Phase Behavior, Densities, and Isothermal Compressibility of the CO_2 + Pentane + Acetone Ternary System in Various Phase Regions

Jiawei Chen, Weize Wu, Zhimin Liu, Tao Jiang, Jun He, Ying Huang, Guanying Yang, and Buxing Han*

Center for Molecular Sciences, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100080, China

The phase behavior and critical parameters of the CO_2 (1) + pentane (2) + acetone (3) ternary system with different compositions were determined experimentally, and their densities were measured in both the subcritical and supercritical (SC) regions. The isothermal compressibility (K_T) at some typical conditions was calculated using the density data. It is demonstrated that the density is sensitive to pressure as the pressure approaches the critical point of the mixtures, i.e, K_T is large and increases sharply. K_T also increases significantly as pressure approaches the dew point or the bubble point at other compositions close to the critical composition. However, K_T is very small and is not sensitive to pressure well above the dew point or the bubble point. When the composition is far from the critical composition, K_T has small values, and the effect of pressure on K_T is very small, even near the phase-separation point.

Introduction

Supercritical fluids (SCFs) have been used in many fields, such as extractions and fractionations,[1−3] chemical reactions,[4,5] and material processing.[2,6] It is well known that chemists have found and synthesized numerous compounds. Unfortunately, the substances that are applicable as supercritical (SC) solvents in practice are very limited because many factors have to be considered in industrial processes, such as critical temperature, critical pressure, toxicity, and price. To tune the processes effectively by changing pressure, the SC solvents should be in their high compressibility region, so that the processes can be optimized by tuning the pressure. The compressibility of a pure SCF is very large in its critical region, but it decreases as the temperature and/or pressure differs from that at the critical point. The critical parameters of a pure substance are fixed, and therefore they cannot be changed according to the requirements of the operation processes. Utilization of mixed fluids in the critical regions may be an effective way to broaden the applications of SCFs.[7] For example, one can obtain many mixed SC solvents with different properties by changing the composition of a mixed system, which may meet different requirements. Therefore, study of the properties of mixtures in their critical region is of importance to both fundamental research and practical applications.

Acetone and pentane are typical solvents. Acetone is polar, while pentane is nonpolar. The phase behaviors of CO_2 (1) + pentane (2) and CO_2 (1) + acetone (2) binary mixtures have been studied by different authors.[8−14] In a previous paper,[15] we studied the phase behavior, critical parameters, densities, and compressibility of CO_2 (1) + pentane (2) and CO_2 (1) + acetone (2) binary mixtures in different phase regions. We are also interested in more complex mixtures. In this work, we study the phase behavior and critical parameters of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures at different conditions, and the densities and compressibility of the mixed fluids are then studied systematically in both the subcritical and SC regions.

Experimental Section

Materials. CO_2, with a purity of 99.995%, was supplied by the Beijing Analytical Instrument Factory. Acetone (>99.9%) and *n*-pentane (>99.8%) were of analytical grade produced by the Beijing Chemical Reagent Plant. The chemicals were used as received.

Apparatus and Procedures. The phase behavior and the densities of the mixtures were determined by a volume tunable view cell. The details were given in our previous work.[16] The accuracies of temperature and pressure measurements were ±0.05 K and ±0.025 MPa, respectively. In a typical experiment, the air in the view cell was removed by vacuum, and a desired amount of the pentane + acetone mixture was charged. CO_2 was then added from a sample bomb. The mass of CO_2 in the view cell was known from the mass difference of the sample bomb before and after charging the system. The estimated uncertainty of the mole fraction was 0.001. The cell was placed into the water bath at the desired temperature. After thermal equilibrium had been reached, the piston in the optical cell was moved up and down to change the volume and the pressure of the system. The volume of the system was known from the position of the piston, which was calibrated accurately by using water as a medium. At the critical point, a very strong opalescence was observed and the meniscus appeared at the half-volume level after slight pressure reduction.

It was estimated that the accuracy of determined density data was better than $\pm 0.001\ g\cdot cm^{-3}$. To calculate the compressibility (K_T), we used a B-spline method to smooth the measured density data and K_T was obtained by differential calculation. It was estimated that the accuracy of the K_T data was better than ±3%.

Results and Discussion

Critical Points of the Mixtures. The critical parameters of CO_2 were measured in this work to verify the

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* To whom correspondence may be addressed. Tel.: 86-10-62562821. Fax: 86-10-62559373. E-mail: Hanbx@iccas.ac.cn.

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Table 1. Critical Temperature and Critical Pressure of CO_2 (1) + Pentane (2) + Acetone (3) Ternary Mixtures with Different Compositions

χ_2:χ_3 = 1:3			χ_2:χ_3 = 1:1			χ_2:χ_3 = 3:1		
χ_1	T_c/K	P_c/MPa	χ_1	T_c/K	P_c/MPa	χ_1	T_c/K	P_c/MPa
0.896	334.85	9.46	0.902	329.75	8.89	0.897	328.15	8.65
0.914	330.95	9.04	0.927	325.15	8.67	0.924	323.15	8.40
0.927	326.85	8.81	0.930	323.15	8.41	0.944	318.15	8.13
0.941	323.15	8.60	0.935	322.35	8.38	0.946	317.65	8.11
0.959	316.95	8.18	0.949	318.15	8.25	0.967	311.95	7.82

