

Volumetric Properties of Sulfur Hexafluoride + Pentane and Sulfur Hexafluoride + Toluene at High Pressures

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The volumetric properties of sulfur hexafluoride + pentane and sulfur hexafluoride + toluene mixtures have been determined at 323, 348, 373, 398, and 423 K at pressures up to 70 MPa using a variable-volume view cell. Densities for pure components and mixtures containing 90, 80, 70, 50, 40, and 30% by mass sulfur hexafluoride are reported as a function of pressure at each temperature.

Introduction

This paper is a continuation of our ongoing investigation of the volumetric properties of binary fluid mixtures at high pressures (Gökmenoglu et al., 1996; Kiran et al., 1996; Pöhler and Kiran 1996, 1997). We have already reported on the densities of binary mixtures of carbon dioxide with pentane, toluene, sulfur hexafluoride, acetone, and ethanol. Binary fluid mixtures at high pressure are often encountered in supercritical fluid processing. Mixture composition, along with pressure and temperature are used to adjust the properties of the fluid to bring about miscibility or phase separation or changes in viscosity, diffusivity, or reactivity in a variety of applications, such as those encountered in polymer processing ranging from reactions and polymerization to particle formation or foaming (Kiran, 1994, 1995).

Sulfur hexafluoride has interesting properties. It has a fairly low critical temperature ($T_c = 318.69$ K) and critical pressure ($P_c = 3.759$ MPa) (Berg and Wagner, 1990). In this respect, it is similar to carbon dioxide ($T_c = 304.15$; $P_c = 7.38$ MPa), but it displays much higher densities (Gökmenoglu et al., 1996). Furthermore, compared to carbon dioxide, SF₆ is a more effective solvent for many polymers at high pressures. For example, mixtures of SF₆ and carbon dioxide have been used to improve the solubility of polystyrene and to extend the precipitation threshold in the supercritical polymerization of styrene (Kiran and Gökmenoglu, 1966). The high density of pure SF₆ is an important feature that can be used to prepare fluid mixtures with a wide range of density values with low critical temperatures, and we have been exploring such mixtures for unique applications such as "isopycnic" or "levitation processing" (Kiran and Gökmenoglu, 1966).

Limited information is available on mixtures of sulfur hexafluoride with other fluids. For sulfur hexafluoride + pentane, vapor–liquid equilibria are reported at pressures below 4.2 MPa and temperatures below 340 K (Berg and Wagner, 1990). Phase equilibrium information for sulfur hexafluoride + propane along with critical data (Clegg and Rowlinson, 1955) and the gas–liquid critical temperatures (without data on critical pressures) for mixtures of SF₆ with toluene (Christou and Young, 1991) have also been reported.

We have recently determined the volumetric properties of sulfur hexafluoride + carbon dioxide mixtures up to 70 MPa (Gökmenoglu et al., 1996). We now present similar data for the sulfur hexafluoride + pentane and the sulfur

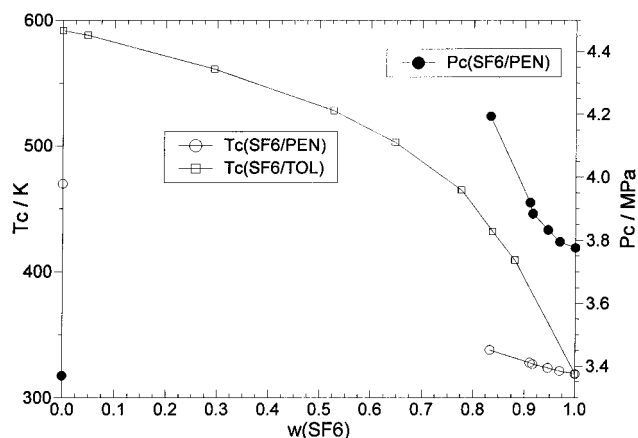


Figure 1. Critical temperature and pressure of the binary mixtures sulfur hexafluoride + pentane (data from Berg and Wagner (1990)) and critical temperatures for binary mixtures of sulfur hexafluoride + toluene [data from Christou et al. (1991)].

hexafluoride + toluene mixtures at pressures up to 70 MPa.

