ranges were 353−373 K and 4.00−7.00 MPa, respectively. The mass fractions of the long-chain alkanes varied between 0 and 0.0326. It was found that the densities of the mixtures increased almost linearly with the mass fractions of the heavy alkanes. The effects of all the long-chain alkanes on the density did not depend on molar mass. Bubble point pressures decreased only very slightly as the mass fractions of the long-chain alkanes increased.

Introduction

The phase diagrams of propane + long-chain alkane systems have been studied previously. Leder et al. (1976) studied the propane + hexatriacontane system and Peters et al. (1989) several systems with C_{32}−C_{50} alkanes as the heavier component. Peters et al. (1992, 1993) have reported detailed phase diagrams for the propane + tetratriacontane and propane + hexacontane systems, respectively. Condo et al. (1992) have reported cloud point pressures and solid−supercritical fluid solubilities in propane + linear polyethylene systems, which are analogous systems at least when fractionated polyethylene samples are used.

Although some data on the phase behavior of these systems are available, the liquid (or supercritical fluid) densities have not been previously reported. Bubble point pressures for the systems propane + tetratriacontane and propane + hexacontane are available (Peters et al., 1992, 1993), but no bubble points are available in the literature for the systems studied in this work. PVT and vapor pressure data are useful both for process design and for developing equation of state models. Long-chain alkanes can be used as monodisperse model components for small polymer (polyethylene) molecules. Propane + long-chain alkane solutions are also examples of very asymmetric mixtures, which are interesting from a modeling point of view. In addition, the measurements reported here have been performed in the vicinity of the critical point of propane.

Experimental Section

Materials. The propane used in these measurements was supplied by AGA Oy (purity 99.95 mol %). The long-chain alkanes were all purchased from Aldrich-Chemie, and their certified purities were 99 mol % (triacontane), 98 mol % (hexatriacontane), 98 mol % (tetracontane), >99 mol % (pentacontane), and 99 mol % (squalane). All materials were used without further purification.

Apparatus. A DB Robinson Jefri sapphire glass cell apparatus was used for measuring the densities and bubble point pressures for the systems studied. The main component of the apparatus is a sapphire glass cylinder, which is mounted vertically in a thermostated air bath (Figure 1). The maximum volume of the cylinder is approximately 56 cm^3. There is a metal piston equipped with two Viton O-rings inside the cylinder. While measurements are done, the fluid sample is placed above the piston while the space below the piston is filled with hydraulic oil. The volume of the sample can be changed by increasing or decreasing the amount of hydraulic oil below the piston. This is done by means of a manually operated pump. The mixing of the sample can be done using a metal mixer, which is moved up and down by a U-shaped magnet moving outside the cylinder.

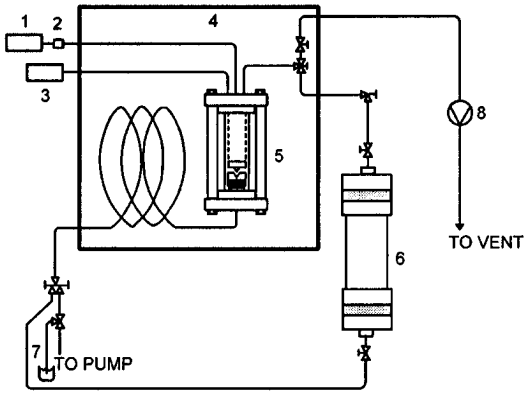

Figure 1. The sapphire cell apparatus: (1) pressure display, (2) pressure transducer, (3) temperature display, (4) thermostatted air bath, (5) sapphire glass cylinder, (6) feed cylinder, (7) oil reservoir, (8) vacuum pump.

The cell temperature was measured with an ERO temperature gauge equipped with a Pt-100 thermoprobe, which was calibrated against a Unomat TRX temperature calibrator. The cell pressure was measured with a Beamex pressure gauge, which was calibrated against a Beamex PC105 pressure calibrator. Temperature and pressure were both measured directly from the experimental fluid. The cell volume was determined by measuring the positions of the piston and the upper end of the cylinder by means

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Table 1. Measured Densities of the Systems Studied