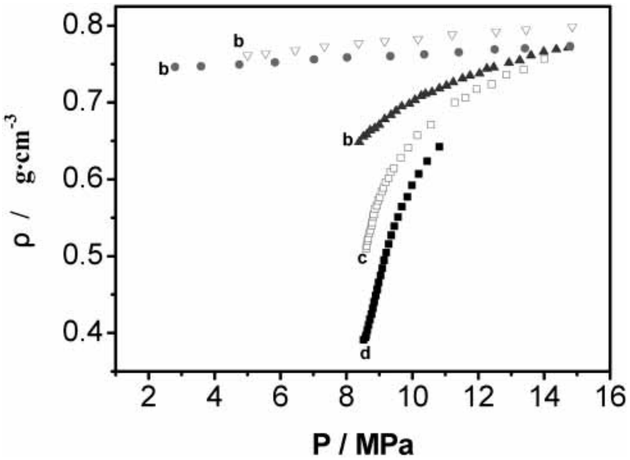

Figure 1. Dependence of the density of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 1:3) on pressures at 323.15 K. ■, χ_1 = 0.960; □, χ_1 = 0.941; ▲, χ_1 = 0.896; ▽, χ_1 = 0.603; ●, χ_1 = 0.357.

reliability of the apparatus. The results obtained of 304.25 K, 7.38 MPa, and 0.462 g·cm^{-3} agreed well with published data.[17]

In this work, we studied how the densities of the CO_2 (1) + pentane (2) + acetone (3) ternary mixtures change with composition and pressure in different phase regions. Critical parameters and phase behavior of the mixture were the basis for selecting suitable experimental conditions. The critical parameters of the mixtures with different compositions are presented in Table 1.

Densities of the Mixtures at Fixed Temperature. The densities of the mixtures were determined at 323.15 K up to 15 MPa in the SC region and subcritical regions. Tables 2−4 list the results of the CO_2 (1) + pentane (2) + acetone (3) ternary mixtures with χ_2:χ_3 = 1:3, 1:1, and 3:1, respectively. The dependence of the densities on pressure and composition at 323.15 K is also illustrated in Figures 1−3. In this paper, χ_1, χ_2, and χ_3 stand for the mole fraction of CO_2, pentane, and acetone, respectively; parts b, c, and d in all the figures and tables stand for the bubble point, critical point, and dew point, respectively.

The results in Figures 1−3 reveal that the sensitivity of density to system pressure depends on both composition and pressure at a fixed temperature. The K_T value of a fluid is a quantitative expression of the sensitivity of density to pressure, which is closely related with the structure of fluids.[18] K_T values of the mixtures have been calculated using the density data in Tables 2−4 by the following equation

$$K_T = \frac{1}{\rho}\left(\frac{\partial \rho}{\partial P}\right)_T \tag{1}$$

where ρ is the density of fluids and P is the pressure.

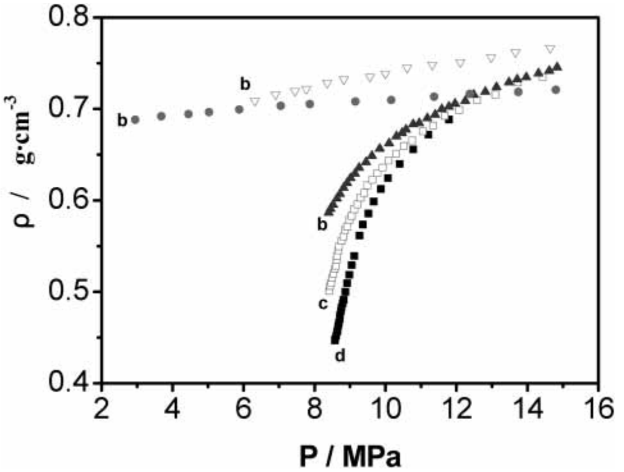

Figure 2. Dependence of the density of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 1:1) on pressures at 323.15 K. ■, χ_1 = 0.938; □, χ_1 = 0.930; ▲, χ_1 = 0.902; ▽, χ_1 = 0.728; ●, χ_1 = 0.364.

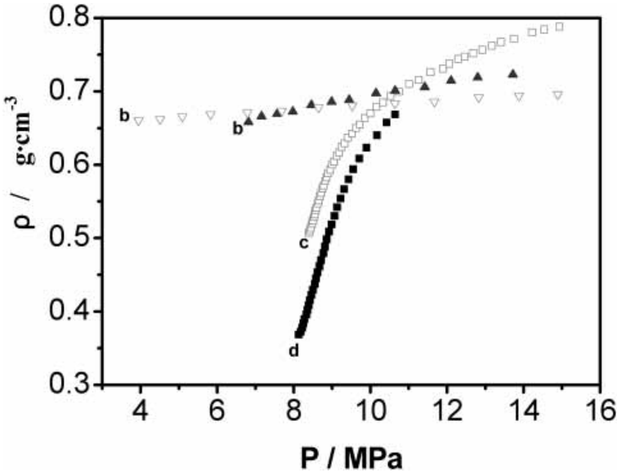

Figure 3. Dependence of the density of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 3:1) on pressures at 323.15 K. ■, χ_1 = 0.949; □, χ_1 = 0.924; ▲, χ_1 = 0.769; ▽, χ_1 = 0.454.

Figures 4−6 show the effect of pressure on the K_T value of the mixed fluid with χ_2:χ_3 = 1:3, 1:1, and 3:1 at 323.15 K. K_T is very large and sensitive to pressure as the pressure approaches the critical point of a mixture, i.e, at the critical composition, K_T increases sharply as pressure approaches the phase-separation pressure. K_T also increases significantly as pressure approaches the dew point or the bubble point at other compositions close to the critical composition. However, K_T is very small and not sensitive to pressure as pressure is well above the bubble point. The data in Figures 4−6 also illustrate that, as the composition is far from the critical composition, the effect of pressure on K_T is very limited, even near the phase-separation point. Interestingly, for CO_2 (1) + pentane (2) + acetone (3) ternary mixtures with χ_2:χ_3 = 1:3, 1:1, and 3:1, the variation of the compressibility K_T shows similar behavior, while the K_T of CO_2 (1) + pentane (2) is larger than that of CO_2 (1) + acetone (2) under the similar conditions.[15]