Experimental Section

Sulfur hexafluoride was instrument grade (Scott Specialty Gases) with a purity of 99.99%. Pentane (99.9% purity) and toluene (99.8% purity) were obtained from Aldrich and Fisher Chemicals, respectively.

A detailed description of the experimental system (a variable-volume view cell with a linear variable differential transformer (LVDT) to monitor the position of the piston, and hence the internal volume) and procedures for determination of the densities are given in our previous publications (Kiran et al., 1996; Gökmenoglu et al., 1996). Pressures and temperatures in the cell are determined with an accuracy of ± 0.03 MPa and ± 0.5 K, respectively. About 15 to 18 g of the fluid is charged to the cell. The mass loading of the cell is determined using a sensitive balance with an accuracy of ± 0.01 g. Volume changes are determined with an accuracy of ± 0.0025 cm³. The maximum internal volume of the cell is 22.43 ± 0.05 cm³. The relative error in density determinations (estimated as $\Delta\rho/\rho = [(\Delta m/m)^2 + (\Delta V/V)^2]^{1/2}$ where m is the mass and V is the volume) is about 0.125%. As demonstrated by comparisons with literature data in our previous publications (Kiran et al., 1996; Gökmenoglu et al., 1996), the densities are determined with an accuracy of $\pm 1.2\%$.

Results and Discussion

Sulfur Hexafluoride + Pentane. Table 1 summarizes the density data for sulfur hexafluoride and pentane and

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Table 1. Densities of Sulfur Hexafluoride (1) + Pentane (2) (w = Mass Fraction)