T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})
Propane											
Experiment 1											
343.1	3.988	415.9	343.1	6.985	439.6	353.2	6.988	415.3	363.3	6.985	392.6
343.0	4.486	421.2	353.1	3.995	385.1	363.1	3.991	336.4	373.1	4.634	270.0
343.0	4.989	424.3	353.2	4.490	391.6	363.2	4.488	352.8	373.3	4.986	306.7
343.1	5.484	428.6	353.2	4.985	397.6	363.2	4.989	364.9	373.3	5.487	329.0
343.1	5.989	432.2	353.2	5.485	402.8	363.2	5.487	373.2	373.3	5.987	343.5
343.1	6.489	435.2	353.2	5.988	408.0	363.3	5.986	381.0	373.3	6.488	354.3
343.1	6.886	438.3	353.2	6.484	412.2	363.3	6.484	387.2	373.2	6.984	362.6
Experiment 2											
343.1	3.991	415.7	353.2	3.992	386.1	363.1	3.994	337.0	373.1	4.611	245.9
343.1	4.490	420.2	353.2	4.489	393.0	363.2	4.484	353.3	373.4	4.988	304.5
343.1	4.988	423.6	353.2	4.990	399.0	363.2	4.986	365.1	373.4	5.481	329.0
343.1	5.488	428.0	353.2	5.488	403.0	363.3	5.487	373.7	373.4	5.983	344.6
343.1	5.986	430.0	353.2	5.988	409.4	363.3	5.986	380.5	373.4	6.487	354.8
343.1	6.486	433.2	353.2	6.485	412.8	363.2	6.486	387.4	373.4	6.982	363.8
343.1	6.986	436.7	353.2	6.985	418.3	363.2	6.985	393.7			
Propane (1) + Triacontane (2)											
$w_2 = 0.0098$											
353.2	3.989	390.7	353.2	6.487	416.6	363.2	5.488	378.4	373.4	4.988	316.0
353.2	4.492	397.3	353.2	6.986	420.5	363.2	5.987	385.4	373.4	5.485	337.8
353.2	4.989	402.9	363.1	3.992	344.9	363.2	6.487	391.0	373.4	5.987	350.8
353.2	5.492	407.9	363.2	4.489	359.8	363.2	6.983	396.8	373.3	6.487	361.3
353.2	5.987	412.7	363.2	4.990	370.7	373.1	4.507	240.6	373.3	6.984	369.2
$w_2 = 0.0206$											
353.1	3.990	394.9	353.2	6.484	419.4	363.2	5.486	383.3	373.3	4.985	324.7
353.1	4.495	400.8	353.2	6.986	423.2	363.2	5.986	388.9	373.3	5.483	342.9
353.1	4.989	406.7	363.1	3.992	352.9	363.2	6.484	394.6	373.3	5.985	355.7
353.2	5.490	411.1	363.1	4.491	365.2	363.2	6.987	399.2	373.3	6.487	363.9
353.2	5.987	415.4	363.2	4.986	374.5	373.0	4.496	275.0	373.2	6.985	372.7
$w_2 = 0.0289$											
353.3	3.994	399.1	353.4	6.486	422.6	363.3	5.489	386.3	373.4	4.982	332.4
353.3	4.493	404.8	353.4	6.987	425.7	363.3	5.989	392.6	373.4	5.489	349.5
353.3	4.988	410.1	363.2	3.993	357.3	363.3	6.486	398.0	373.4	5.983	360.8
353.4	5.489	415.0	363.2	4.487	369.0	363.3	6.987	402.5	373.4	6.485	369.1
353.4	5.986	418.9	363.3	4.989	379.0	373.2	4.492	291.6	373.4	6.983	377.0