Effect of Temperature and Pressure on the Density at Fixed Composition. As discussed above, the critical parameters of mixtures change with composition. At fixed composition, the effect of pressure on the density at different temperatures is also interesting. Figure 7 shows the dependence of the densities of CO_2 (1) + pentane (2) + acetone (3) mixture with χ_2:χ_3 = 1:1 and χ_1 = 0.902 on pressure at different temperatures. The density is sensitive to pressure near the phase-separation pressures, especially

Table 2. Densities of CO_2 (1) + Pentane (2) + Acetone (3) Ternary Mixtures (χ_2:χ_3 = 1:3) at Different Conditions

P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}
T = 323.15 K, $\chi_1 = 0.960^{d}$													
8.52^{d}	0.391^{d}	8.67	0.410	8.84	0.440	9.04	0.475	9.28	0.516	9.68	0.564	10.45	0.624
8.58	0.394	8.71	0.417	8.89	0.448	9.09	0.484	9.36	0.527	9.85	0.577	10.82	0.642
8.60	0.397	8.76	0.425	8.93	0.457	9.14	0.494	9.45	0.539	9.99	0.592		
8.63	0.403	8.80	0.432	8.98	0.466	9.21	0.505	9.56	0.551	10.19	0.607		
T = 323.15 K, $\chi_1 = 0.941^{c}$													
8.60^{c}	0.509^{c}	8.72	0.534	8.87	0.561	9.12	0.589	9.65	0.628	11.61	0.706	14.00	0.757
8.61	0.514	8.76	0.540	8.92	0.565	9.19	0.596	9.88	0.641	11.95	0.717		
8.64	0.520	8.78	0.544	8.96	0.572	9.28	0.601	10.15	0.657	12.41	0.724		
8.66	0.523	8.81	0.551	9.01	0.576	9.34	0.609	10.56	0.671	12.85	0.736		
8.69	0.530	8.83	0.554	9.07	0.584	9.44	0.614	11.28	0.700	13.38	0.743		
T = 323.15 K, $\chi_1 = 0.896^{b}$													
8.39^{b}	0.648^{b}	9.00	0.671	9.91	0.698	10.82	0.718	12.04	0.738	13.60	0.761		
8.51	0.655	9.16	0.678	10.08	0.703	11.04	0.722	12.26	0.744	13.99	0.766		
8.62	0.658	9.35	0.684	10.27	0.709	11.24	0.726	12.47	0.746	14.37	0.769		
8.72	0.664	9.51	0.689	10.40	0.711	11.52	0.731	12.93	0.752	14.73	0.771		
8.86	0.666	9.68	0.695	10.60	0.713	11.75	0.735	13.26	0.755				
T = 323.15 K, $\chi_1 = 0.603^{b}$													
5.00^{b}	0.761^{b}	6.45	0.768	8.37	0.777	10.17	0.783	12.54	0.792	14.85	0.799		
5.55	0.764	7.33	0.773	9.16	0.780	11.21	0.788	13.45	0.795				
T = 323.15 K, $\chi_1 = 0.357^{b}$													
2.80^{b}	0.746^{b}	4.75	0.749	7.02	0.756	9.33	0.760	11.41	0.765	13.42	0.771		
3.59	0.747	5.83	0.752	8.02	0.758	10.36	0.763	12.50	0.769	14.79	0.773		
T = 318.15 K, $\chi_1 = 0.941^{b}$													
8.15^{b}	0.614^{b}	8.40	0.637	8.73	0.657	9.21	0.683	9.94	0.706	11.02	0.737	12.35	0.761
8.22	0.622	8.50	0.642	8.88	0.666	9.42	0.689	10.23	0.718	11.50	0.745		
8.30	0.628	8.61	0.651	9.03	0.672	9.66	0.700	10.62	0.725	12.08	0.757		
T = 334.85 K, $\chi_1 = 0.896^{b}$													
9.46^{b}	0.511^{b}	9.69	0.535	9.98	0.563	10.44	0.593	11.16	0.624	12.35	0.663	13.98	0.697
9.48	0.514	9.72	0.538	10.05	0.566	10.50	0.596	11.27	0.629	12.59	0.667	14.15	0.701
9.51	0.517	9.75	0.542	10.10	0.570	10.60	0.600	11.38	0.634	12.79	0.672	14.38	0.705
9.53	0.520	9.80	0.546	10.15	0.573	10.68	0.605	11.55	0.639	13.01	0.677	14.58	0.709
9.56	0.523	9.82	0.549	10.21	0.577	10.74	0.608	11.71	0.644	13.24	0.681	14.77	0.712
9.59	0.525	9.85	0.552	10.26	0.580	10.85	0.613	11.83	0.649	13.52	0.687	14.98	0.713
9.63	0.529	9.89	0.555	10.33	0.585	10.97	0.618	12.02	0.653	13.69	0.692		
9.65	0.531	9.94	0.559	10.39	0.589	11.05	0.621	12.19	0.659	13.80	0.696		
T = 328.15 K, $\chi_1 = 0.941^{d}$													
8.89^{d}	0.433^{d}	9.04	0.449	9.18	0.469	9.37	0.496	9.74	0.542	10.42	0.601	12.63	0.689
8.92	0.435	9.07	0.454	9.20	0.471	9.45	0.505	9.84	0.554	10.63	0.614	13.30	0.706
8.94	0.440	9.09	0.456	9.23	0.476	9.49	0.513	9.95	0.565	10.88	0.628	14.23	0.724
8.99	0.442	9.11	0.461	9.26	0.479	9.56	0.523	10.09	0.576	11.20	0.643		
9.01	0.447	9.14	0.464	9.32	0.487	9.64	0.533	10.23	0.589	11.56	0.658		
T = 330.95 K, $\chi_1 = 0.914^{c}$													
9.04^{c}	0.505^{c}	9.14	0.523	9.34	0.547	9.69	0.581	10.24	0.618	11.27	0.661	13.27	0.712
9.05	0.508	9.19	0.527	9.44	0.558	9.85	0.592	10.52	0.633	11.78	0.677	14.28	0.730
9.07	0.513	9.22	0.532	9.55	0.569	10.02	0.606	10.85	0.647	12.45	0.694		
9.10	0.517	9.26	0.537										
T = 326.85 K, $\chi_1 = 0.927^{c}$													
8.81^{c}	0.510^{c}	8.87	0.524	9.01	0.542	9.15	0.562	9.43	0.587	10.18	0.636	11.90	0.691
8.82	0.514	8.92	0.527	9.04	0.545	9.21	0.565	9.57	0.599	10.51	0.648	12.55	0.707
8.83	0.516	8.94	0.533	9.08	0.552	9.26	0.572	9.74	0.610	10.88	0.662	13.40	0.722
8.85	0.519	8.98	0.537	9.11	0.555	9.31	0.575	9.94	0.623	11.37	0.676	14.45	0.739
T = 316.95 K, $\chi_1 = 0.959^{c}$													
8.18^{c}	0.506^{c}	8.26	0.524	8.39	0.558	8.71	0.604	9.45	0.659	11.32	0.724		
8.20	0.510	8.27	0.528	8.45	0.569	8.83	0.617	9.78	0.674	12.19	0.743		
8.22	0.515	8.31	0.538	8.53	0.580	8.98	0.630	10.14	0.691	13.30	0.764		
8.24	0.519	8.35	0.548	8.62	0.592	9.19	0.644	10.66	0.707	14.50	0.783		