323 K		348 K		373 K		398 K		423 K	
p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$
$w_1 = 1$									
4.03	0.6128	5.66	0.6166	7.13	0.6163	8.65	0.6155	10.09	0.6150
4.06	0.7913	5.89	0.6681	7.48	0.6590	9.12	0.6537	10.70	0.6530
4.06	0.8198	6.11	0.7213	7.85	0.7038	9.74	0.7036	11.46	0.6982
4.12	0.8422	6.35	0.7797	8.27	0.7541	10.31	0.7509	12.27	0.7464
4.17	0.8954	6.13	0.7960	8.19	0.7954	10.56	0.7950	12.85	0.7940
4.26	0.9281	6.58	0.8284	8.77	0.8106	11.03	0.8028	13.27	0.8017
4.28	0.9549	6.36	0.8451	8.65	0.8441	11.26	0.8428	13.84	0.8427
4.37	1.0122	6.89	0.8899	9.44	0.8772	11.99	0.8673	14.56	0.8657
4.51	1.0195	6.67	0.9017	9.16	0.8947	12.13	0.8984	15.13	0.8999
4.54	1.0800	7.08	0.9641	9.91	0.9550	13.22	0.9394	16.19	0.9389
4.72	1.0929	7.46	0.9765	10.35	0.9568	13.38	0.9666	16.72	0.9636
5.15	1.1854	7.61	1.0321	10.91	1.0242	15.03	1.0315	18.42	1.0263
6.64	1.2953	8.40	1.0815	11.68	1.0515	14.92	1.0370	18.77	1.0341
10.71	1.4360	8.41	1.1026	12.39	1.1046	17.15	1.1204	21.36	1.1070
		9.76	1.1894	14.73	1.1962	20.61	1.2149	25.07	1.1910
		12.21	1.2918	18.83	1.3100	26.31	1.3297	30.82	1.2925
		17.30	1.4141	24.74	1.4188	33.69	1.4312	39.20	1.3998
$w_1 = 0.89$									
3.13	0.7201	4.59	0.7193	6.83	0.7174	9.40	0.7143	11.94	0.7132
3.83	1.0590	6.18	0.9425	8.38	0.8434	11.57	0.8281	13.66	0.7820
6.05	1.1525	7.23	0.9971	10.00	0.9240	13.61	0.9025	15.59	0.8442
7.89	1.1886	10.34	1.0949	11.68	0.9849	15.45	0.9537	19.36	0.9379
9.81	1.2218	12.29	1.1335	13.57	1.0338	19.21	1.0318	23.03	1.0022
11.50	1.2421	13.64	1.1571	15.47	1.0722	23.02	1.0884	26.78	1.0550
13.49	1.2663	15.57	1.1857	19.29	1.1321	26.95	1.1352	30.04	1.0931
14.89	1.2814	19.24	1.2285	23.25	1.1814	30.50	1.1726	34.52	1.1375
		23.68	1.2713	26.80	1.2166	34.41	1.2065	38.17	1.1699
				30.57	1.2473	38.20	1.2347	41.93	1.1984
								45.86	1.2246
$w_1 = 0.8$									
4.23	0.9977	9.74	0.9656	8.26	0.8172	10.31	0.7495	14.24	0.7477
7.77	1.0422	12.54	0.9980	11.71	0.8956	15.81	0.8677	19.15	0.8352
11.77	1.0772	15.94	1.0304	15.37	0.9477	19.58	0.9173	23.55	0.8894
15.44	1.1030	19.15	1.0553	19.34	0.9880	23.27	0.9542	26.99	0.9233
19.50	1.1273	23.16	1.0807	23.31	1.0203	27.34	0.9879	30.96	0.9555
23.16	1.1456	26.82	1.1012	27.00	1.0458	30.91	1.0124	34.48	0.9806
26.79	1.1620	30.83	1.1213	30.82	1.0676	34.52	1.0330	38.27	1.0031
30.70	1.1785	34.56	1.1385	34.60	1.0877	38.36	1.0545	42.25	1.0252
35.46	1.1963	38.28	1.1534	38.23	1.1042	42.20	1.0737	45.90	1.0435
		42.01	1.1674	41.94	1.1200	45.95	1.0902	49.78	1.0604
		45.80	1.1800	45.98	1.1364	49.81	1.1062	53.72	1.0779
				49.90	1.1505	53.48	1.1201	57.18	1.0906
				53.46	1.1629	56.99	1.1329	61.17	1.1057
						60.93	1.1458	64.77	1.1172
						65.08	1.1585		
$w_1 = 0.64$									
4.19	0.8825	8.59	0.8476	15.41	0.8385	18.32	0.8032	26.11	0.8035
7.77	0.9036	11.56	0.8664	19.31	0.8592	23.21	0.8315	30.71	0.8254
11.53	0.9201	15.41	0.8865	23.17	0.8777	26.86	0.8490	34.60	0.8422
15.57	0.9364	19.16	0.9032	26.99	0.8939	30.68	0.8659	38.54	0.8580
19.22	0.9495	23.05	0.9188	30.62	0.9060	34.50	0.8807	42.28	0.8719
23.13	0.9612	26.76	0.9317	34.18	0.9182	38.31	0.8933	45.21	0.8807
26.89	0.9728	30.57	0.9432	38.31	0.9305	42.05	0.9054	49.72	0.8942
30.62	0.9826	34.52	0.9540	42.40	0.9424	45.86	0.9165	53.50	0.9058
34.58	0.9924	38.66	0.9650	45.95	0.9511	49.89	0.9278	57.31	0.9153
38.12	1.0008	42.06	0.9740	49.56	0.9603	53.55	0.9374	61.60	0.9264
41.69	1.0072	45.69	0.9827	53.37	0.9688	57.11	0.9456	65.14	0.9343
45.73	1.0165	49.41	0.9909	57.17	0.9777	60.95	0.9547		
49.29	1.0234	53.10	0.9985	60.98	0.9842	64.74	0.9620		
52.11	1.0288	57.29	1.0072	64.82	0.9921				
		60.14	1.0121						
$w_1 = 0.4$									
4.25	0.7632	11.75	0.7472	17.48	0.7295	28.12	0.7302	34.05	0.7182
8.02	0.7765	15.44	0.7598	20.74	0.7397	30.88	0.7385	38.57	0.7318
11.60	0.7871	19.42	0.7701	23.42	0.7472	34.62	0.7470	42.15	0.7390
15.51	0.7975	23.17	0.7800	26.99	0.7570	38.43	0.7579	45.95	0.7496
19.28	0.8054	27.04	0.7899	30.73	0.7676	42.07	0.7657	49.65	0.7574
23.19	0.8142	30.68	0.7974	34.62	0.7768	46.11	0.7745	53.53	0.7651
26.67	0.8209	34.75	0.8053	38.44	0.7849	49.78	0.7816	57.17	0.7723
31.18	0.8294	38.43	0.8125	42.27	0.7921	53.60	0.7884	61.17	0.7805
34.64	0.8350	41.99	0.8197	46.10	0.7993	57.15	0.7948	64.67	0.7868
38.28	0.8417	45.96	0.8252	49.68	0.8056	61.30	0.8019		
42.13	0.8475	49.64	0.8314	53.56	0.8120	64.68	0.8073		