Propane (1) + Hexatriacontane (2)											
$w_2 = 0.0108$											
353.2	3.990	390.8	353.3	6.484	416.5	363.3	5.486	377.9	373.4	5.482	335.6
353.2	4.489	396.8	353.2	6.987	420.0	363.2	5.986	386.3	373.4	5.986	349.3
353.2	4.987	402.5	363.2	3.993	344.6	363.2	6.487	391.8	373.5	6.483	359.4
353.2	5.484	406.9	363.2	4.487	358.8	363.2	6.983	397.1	373.4	6.986	368.9
353.2	5.988	411.9	363.3	4.987	369.9	373.4	4.986	314.3			
$w_2 = 0.0227$											
353.2	3.998	395.8	353.3	6.488	420.9	363.2	5.488	384.9	373.4	5.484	343.0
353.2	4.494	402.3	353.2	6.982	424.5	363.2	5.987	391.1	373.4	5.982	356.3
353.2	4.991	407.5	363.1	3.993	353.3	363.3	6.480	397.0	373.4	6.485	366.3
353.2	5.490	412.4	363.2	4.491	366.1	363.3	6.986	401.9	373.4	6.984	373.7
353.2	5.988	416.3	363.2	4.987	376.6	373.4	4.984	323.8			
$w_2 = 0.0308$											
353.1	3.992	402.0	353.2	6.483	425.4	363.2	5.490	389.7	373.4	5.488	352.0
353.2	4.493	407.5	353.2	6.988	429.1	363.2	5.990	395.7	373.3	5.984	363.3
353.2	4.989	412.9	363.1	3.992	360.7	363.2	6.485	401.0	373.3	6.484	372.0
353.2	5.490	417.0	363.1	4.489	373.1	363.2	6.986	406.4	373.4	6.985	379.4
353.2	5.986	421.4	363.1	4.990	382.4	373.4	4.987	333.7			
$w_2 = 0.0319$											
353.2	3.994	401.3	353.2	6.488	424.9	363.2	5.487	388.4	373.4	5.484	350.9
353.2	4.492	407.4	353.2	6.982	429.8	363.2	5.989	395.4	373.4	5.985	363.0
353.2	4.988	413.1	363.1	3.992	359.8	363.2	6.484	400.3	373.4	6.487	372.4
353.2	5.490	417.1	363.2	4.487	372.3	363.2	6.986	405.5	373.4	6.978	378.9
353.2	5.988	421.8	363.2	4.989	380.9	373.5	4.983	331.1			
Propane (1) + Tetracontane (2)											
$w_2 = 0.0096$											
353.2	3.990	391.7	353.2	6.483	416.5	363.2	5.485	378.1	373.4	5.486	337.3
353.1	4.491	398.2	353.2	6.982	420.6	363.2	5.984	385.4	373.4	5.984	350.2
353.2	4.992	403.4	363.0	3.991	345.1	363.2	6.488	391.3	373.4	6.484	360.7
353.2	5.486	408.4	363.1	4.487	359.5	363.2	6.980	396.3	373.3	6.985	369.0
353.2	5.987	412.8	363.2	4.989	370.1	373.3	4.986	315.7			
$w_2 = 0.0190$											
353.1	3.992	395.0	353.2	6.486	419.7	363.2	5.489	382.5	373.3	5.487	343.5
353.2	4.491	401.1	353.2	6.982	422.9	363.2	5.986	389.8	373.3	5.986	356.1
353.2	4.990	406.4	363.1	3.990	351.1	363.2	6.486	395.0	373.3	6.485	365.5
353.2	5.490	411.3	363.1	4.489	364.6	363.2	6.983	399.6	373.3	6.986	373.7
353.2	5.987	415.3	363.2	4.988	374.5	373.4	4.985	323.6			

