The corresponding van't Hoff parameters are given in the last column of Table IV. [The solubility data in terms of temperature and % P_2O_5 concentration are given by % $CaSO_4$ = 2.58353564 + 0.01479136t − 0.08054436(% P_2O_5) + 0.00054433(% P_2O_5)^2 − 0.00014460t(% P_2O_5).]

Finally, it should be noted that eqs 1, 2, 4, 5, and 7 give good estimates of the solubility of $CaSO_4 \cdot 0.5H_2O$ only when employed within the range of the experimental variables (Tables I–III and Table I of ref 1). They cannot be employed for extrapolations outside these ranges. Moderate extrapolations of the data may be made by using the van't Hoff equation.

Glossary

t	temperature, °C
R^2	correlation coefficient
ΔH	apparent heat of solution at saturation, cal/mol
C	integration constant
T	absolute temperature, K
R	gas constant, 1.9872 cal/(deg·mol)
K_{sp}	apparent solubility product constant, (% Ca)(% SO_4)
ΔS	apparent entropy of solution at saturation, cal/(deg·mol)

Literature Cited

(1) Sullivan, J. M.; Kohler, J. J.; Grinstead, J. H., Jr. J. Chem. Eng. Data 1988, 33, 367–370.
(2) Kremenetskaya, E. V.; Petropavlovskii, I. A.; Karmyshov, V. F.; Spirdonova, I. A. Khim. Promst. (Moscow) 1987, 19 (3), 31–33.
(3) Shpunt, S. Ya.; Shul'gina, N. N.; Guseva, Z. I. Zh. Prikl. Khim. (Leningrad) 1987, 40 (6), 1236–1242.
(4) Glazyrina, L. N.; Savinkova, E. I.; Grinevich, A. V.; Akhmedova, L. E. Zh. Prikl. Khim. (Leningrad) 1980, 53 (11), 2524–2527.
(5) Glazyrina, L. N.; Grinevich, A. V.; Savinkova, E. I.; Danilyuk, O. G.; Gubanova, G. V. Zh. Prikl. Khim. (Leningrad) 1980, 53 (10), 2327–2330.

High-Pressure Binary Vapor–Liquid Equilibrium Measurements of Carbon Dioxide with Aromatic Hydrocarbons

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High-pressure binary vapor–liquid equilibria of carbon dioxide with anisole and methylanisole isomers were determined at 333, 363, and 393 K and pressures up to 20 MPa. A circulation-type apparatus was built and tested by comparing the measured vapor–liquid equilibrium data of the carbon dioxide–toluene system at 393 K with literature values. In addition to temperature, pressure, and phase compositions, the vapor- and liquid-phase densities were measured for the binary systems of carbon dioxide with anisole and methylanisole isomers.

Introduction

The design and operation of separation processes requires knowledge of vapor–liquid equilibrium data. In particular, high-pressure phase equilibrium data for systems containing aromatic hydrocarbons will be needed to design and operate coal processing plants.

In this study, we have measured binary-phase equilibria of systems containing aromatic hydrocarbons (anisole and methylanisole isomers) as one of the components and carbon dioxide as the other component.

A circulation-type apparatus was built to measure temperature, pressure, vapor- and liquid-phase compositions, and vapor- and liquid-phase densities and was tested by comparing the vapor–liquid equilibrium data for the carbon dioxide–toluene system at 393 K with those of Ng and Robinson (1). This comparison showed good agreement. After testing the experimental apparatus, vapor–liquid equilibrium data for carbon

dioxide–anisole, carbon dioxide–2-methylanisole, carbon dioxide–3-methylanisole, and carbon dioxide–4-methylanisole were measured at 333, 363, and 393 K and pressures ranging up to the mixture critical pressures.