Table 2. Densities of Sulfur Hexafluoride (1) + Toluene (2) (w = Mass Fraction)

323 K		348 K		373 K		398 K		423 K	
p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$
$w_1 = 0.87$									
3.53	0.6785	5.21	0.6764	6.77	0.6781	8.29	0.6795	10.19	0.6802
3.57	0.7164	5.73	0.8540	6.98	0.7140	8.61	0.7162	10.96	0.7200
3.59	0.7582	5.97	0.9141	7.20	0.7554	9.06	0.7616	11.92	0.7635
3.64	0.8144	6.25	0.9637	7.49	0.8033	9.83	0.8100	13.16	0.8121
3.66	0.8685	6.80	1.0043	7.89	0.8058	11.06	0.8691	14.88	0.8669
3.71	0.9302	7.36	1.0376	8.85	0.9188	12.95	0.9355	17.37	0.9307
3.80	1.0015	7.79	1.0594	10.37	0.9920	15.07	0.9899	21.07	1.0003
4.12	1.0856	8.54	1.0871	13.50	1.0738	18.99	1.0624	23.79	1.0421
5.21	1.1574	9.62	1.1180	18.15	1.1461	24.28	1.1313	27.01	1.0823
		11.51	1.1598					30.90	1.1235
$w_1 = 0.81$									
3.59	0.6795	5.18	0.6785	6.67	0.6800	8.08	0.6802	9.53	0.6832
3.61	0.7162	5.28	0.7162	6.91	0.7190	8.44	0.7183	10.06	0.7270
3.63	0.7522	5.40	0.7571	7.17	0.7615	8.96	0.7738	10.62	0.7748
3.64	0.7967	5.51	0.8049	7.48	0.8084	9.47	0.8231	11.60	0.8274
3.68	0.8494	5.67	0.8570	7.88	0.8612	10.18	0.8835	13.45	0.8866
3.74	0.9033	5.92	0.9193	8.45	0.9208	11.33	0.9454	15.78	0.9391
3.82	0.9682	6.52	0.9904	9.61	0.9924	14.67	1.0186	19.61	1.0012
4.03	1.0395	7.99	1.0651	12.61	1.0713	19.51	1.0839	23.24	1.0450
5.06	1.1274	11.65	1.1522	15.68	1.1194	23.13	1.1215	27.08	1.0835
9.68	1.2227	15.51	1.2005	19.74	1.1602	26.95	1.1532	30.86	1.1166
		19.61	1.2340	23.12	1.1929	30.68	1.1800	34.59	1.1434
				26.73	1.2181	34.67	1.2057	38.33	1.1672
								42.07	1.1883
								44.43	1.2008
$w_1 = 0.73$									
3.63	0.7599	5.32	0.7592	6.92	0.7633	8.43	0.7632	9.88	0.7656
3.65	0.7954	5.46	0.8000	7.21	0.8075	8.83	0.8040	11.58	0.8682
3.67	0.8372	5.59	0.8455	7.53	0.8519	9.32	0.8491	15.66	0.9638
3.72	0.8845	5.76	0.8931	8.01	0.9040	10.06	0.9015	19.64	1.0119
3.78	0.9397	6.10	0.9496	8.83	0.9621	11.18	0.9593	23.29	1.0464
3.91	1.0142	6.82	1.0124	10.60	1.0284	13.90	1.0228	26.92	1.0730
4.45	1.0873	8.82	1.0795	15.07	1.0965	18.67	1.0744	30.83	1.0995
7.52	1.1646	11.49	1.1279	19.11	1.1343	23.15	1.1099	34.66	1.1214
11.73	1.2038	15.47	1.1700	22.94	1.1628	27.04	1.1357	38.28	1.1403