Table 1 (Continued)

T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})	T/K	P/MPa	ρ/(kg·m^{-3})
Propane (1) + Tetracontane (2)											
$w_2 = 0.0318$											
353.1	3.990	399.8	353.2	6.487	424.4	363.2	5.488	388.1	373.4	5.483	347.7
353.1	4.492	405.8	353.2	6.987	426.6	363.2	5.984	394.1	373.4	5.985	359.7
353.2	4.990	410.4	363.1	3.994	358.9	363.2	6.484	399.5	373.3	6.485	368.7
353.2	5.489	414.7	363.2	4.491	371.3	363.2	6.984	404.2	373.3	6.985	377.2
353.2	5.987	419.2	363.2	4.990	380.5	373.4	4.984	331.0			
Propane (1) + Pentacontane (2)											
$w_2 = 0.0100$											
353.2	3.991	390.6	353.3	6.486	416.7	363.3	5.487	378.9	373.5	5.488	337.3
353.2	4.490	397.2	353.3	6.985	419.8	363.3	5.988	385.1	373.4	5.987	350.9
353.2	4.989	402.6	363.1	3.992	344.8	363.3	6.485	391.5	373.4	6.486	361.1
353.3	5.484	407.6	363.2	4.491	359.9	363.3	6.982	397.2	373.4	6.984	368.4
353.2	5.989	412.2	363.3	4.984	370.8	373.5	4.987	315.3			
$w_2 = 0.0199$											
353.1	3.992	396.0	353.3	6.487	420.5	363.3	5.488	383.7	373.4	5.984	356.7
353.2	4.489	402.3	353.3	6.988	424.0	363.3	5.985	390.8	373.4	6.486	366.7
353.2	4.988	407.7	363.2	3.992	352.7	363.3	6.485	396.7	373.4	6.983	374.7
353.3	5.486	411.6	363.2	4.490	366.3	363.3	6.987	401.7			
353.3	5.985	416.7	363.3	4.983	376.7	373.5	5.484	343.9			
$w_2 = 0.0201$											
353.1	3.991	395.3	353.2	6.486	420.1	363.2	5.486	383.9	373.4	5.987	356.7
353.1	4.493	401.8	353.2	6.987	423.6	363.2	5.987	390.7	373.3	6.484	366.6
353.2	4.988	406.8	363.1	3.993	352.7	363.2	6.487	396.5	373.3	6.987	374.5
353.1	5.488	411.7	363.1	4.487	366.1	363.2	6.984	401.5			
353.2	5.987	415.8	363.2	4.987	376.3	373.4	5.485	344.2			
$w_2 = 0.0279$											
353.2	3.991	400.1	353.2	6.486	424.3	363.2	5.486	388.6	373.4	5.984	361.4
353.2	4.494	406.3	353.2	6.983	427.8	363.2	5.987	394.5	373.4	6.485	371.2
353.2	4.992	411.7	363.1	3.995	357.6	363.3	6.487	400.6	373.4	6.986	378.8
353.2	5.490	415.7	363.2	4.487	371.0	363.2	6.982	405.3			
353.2	5.988	420.0	363.2	4.990	380.5	373.4	5.484	349.4			
Propane (1) + Squalane (2)											
$w_2 = 0.0326$											
353.2	3.987	400.0	353.2	6.485	423.5	363.2	5.484	388.1	373.3	5.486	349.8
353.2	4.490	406.6	353.2	6.986	427.2	363.2	5.989	394.9	373.3	5.982	361.1
353.2	4.989	410.9	363.0	3.992	359.3	363.2	6.484	399.9	373.3	6.482	370.6
353.2	5.489	415.9	363.1	4.489	371.6	363.2	6.983	405.2	373.3	6.983	378.2
353.2	5.985	419.9	363.1	4.989	381.1	373.3	4.986	333.0			

of a cathetometer, and calculating the volume on the basis of the known cross-sectional area of the cylinder.

Procedure. The solid long-chain alkane (liquid, in the case of squalane) was weighed and placed in the cylinder, which was dismantled at this stage. When densities for pure propane were measured, the previous step was omitted. Then the cylinder was assembled again, tested for leaks, and evacuated three times, flushing with gaseous propane in between the evacuations. After the last evacuation, liquid propane was transferred to the cylinder from a pressure vessel at room temperature. The propane added into the cylinder was compressed until the vapor phase formed in the cylinder had disappeared and the pressure was clearly above the vapor pressure of propane. The mass of propane was determined by measuring the volume of the cylinder at this stage, and using the Hankinson−Brobst−Thomson (HBT) correlation (Thomson et al., 1982) to obtain the density of propane at the temperature and pressure of the system. The HBT correlation, also known as COSTALD, is a highly accurate corresponding states correlation developed for the calculation of saturated and compressed liquid densities. The propane densities calculated from HBT correlation were in excellent agreement with the density values recommended by Younglove and Ely (1987) in the typical filling conditions. The average relative error in HBT predictions was 0.0201%. The volume of propane was corrected for the volume taken by the long-chain alkane. The solid density of hexatriacontane at 20 °C, which is 0.962 g/cm^3 (Landolt-Börnstein, 1971) was used for all solid alkanes. Since the volume of the long-chain alkane is very small compared to the volume of propane, an error of ±0.050 g/cm^3 in the density of the long-chain alkane causes only ±0.08% error in the final results of the density measurements. The density of squalane at 20 °C is 0.810 g/cm^3 as given by the supplier.

The thermostated air bath was heated to the desired temperature, and approximately 1 h was allowed for equilibration before actual measurements were started. Bubble point pressures were measured by bringing the system to a state where only a small bubble of gas could be seen in the cylinder. At that stage, the temperature and the pressure of the system were recorded. Then the system was compressed to the next pressure, where fluid density was to be measured. The temperature and pressure of the system were recorded, as well as the position of the piston. Positions of the piston (sample volumes) were measured similarly at all pressures, keeping the temperature constant. Then the temperature of the system was changed, approximately 1 h was allowed for equilibration, and the bubble point pressures and densities were measured again. Densities were calculated on the basis of known sample mass and volume; volume was calculated from the position of the piston.