[b] Bubble point. [c] Critical point. [d] Dew point.

near the critical temperature at this composition. Similarly, the density of the fluids at pressures much higher than phase-separation pressures is not sensitive to pressure. For χ_2:χ_3 = 1:3 with $\chi_1 = 0.941$ and χ_2:χ_3 = 3:1 with $\chi_1 = 0.924$, the dependence of the density on pressure and temperature is similar, which can be seen from the data in Tables 2−4.

Effect of Pressure on the Density of the Mixtures with Critical Compositions. For a mixed system, T_c and P_c vary with composition. Figure 8 illustrates the effect of pressure on the densities of the mixed fluids (χ_2:χ_3 = 1:3) with critical compositions at different temperatures. At fixed pressure, the density depends strongly on composition and temperature, as shown clearly in the figure. However, the curves with different compositions are similar in that they change dramatically with pressure near the critical point, although their temperatures and composition are

Table 3. Densities of CO_2(1) + Pentane(2) + Acetone(3) Ternary Mixtures (χ_2:χ_3=1:1) at Different Conditions

P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}
T = 323.15 K, χ_1 = 0.938^{d}													
8.57^{d}	0.447^{d}	8.65	0.459	8.72	0.475	8.82	0.491	9.05	0.529	9.52	0.586	10.40	0.640
8.58	0.448	8.67	0.463	8.74	0.479	8.87	0.500	9.12	0.539	9.67	0.599	10.78	0.656
8.61	0.452	8.69	0.467	8.76	0.483	8.92	0.509	9.27	0.561	9.87	0.613	11.21	0.672
8.63	0.456	8.71	0.471	8.79	0.487	8.98	0.519	9.36	0.574	10.07	0.624	11.79	0.688
T = 323.15 K, χ_1 = 0.930^{c}													
8.41^{c}	0.501^{c}	8.60	0.528	8.81	0.560	9.21	0.596	9.92	0.636	11.34	0.682	14.43	0.735
8.44	0.506	8.62	0.535	8.86	0.567	9.30	0.603	10.09	0.643	11.67	0.692		
8.47	0.510	8.63	0.539	8.92	0.571	9.40	0.608	10.30	0.651	12.08	0.699		
8.49	0.515	8.67	0.545	8.98	0.579	9.49	0.616	10.52	0.660	12.58	0.710		
8.53	0.520	8.69	0.550	9.05	0.583	9.63	0.622	10.75	0.665	13.10	0.716		
8.57	0.525	8.76	0.556	9.12	0.590	9.77	0.630	11.05	0.675	13.73	0.729		
T = 323.15 K, χ_1 = 0.902^{b}													
8.40^{b}	0.586^{b}	8.82	0.614	9.45	0.642	10.47	0.674	11.40	0.694	12.49	0.716	13.97	0.735
8.45	0.591	8.92	0.619	9.62	0.649	10.58	0.677	11.59	0.700	12.81	0.719	14.31	0.739
8.52	0.595	9.02	0.624	9.84	0.657	10.78	0.683	11.79	0.702	13.13	0.724	14.62	0.742
8.63	0.602	9.14	0.629	10.10	0.662	10.95	0.685	11.96	0.706	13.48	0.729	14.84	0.746
8.72	0.607	9.26	0.636	10.31	0.670	11.18	0.690	12.25	0.709	13.71	0.732		
T = 323.15 K, χ_1 = 0.728^{b}													
6.32^{b}	0.709^{b}	7.45	0.719	8.36	0.729	9.56	0.735	10.61	0.745	12.11	0.751	13.66	0.762
6.90	0.716	7.77	0.722	8.83	0.732	10.00	0.739	11.32	0.748	12.98	0.757	14.64	0.767
T = 323.15 K, χ_1 = 0.364^{b}													
2.95^{b}	0.688^{b}	4.45	0.694	5.88	0.699	7.87	0.705	10.16	0.710	12.38	0.716	14.80	0.721
3.68	0.692	5.03	0.697	7.05	0.703	9.15	0.708	11.37	0.713	13.74	0.719		
T = 328.15 K, χ_1 = 0.902^{b}													
8.81^{b}	0.512^{b}	9.05	0.541	9.42	0.574	9.93	0.606	10.77	0.640	12.19	0.675	14.18	0.709
8.83	0.515	9.09	0.544	9.48	0.578	10.00	0.609	10.88	0.642	12.38	0.679	14.32	0.712
8.85	0.518	9.12	0.549	9.51	0.580	10.07	0.613	11.02	0.645	12.58	0.684	14.49	0.715