15.58	1.2447	19.09	1.2005	26.70	1.1851	30.47	1.1530	41.99	1.1580
19.28	1.2663	23.25	1.2264	30.98	1.2082	34.23	1.1736	46.07	1.1765
23.09	1.2856	26.83	1.2452	34.64	1.2249	38.27	1.1902	49.93	1.1901
27.17	1.3030	30.78	1.2636	38.38	1.2414	42.22	1.2084	53.42	1.2049
		34.41	1.2791	42.22	1.2545	45.74	1.2213	57.03	1.2184
		38.36	1.2922	46.01	1.2688	49.68	1.2359	61.32	1.2315
						53.15	1.2475	64.90	1.2432
						57.02	1.2594		
$w_1 = 0.5$									
3.63	0.6574	4.56	0.6575	5.51	0.6582	6.33	0.6585	7.23	0.6595
3.67	0.6892	4.75	0.6905	5.83	0.6890	6.79	0.6922	7.79	0.6926
3.70	0.7263	4.94	0.7276	6.15	0.7270	7.26	0.7281	8.41	0.7284
3.72	0.8051	5.16	0.7702	6.52	0.7668	7.80	0.7687	9.19	0.7697
3.75	0.8502	5.40	0.8158	6.97	0.8117	8.53	0.8149	10.35	0.8178
3.79	0.9024	5.68	0.8674	7.65	0.8656	9.75	0.8662	11.37	0.8710
3.87	0.9663	6.36	0.9245	8.67	0.9263	11.79	0.9316	15.63	0.9010
4.20	1.0321	9.29	0.9863	12.05	0.9720	15.57	0.9480	19.44	0.9195
8.22	1.0510	12.44	1.0109	15.72	0.9902	19.37	0.9625	23.17	0.9356
12.00	1.0648	15.60	1.0245	19.48	1.0035	22.97	0.9744	26.95	0.9491
15.51	1.0764	19.37	1.0387	23.22	1.0148	26.92	0.9872	30.98	0.9624
19.28	1.0884	23.37	1.0504	27.22	1.0241	30.81	0.9983	34.49	0.9729
23.10	1.0973	27.12	1.0601	30.71	1.0339	34.44	1.0071	38.07	0.9823
26.97	1.1052	30.78	1.0695	34.43	1.0422	38.31	1.0175	41.91	0.9920
30.73	1.1052	34.53	1.0772	38.32	1.0508	42.10	1.0256	45.74	1.0012
32.92	1.1107	38.37	1.0868	42.07	1.0584	46.02	1.0347	49.90	1.0115
		42.29	1.0946	46.08	1.0666	49.84	1.0425	53.38	1.0187
				49.47	1.0729	53.23	1.0486	57.18	1.0275
								61.05	1.0336
								64.69	1.0404
$w_1 = 0$									
3.42	0.8396	10.73	0.8249	17.70	0.8104	25.08	0.7982	32.35	0.7875
8.23	0.8438	14.26	0.8272	21.44	0.8143	28.55	0.8026	35.58	0.7902
11.95	0.8460	16.68	0.8294	24.16	0.8170	31.58	0.8056	39.11	0.7941
15.75	0.8506	19.54	0.8320	26.90	0.8207	34.88	0.8089	42.05	0.7978
19.61	0.8524	23.53	0.8353	30.48	0.8237	38.41	0.8111	45.81	0.8016
23.43	0.8552	27.01	0.8384	34.51	0.8263	42.13	0.8159	49.59	0.8040
26.91	0.8580	30.55	0.8412	38.34	0.8306	46.11	0.8183	53.59	0.8081
30.87	0.8617	34.44	0.8445	42.25	0.8339	49.81	0.8213	57.18	0.8110