The calculations performed to obtain the experimental densities are described here. The mass of the long-chain alkane m_2 was obtained by weighing. The mass of propane m_1 was calculated from eq 1, where ρ_{HBT} is the density of

$$m_1 = \rho_{HBT}[(y_2 - y_1)A + V_0 - V_2] \quad (1)$$

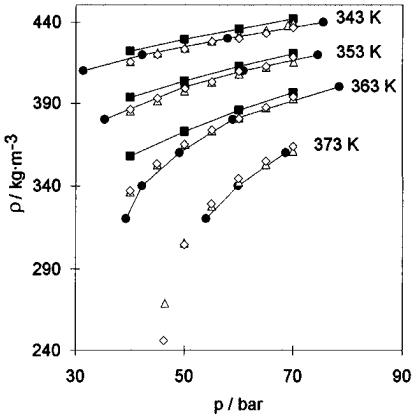

Figure 2. Density of pure propane: comparison of literature values and this work. The symbols are as follows: ■, Vargaftik (1975); ●, Dittmar et al. (1962); △, ◇, this work. The temperatures of the isotherms are given in the figure.

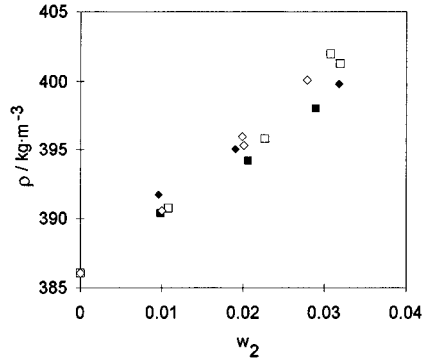

Figure 3. Densities of the propane (1) + long-chain alkane (2) systems as functions of w_2, the mass fraction of the long-chain alkane (T = 353 K, p = 4.0 MPa). Component (2) is as follows: ■, triacontane; □, hexatriacontane; ◆, tetracontane; ◇, pentacontane. Similar figures can be obtained by plotting the experimental data at any other temperature and pressure.

propane predicted by HBT at the filling conditions, y_2 is the position of the upper end of the cylinder, y_1 is the position of the piston, A is the cross-sectional area of the cylinder, V_0 is a volumetric correction due to the shape of the piston and the mixer, and V_2 is the volume of the long-chain alkane. V_2 is calculated from eq 2, where m_2 is the

$$V_2 = m_2/\rho_2 \tag{2}$$

mass and ρ_2 the density of the long-chain alkane. The experimental densities were then obtained from eq 3 using the y_2 and y_1 determined at the experimental conditions.

$$\rho_{exp} = \frac{m_1 + m_2}{(y_2 - y_1)A + V_0} \tag{3}$$

Accuracy. The uncertainty for the measured temperatures is ±0.05 K, and it is ±0.001 MPa for the measured pressures. The uncertainties in the position of the upper end of the cylinder and the position of the piston are ±0.001 and ±0.001 cm, respectively. Sources of error in the determination of the mass of propane are error in the HBT prediction of the density (±0.0201%), error in the predicted density due to uncertainties in temperature and pressure (±0.084 kg/m^3), error in the volume of the long-chain alkane (±50 kg/m^3 causes an error of ±0.022 cm^3 in the volume), and error in the determination of the volume of the cylinder (±0.0101 cm^3). The uncertainty in the mass of the long-chain alkane is ±0.0001 g. Thermal expansion of the cylinder was not taken into account.

The error analysis was performed by calculating first a density value assuming all the properties errorless. Then