8.87	0.520	9.16	0.551	9.55	0.584	10.13	0.615	11.14	0.649	12.77	0.686	14.70	0.717
8.90	0.523	9.19	0.555	9.62	0.588	10.22	0.617	11.26	0.654	12.98	0.692	14.97	0.720
8.93	0.527	9.22	0.558	9.66	0.590	10.30	0.621	11.40	0.655	13.20	0.696		
8.96	0.530	9.29	0.561	9.71	0.593	10.37	0.626	11.53	0.660	13.40	0.698		
8.98	0.532	9.32	0.565	9.76	0.597	10.45	0.628	11.68	0.665	13.58	0.699		
9.00	0.535	9.36	0.568	9.84	0.601	10.58	0.631	11.80	0.667	13.77	0.702		
9.02	0.538	9.39	0.571	9.86	0.603	10.68	0.636	12.02	0.671	13.99	0.708		
T = 329.75 K, χ_1 = 0.902^{c}													
8.89^{c}	0.483^{c}	9.15	0.515	9.49	0.550	10.10	0.593	11.10	0.635	12.50	0.671	14.23	0.701
8.91	0.489	9.18	0.520	9.54	0.556	10.22	0.599	11.20	0.638	12.70	0.674	14.46	0.706
8.95	0.492	9.22	0.523	9.60	0.561	10.35	0.604	11.34	0.642	12.91	0.679	14.74	0.710
8.98	0.496	9.26	0.529	9.66	0.567	10.48	0.612	11.52	0.648	13.09	0.684	15.04	0.715
9.00	0.498	9.30	0.533	9.74	0.571	10.61	0.616	11.70	0.654	13.26	0.685		
9.04	0.502	9.35	0.539	9.82	0.577	10.74	0.621	11.88	0.656	13.47	0.691		
9.08	0.506	9.38	0.541	9.91	0.582	10.82	0.624	12.08	0.661	13.70	0.695		
9.10	0.511	9.42	0.546	10.00	0.588	10.94	0.628	12.29	0.667	13.93	0.698		
T = 333.15 K, χ_1 = 0.902^{d}													
9.05^{d}	0.434^{d}	9.25	0.457	9.54	0.489	9.92	0.529	10.58	0.573	11.86	0.626	13.54	0.669
9.07	0.436	9.29	0.459	9.58	0.494	9.97	0.533	10.67	0.579	12.09	0.632	13.71	0.671
9.09	0.438	9.31	0.463	9.62	0.497	10.03	0.538	10.71	0.584	12.33	0.640	13.86	0.675
9.12	0.440	9.35	0.466	9.65	0.503	10.09	0.542	10.91	0.590	12.58	0.645	14.10	0.681
9.14	0.443	9.37	0.470	9.70	0.507	10.16	0.547	11.03	0.595	12.74	0.650	14.28	0.683
9.15	0.445	9.40	0.474	9.72	0.511	10.23	0.552	11.18	0.602	12.88	0.654	14.47	0.685
9.17	0.447	9.43	0.477	9.79	0.515	10.32	0.558	11.32	0.607	13.07	0.657	14.72	0.690
9.19	0.450	9.48	0.481	9.83	0.520	10.40	0.562	11.47	0.613	13.23	0.662		
9.22	0.452	9.50	0.486	9.88	0.523	10.50	0.568	11.65	0.619	13.37	0.665		
T = 318.15 K, χ_1 = 0.902^{b}													
7.83^{b}	0.640^{b}	8.67	0.671	9.32	0.691	10.34	0.713	11.55	0.733	13.00	0.752	14.80	0.770
7.95	0.644	8.82	0.676	9.54	0.697	10.54	0.716	11.84	0.734	13.30	0.754		
8.12	0.653	8.91	0.680	9.70	0.701	10.74	0.719	12.16	0.739	13.61	0.757		
8.29	0.658	9.02	0.682	9.89	0.703	11.04	0.723	12.42	0.744	13.97	0.760		
8.51	0.666	9.18	0.685	10.07	0.706	11.27	0.729	12.69	0.748	14.48	0.767		
T = 325.15 K, χ_1 = 0.927^{c}													
8.67^{c}	0.474^{c}	8.78	0.491	8.89	0.508	9.09	0.538	9.53	0.584	10.45	0.638	12.79	0.706
8.69	0.478	8.81	0.496	8.92	0.515	9.19	0.549	9.70	0.597	10.85	0.654	13.87	0.725
8.71	0.482	8.83	0.499	8.95	0.518	9.30	0.561	9.90	0.611	11.37	0.670		
8.74	0.487	8.86	0.505	9.02	0.528	9.40	0.572	10.15	0.623	11.99	0.687		
T = 318.15 K, χ_1 = 0.949^{c}													
8.25^{c}	0.487^{c}	8.34	0.512	8.59	0.562	9.17	0.622	10.85	0.696	12.86	0.745		
8.27	0.491	8.38	0.521	8.67	0.572	9.36	0.636	11.18	0.708	13.40	0.752		
8.28	0.495	8.43	0.531	8.75	0.584	9.62	0.650	11.53	0.714	14.01	0.764		
8.30	0.501	8.49	0.542	8.87	0.596	9.94	0.665	11.87	0.726	14.67	0.770		
8.31	0.504	8.53	0.552	9.00	0.609	10.34	0.680	12.29	0.731				

[b] Bubble point. [c] Critical point. [d] Dew point.