Kim et al. (2) reported solubility data for carbon dioxide–anisole at 343.2 and 372.3 K and pressure ranging up to 16.8 MPa. However, no data are reported in the literature for carbon dioxide–2-methylanisole, carbon dioxide–3-methylanisole, and carbon dioxide–4-methylanisole systems.
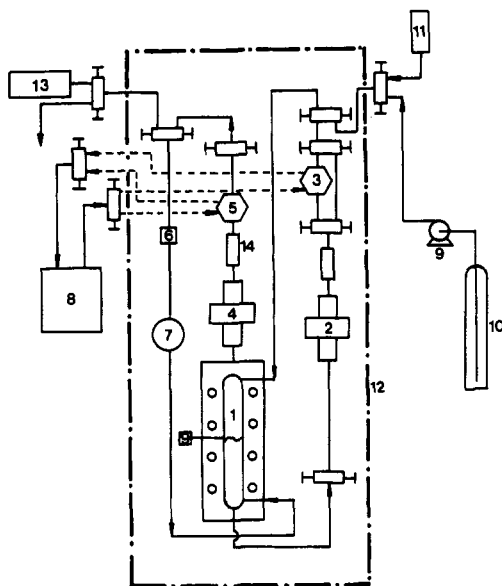
Experimental Section

Apparatus and Procedure. Vapor- and liquid-phase densities, as well as vapor and liquid equilibrium compositions, can be measured at various temperatures, ranging from ambient temperature to 420 K, and various pressures, ranging from ambient pressure to 68 MPa, by using the apparatus shown in Figure 1.

The equilibrium cell, made of stainless steel type-316, has an internal volume of 50 cm³ and is equipped with two glass windows. The vapor and liquid phases of the cell are circulated to obtain a rapid equilibration by using the magnetically operated pumps. Equilibrium is usually obtained in about 10 min. As shown in Figure 1, the vapor phase is drawn from the top of the cell and returns through the bottom right port, bubbling through the liquid phase. The liquid phase leaves the cell at the bottom and enters through the port at the top right.

Vapor and liquid samples can be injected directly into a Hewlett-Packard gas chromatograph through sampling valves. A Rheodyne six-port sampling valve with an external sampling loop of 20 μL is used to sample the vapor phase. The liquid sampling valve is another Rheodyne six-port valve with an internal loop disk volume of 1 μL. The vapor and liquid samples are transported from the sampling valves to the gas chroma-

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Figure 1. Schematic diagram of experimental apparatus: 1, equilibrium cell; 2, liquid circulation pump; 3, liquid sampling valve; 4, vapor circulation pump; 5, vapor sampling valve; 6, relief valve; 7, pressure gauge; 8, gas chromatograph; 9, metering pump; 10, carbon dioxide; 11, liquid reservoir; 12, air bath; 13, vacuum pump; 14, check valve.

tograph by helium carrier gas and are analyzed by using a thermal conductivity detector. The column, which is 7.4 m long with a $1/_4$-in.-o.d. stainless steel tube, is packed with Chromosorb W, containing 10 wt % diethylene glycol adipate.

The equilibrium cell, circulation pumps, sampling valves, and most of the connection valves are contained in a temperature-controlled air bath. Heating is supplied by 3-kW electrical heaters, and the air-bath temperature is controlled to about ±0.1 K. In order to operate the experimental apparatus without opening the air bath, the stems of the sampling valves and the connection valves are extended and handles are located outside the air bath. The temperature of the system is measured with a chromel–alumel thermocouple (K type), which is located inside the equilibrium cell. The estimated error in the temperature measurement is about ±0.2 K. The pressure in the equilibrium cell is measured by using a Heise gauge (0–40 MPa). The maximum error of this gauge is ±0.5% of the full scale reading.