Table 2. Measured Bubble Point Pressures of the Systems Studied

T/K	P/MPa	T/K	P/MPa	T/K	P/MPa
Propane		Propane (1) + Triacontane (2)		Propane (1) + Hexatriacontane (2)	
experiment 1		w_2 = 0.0098		w_2 = 0.0108	
343.1	2.615	353.1	3.159	353.1	3.166
353.0	3.170	363.0	3.786	363.1	3.798
363.1	3.798				
343.0	2.608	w_2 = 0.0206		w_2 = 0.0227	
		353.0	3.151	353.1	3.166
experiment 2		363.0	3.773	363.1	3.808
353.1	3.174	373.0	*a*		
363.0	3.810			w_2 =0.0308	
		w_2 = 0.0289		353.1	3.160
		353.2	3.151	363.0	3.793
		363.1	3.770		
		373.2	4.488	w_2 = 0.0319	
				353.1	3.176
				363.1	3.804
Propane (1) + Tetracontane (2)		Propane (1) + Pentacontane (2)		Propane (1) + Squalane (2)	
w_2 = 0.0096		w_2 = 0.0100		w_2 = 0.0326	
353.0	3.161	353.1	3.160	353.1	3.155
362.9	3.787	363.0	3.790	363.0	3.778
				373.1	4.509
w_2 = 0.0190		w_2 = 0.0199			
353.0	3.156	353.0	3.157		
363.0	3.784	363.1	3.793		
w_2 = 0.0318		w_2 = 0.0201			
353.0	3.153	353.0	3.158		
363.0	3.780	363.0	3.790		
		w_2 = 0.0279			
		353.1	3.153		
		363.0	3.786		

[a] Subcritical, but too much fluid in the cylinder, bubble point pressure not measured.

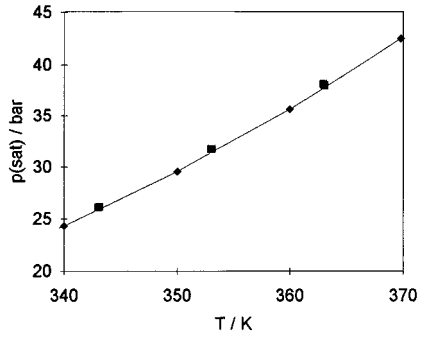

Figure 4. Vapor pressure of pure propane: comparison of literature values and this work. The symbols are as follows: ■, this work; ◆, Younglove and Ely (1987); — Reid et al. (1987).

all the uncertainties were introduced in the way that they tend to increase the calculated density. Next all the uncertainties were introduced so that the calculated density decreased. By comparing the higher and lower density values to the first one, it was concluded that the accuracy of the reported density values should be within ±0.19%. The relative error in the mass fraction was studied with the use of the same method, and it was found to be ±0.17%.

Results and Discussion

Density. Densities were measured for pure propane and propane (1) + long-chain alkane (2) with $w_2 \approx 0.01$, 0.02, and 0.03, except for propane (1) + squalane (2) where only $w_2 = 0.0326$ was studied. The densities of the samples were measured at 353, 363, and 373 K temperatures and at 4.0, 4.5, 5.0, 5.5, 6.0, 6.5, and 7.0 MPa pressures. In addition, the densities of pure propane were measured at 343 K. The densities of the systems studied are reported in Table 1. The critical temperature of pure propane is 369.85 K (Younglove and Ely, 1987), below which all the systems are in a subcritical state. At 373 K, propane (1) + squalane (2) with $w_2 = 0.0326$ and propane (1) + triacontane (2) with $w_2 = 0.0206$ and 0.0289 systems are in a subcritical state, while other systems are supercritical.

The densities obtained for pure propane agree well with values reported in the literature (Figure 2). A comparison of the mixture densities was not possible, because no density data were found for the mixtures studied. The dependence of the fluid density on w_2 (the mass fraction of the long-chain alkane) seems to be almost linear in the region studied (Figure 3). The effect of all long-chain alkanes on the density of the fluid is approximately of the same magnitude regardless of the molar mass.

Bubble Point Pressure. Bubble point pressures were measured for pure propane and the binary mixtures at the same temperatures where densities were determined. The results are listed in Table 2. The vapor pressures measured for pure propane were compared to the Wagner type vapor pressure correlation taken from Reid et al. (1987), and the measured values were on the average 0.039 MPa higher than the calculated ones (Figure 4). The vapor pressures recommended by Younglove and Ely (1987) were on the average 0.0005 MPa lower than the calculated values. However, the precision of the measurements allows a study of the effect of the long-chain alkane mass fraction on the bubble point pressure. The bubble point pressures of the propane + long-chain alkane solutions decrease only slightly when the mass fraction of the heavy alkane is increased.

Symbols

A	cross-sectional area of the cylinder
m_1	mass of propane
m_2	mass of long-chain alkane
V_0	volume correction
V_2	volume of the long-chain alkane
w_2	mass fraction of the long-chain alkane
y_1	position of the piston
y_2	position of the upper end of the cylinder
ρ_{HBT}	density predicted by HBT
ρ_2	density of the long-chain alkane

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