Table 4. Densities of CO_2 (1) + Pentane (2) + Acetone (3) Ternary Mixtures (χ_2:χ_3 = 3:1) at Different Conditions

P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}	P/MPa	ρ/g·cm^{-3}
T = 323.15 K, χ_1 = 0.949^{d}													
8.13^{d}	0.369^{d}	8.33	0.396	8.50	0.430	8.71	0.470	8.99	0.519	9.43	0.580	10.42	0.658
8.18	0.372	8.36	0.402	8.55	0.437	8.77	0.479	9.06	0.530	9.55	0.594	10.64	0.669
8.22	0.378	8.39	0.409	8.58	0.445	8.81	0.488	9.13	0.542	9.71	0.608		
8.25	0.384	8.42	0.416	8.62	0.453	8.86	0.498	9.22	0.554	9.90	0.623		
8.28	0.390	8.46	0.423	8.67	0.462	8.92	0.509	9.31	0.567	10.17	0.640		
T = 323.15 K, χ_1 = 0.924^{c}													
8.40^{c}	0.507^{c}	8.59	0.541	8.85	0.583	9.33	0.629	10.33	0.685	12.29	0.744	14.54	0.784
8.42	0.511	8.63	0.547	8.89	0.588	9.41	0.637	10.53	0.694	12.46	0.747	14.93	0.788
8.45	0.514	8.65	0.551	8.95	0.593	9.52	0.642	10.75	0.699	12.68	0.752		
8.48	0.520	8.68	0.557	9.00	0.600	9.63	0.650	11.01	0.710	12.92	0.756		
8.50	0.523	8.71	0.562	9.05	0.604	9.72	0.655	11.26	0.715	13.17	0.762		
8.53	0.528	8.75	0.567	9.12	0.612	9.86	0.664	11.57	0.726	13.41	0.767		
8.55	0.532	8.78	0.572	9.18	0.617	10.00	0.670	11.90	0.731	13.75	0.772		
8.57	0.538	8.82	0.578	9.25	0.624	10.16	0.678	12.06	0.738	14.22	0.780		
T = 323.15 K, χ_1 = 0.769^{b}													
6.82^{b}	0.658^{b}	7.57	0.670	8.46	0.681	9.45	0.689	10.65	0.701	12.11	0.714	13.72	0.723
7.16	0.666	7.98	0.672	8.99	0.686	10.15	0.697	11.42	0.706	12.80	0.719		
T = 323.15 K, χ_1 = 0.454^{b}													
3.95^{b}	0.660^{b}	5.09	0.666	6.79	0.671	8.65	0.678	10.64	0.684	12.83	0.692	14.89	0.696
4.51	0.662	5.83	0.669	7.69	0.674	9.53	0.681	11.67	0.686	13.88	0.694		
T = 328.15 K, χ_1 = 0.924^{d}													
8.64^{d}	0.425^{d}	8.84	0.454	9.08	0.489	9.38	0.532	9.85	0.582	10.75	0.642	12.90	0.717
8.65	0.427	8.85	0.458	9.10	0.494	9.40	0.537	9.90	0.588	10.89	0.650	13.26	0.726
8.67	0.429	8.89	0.461	9.13	0.497	9.46	0.541	9.98	0.592	11.02	0.656	13.65	0.734
8.69	0.433	8.91	0.465	9.16	0.502	9.50	0.546	10.05	0.600	11.18	0.663	14.06	0.744
8.71	0.435	8.93	0.468	9.19	0.506	9.54	0.550	10.14	0.605	11.37	0.670	14.50	0.753
8.73	0.439	8.96	0.472	9.22	0.511	9.57	0.557	10.21	0.612	11.57	0.678	14.73	0.754
8.75	0.440	8.98	0.475	9.26	0.514	9.63	0.561	10.31	0.616	11.79	0.685		
8.78	0.445	9.00	0.479	9.29	0.519	9.68	0.567	10.41	0.624	12.03	0.694		
8.80	0.448	9.03	0.482	9.32	0.523	9.72	0.571	10.51	0.630	12.29	0.701		
8.82	0.451	9.05	0.487	9.35	0.528	9.79	0.577	10.64	0.637	12.58	0.709		
T = 308.15 K, χ_1 = 0.924^{b}													
6.89^{b}	0.740^{b}	7.29	0.755	7.68	0.768	8.21	0.782	8.91	0.795	9.71	0.810	10.72	0.826
6.98	0.746	7.36	0.761	7.88	0.774	8.41	0.786	9.09	0.800	10.03	0.817	11.16	0.829
7.15	0.750	7.50	0.764	8.02	0.779	8.64	0.790	9.37	0.804	10.41	0.822	11.21	0.832
T = 313.15 K, χ_1 = 0.924^{b}													
7.44^{b}	0.694^{b}	7.77	0.711	8.32	0.732	9.14	0.758	10.19	0.782	11.76	0.805	13.59	0.831
7.49	0.697	7.90	0.715	8.51	0.739	9.36	0.764	10.58	0.787	12.30	0.816		
7.57	0.700	8.01	0.720	8.69	0.746	9.62	0.768	10.91	0.795	12.76	0.823		
7.65	0.704	8.14	0.727	8.96	0.751	9.93	0.776	11.24	0.802	13.28	0.825		
T = 318.15 K, χ_1 = 0.924^{b}													
8.05^{b}	0.612^{b}	8.37	0.642	8.90	0.679	9.87	0.716	11.08	0.750	12.49	0.777	14.30	0.802
8.09	0.617	8.46	0.650	9.06	0.685	10.15	0.727	11.33	0.755	12.76	0.782	14.75	0.810
8.16	0.624	8.55	0.655	9.22	0.694	10.48	0.734	11.60	0.763	13.08	0.786	15.00	0.818
8.23	0.630	8.65	0.663	9.40	0.700	10.58	0.737	11.85	0.765	13.51	0.792		
8.30	0.637	8.76	0.669	9.63	0.710	10.79	0.744	12.16	0.771	14.00	0.801		
T = 317.65 K, χ_1= 0.946^{c}													
8.11^{c}	0.499^{c}	8.25	0.532	8.56	0.593	9.54	0.679	11.00	0.733	12.56	0.768	15.05	0.810
8.13	0.503	8.29	0.541	8.64	0.605	9.70	0.685	11.20	0.738	12.88	0.775		
8.15	0.506	8.33	0.552	8.73	0.617	10.06	0.700	11.39	0.745	13.21	0.783		
8.17	0.511	8.38	0.561	8.85	0.630	10.27	0.710	11.70	0.751	13.77	0.789		
8.19	0.514	8.42	0.571	9.09	0.650	10.50	0.717	11.92	0.754	14.10	0.796		
8.22	0.523	8.49	0.582	9.30	0.664	10.75	0.727	12.20	0.763	14.44	0.803		
T = 311.95 K, χ_1 = 0.967^{c}													
7.82^{c}	0.495^{c}	7.89	0.522	8.00	0.560	8.20	0.606	8.63	0.660	9.81	0.723	11.40	0.771
7.83	0.501	7.90	0.531	8.04	0.571	8.27	0.620	8.84	0.674	10.11	0.736	11.93	0.779
7.84	0.506	7.94	0.541	8.08	0.583	8.35	0.631	9.05	0.689	10.39	0.743		
7.86	0.513	7.97	0.551	8.14	0.593	8.48	0.645	9.36	0.705	10.93	0.758		