Table 2 (Continued)

323 K		348 K		373 K		398 K		423 K	
p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$	p/MPa	$\rho/\text{g cm}^{-3}$
34.60	0.8646	38.38	0.8475	46.26	0.8361	53.95	0.8250	60.91	0.8145
38.37	0.8660	42.50	0.8518	49.65	0.8398	57.43	0.8286	64.46	0.8174
42.00	0.8699	45.94	0.8532	53.34	0.8420	60.83	0.8305		
45.92	0.8715	49.71	0.8561	57.09	0.8445	64.74	0.8333		
49.59	0.8747	53.46	0.8597	60.93	0.8472				
53.60	0.8763	57.23	0.8612	64.58	0.8507				
57.33	0.8797	61.39	0.8643						
61.07	0.8818	64.55	0.8662						
64.77	0.8831								

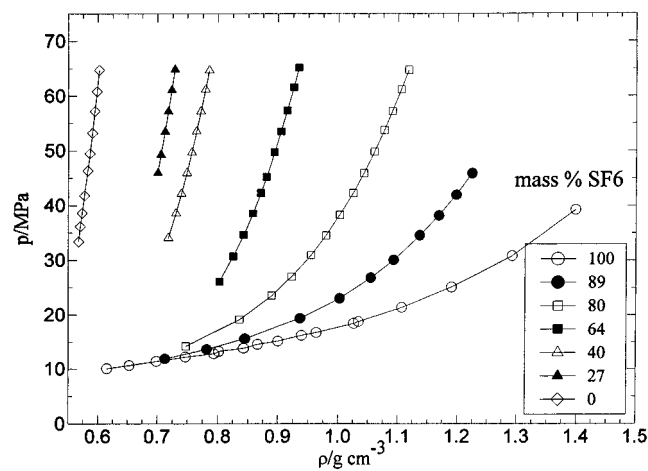


Figure 4. Pressure dependence of density for the binary mixtures of sulfur hexafluoride and pentane at 423 K (compositions in mass percent).

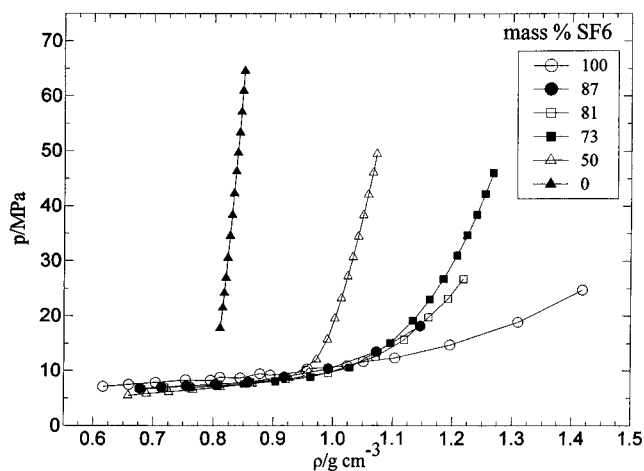


Figure 6. Pressure dependence of density for the binary mixtures of sulfur hexafluoride and toluene at 373 K (compositions in mass percent).