^{b} Bubble point. ^{c} Critical point. ^{d} Dew point.

different. This is one of the features of fluids in the critical region. For χ_2:χ_3 = 1:1 and 3:1, the similar phenomenon exists, which can be seen from the data in Tables 2−4.

The Density of the Mixed Fluids at Phase-Separation Points in Different Phase Regions. Figure 9 shows the densities of the mixed fluids at the bubble points, dew points, and critical points at 323.15 K. The ratios χ_2:χ_3 are 1:3, 1:1, and 3:1, respectively.

Obviously, for the subcritical fluids far from critical points, the densities of the mixtures are not sensitive to χ_1 at fixed χ_2:χ_3 ratios. At a given χ_1, the density of the mixtures increases with increasing χ_3 or the concentration

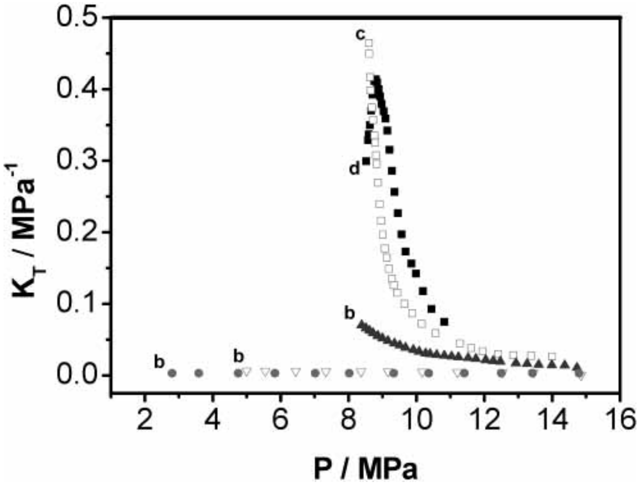

Figure 4. Isothermal compressibility K_T of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 1:3) on pressures at 323.15 K. ■, $\chi_1 = 0.960$; □, $\chi_1 = 0.941$; ▲, $\chi_1 = 0.896$; ●, $\chi_1 = 0.357$.

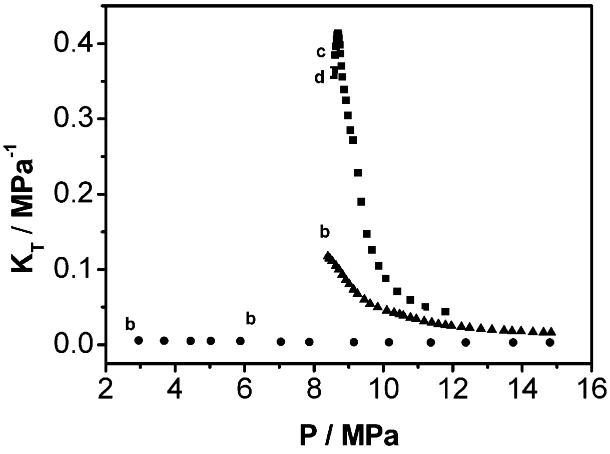

Figure 5. Isothermal compressibility K_T of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 1:1) on pressures at 323.15 K. ■, $\chi_1 = 0.938$; □, $\chi_1 = 0.930$; ▲, $\chi_1 = 0.902$; ▽, $\chi_1 = 0.728$; ●, $\chi_1 = 0.364$.

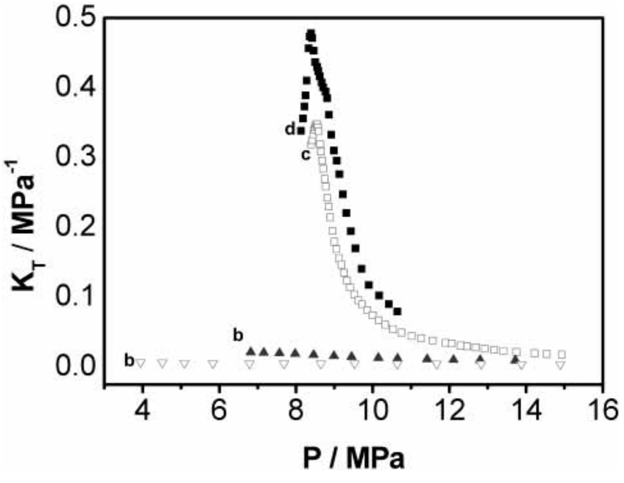

Figure 6. Isothermal compressibility K_T of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 3:1) on pressures at 323.15 K. ■, $\chi_1 = 0.949$; □, $\chi_1 = 0.924$; ▲, $\chi_1 = 0.769$; ▽, $\chi_1 = 0.454$.

of acetone. The main reason is that the subcritical fluids far from the critical composition are more similar to the liquid mixtures, and the density of acetone is larger than that of pentane.

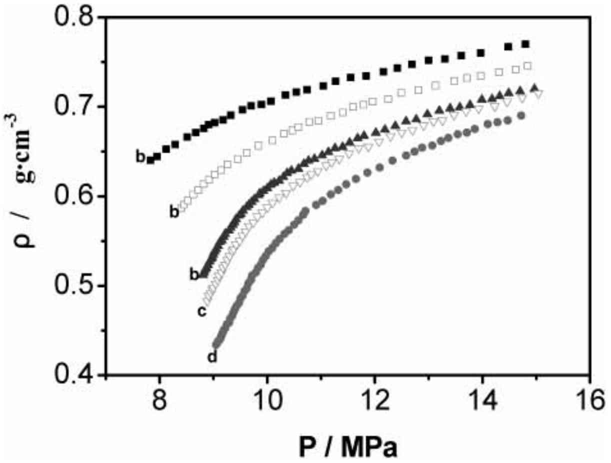

Figure 7. Dependence of the density of CO_2 (1) + pentane (2) + acetone (3) mixtures (χ_2:χ_3 = 1:1) with $\chi_1 = 0.902$ on temperature and pressure in the critical region. ■, T = 318.15 K; □, T = 323.15 K; ▲, T = 328.15 K; ▽, T = 329.75 K; ●, T = 333.15 K.

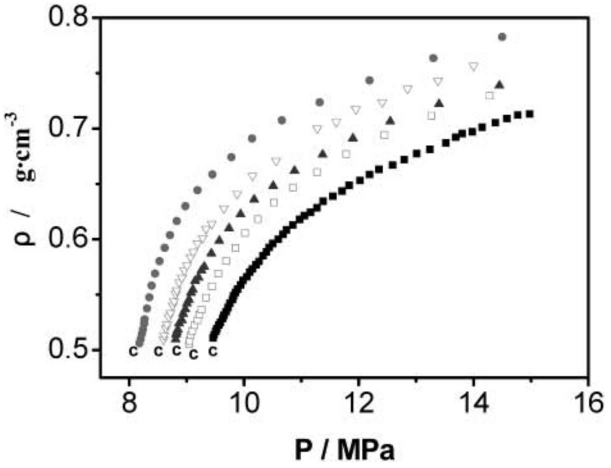

Figure 8. Effect of pressure on the density of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures (χ_2:χ_3 = 1:3) with critical composition. ■, $\chi_1 = 0.896$, T = 334.85 K; □, $\chi_1 = 0.914$, T = 330.95 K; ▲, $\chi_1 = 0.927$, T = 326.85 K; ▽, $\chi_1 = 0.941$, T = 323.15 K; ●, $\chi_1 = 0.959$, T = 316.95 K.

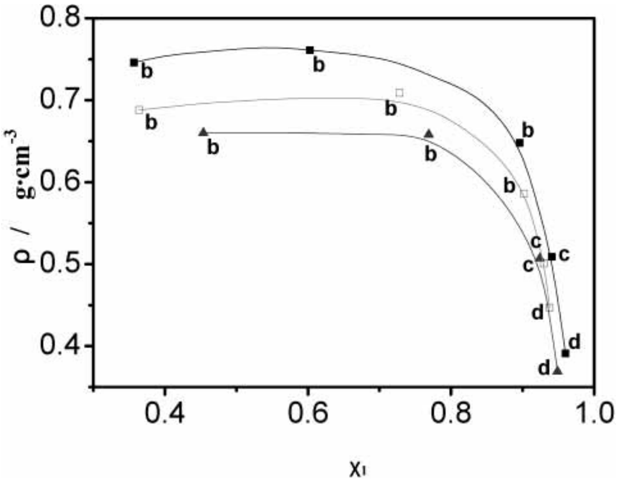

Figure 9. Density of CO_2 (1) + pentane (2) + acetone (3) ternary mixtures at phase-separation points in different phase regions at 323.15 K. ■, χ_2:χ_3 = 1:3; □, χ_2:χ_3 = 1:1; ▲, χ_2:χ_3 = 3:1.

It is interesting that, for all the mixtures with different χ_2:χ_3 ratios, the density is very sensitive to χ_1 near the critical composition and decreases dramatically with increasing χ_1. This suggests that the density and density-dependent properties of the mixed fluids near the critical point can be tuned effectively by varying composition of the mixtures.

Conclusion

Study on the density and compressibility of CO_2 (1) + pentane (2) + acetone (3) in different phase regions reveals that the density is sensitive to pressure near the critical points of the mixtures, i.e., the isothermal compressibility (K_T) is large and sensitive to pressure as the pressure approaches the critical point, dew point, or bubble point in the critical region, and K_T is largest at the phase-separation point at a fixed composition. As pressure is much higher than the phase-separation pressure or the composition is far from the critical composition, K_T is very small and the effect of pressure on K_T is very limited. To tune the properties of a mixed fluid effectively by changing pressure, both the composition and pressure should be close to the critical point of the mixture.

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