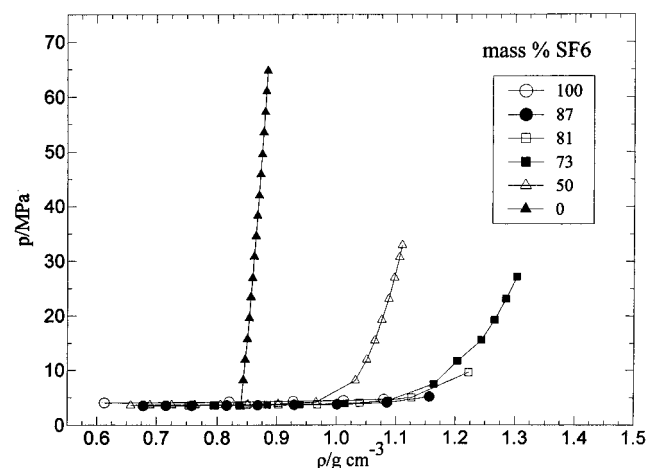


Figure 5. Pressure dependence of density for the binary mixtures of sulfur hexafluoride and toluene at 323 K (compositions in mass percent).

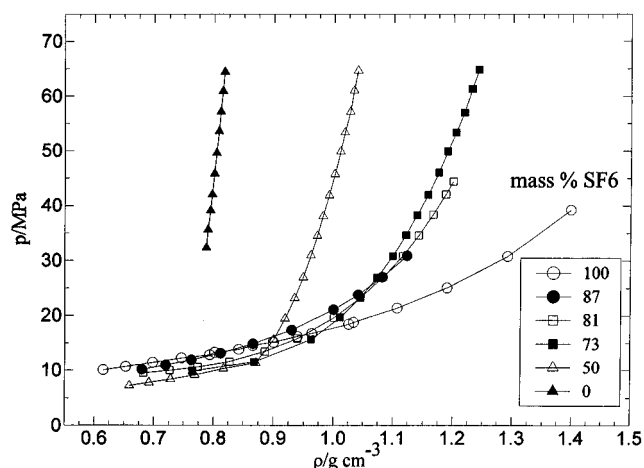


Figure 7. Pressure dependence of density for the binary mixtures of sulfur hexafluoride and toluene at 423 K (compositions in mass percent).

about 0.6 to 1.5 g/cm³ are readily achievable. The compressibility of the mixtures becomes less with the increasing amount of pentane in the system.

Sulfur Hexafluoride + Toluene. The limited data on the critical temperatures of these mixtures are also included in Figure 1. Unlike mixtures of SF₆ with pentane, even with small additions of toluene, the critical temperature shows a rapid rise with this mixture. Table 2 summarizes the density data for pure sulfur hexafluoride and toluene and their binary mixtures containing 50, 73, 81, and 87 mass % sulfur hexafluoride which were also measured at 323, 348, 373, 398, and 423 K at pressures up to 70 MPa. The compositions with higher than 50% toluene were not evaluated at this time. Figures 5–7 show

the variation of density with pressure for these mixtures at 323, 373, and 423 K. For these systems, densities ranging from about 0.8 to 1.4 g/cm³ are readily achievable. Compressibility becomes less with the increasing amount of toluene in the system.

Conclusions

A wide range of densities can be obtained and further changed with pressure or temperature using binary mixtures of sulfur hexafluoride with pentane or toluene. A density range of about 0.6 to 1.5 g/cm³ is displayed in a pressure range up to 70 MPa.

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