Since the thermal conductivity detector responds differently to different compounds, it is necessary to determine its response factors. Calibration curves are constructed by injecting various known amounts of the pure components and determining the corresponding peak areas with use of the vapor sampling valve, the liquid sampling valve, and an SGE syringe. The sampling volumes of the vapor and liquid sampling valves were determined in a manner suggested by McHugh (*3*). The vapor sampling loop was pressurized with carbon dioxide to three different pressures of 14.0, 18.0, and 25.0 MPa, measured with Heise gauge at 310.1 K. The carbon dioxide in the sampling loop was then vented into a graduated tube filled with water at a known temperature. The volume of carbon dioxide was determined by the volume of water displaced. Therefore, the volume of the vapor sampling loop was equal to the molar volume of carbon dioxide at the bath temperature and pressure multiplied by the number of moles of carbon dioxide vented from the sampling loop determined from the volume of water displaced by using the ideal gas equation of state. The molar volume of carbon dioxide was interpolated by using the data obtained from Vargaftik (*4*). The volume of the 20-µL external sampling loop of the vapor sampling valve was determined to be 23.52 µL. From calibration curves for supercritical carbon dioxide prepared from the vapor sampling valve and liquid
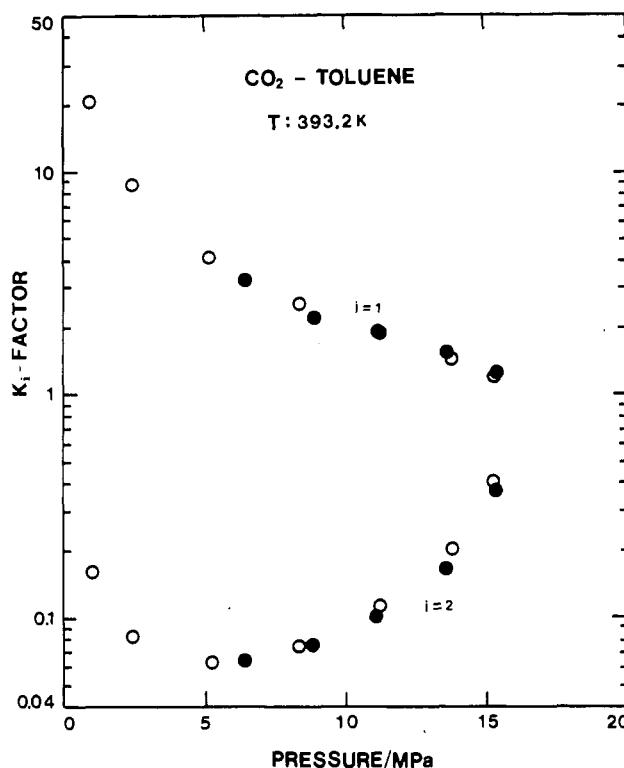
Figure 2. Comparison of K_i factors for the carbon dioxide–toluene system: ●, this work; O, Ng and Robinson (*1*).

sampling valve, the 1-µL internal sampling loop of the liquid sampling valve was determined to be 1.334 µL. The relationship between the peak area and the number of moles was fitted to a second-order polynomial for carbon dioxide and was found to be linear for all the aromatic hydrocarbons tested. Using peak areas along with the calibrated sampling loop volumes allows determination of compositions and densities for both phases. The liquid-phase mole fractions usually were reproducible to within ±0.001, while the vapor-phase mole fractions were reproducible to ±0.002.

Materials. The carbon dioxide with a minimum purity of 99.9% used in this study was supplied by Anjon Gas Co. The anisole, 2-methylanisole, 3-methylanisole, and 4-methylanisole, all of 99% minimum purity, were obtained from Aldrich Chemical. None of these chemicals were further purified since gas chromatographic analysis indicated no detectable impurities.

Results and Discussion

The accuracy of this newly built equilibrium apparatus was tested by comparing measured vapor–liquid equilibrium data for the carbon dioxide–toluene system with those of Ng and Robinson (*1*) at 393 K. For this system, K_i factors are plotted in Figure 2. As shown in Figure 2, the K_i factors at 393 K compare well with those of Ng and Robinson (*1*).

Binary data for the carbon dioxide–anisole system at 333.2, 363.3, and 393.2 K and pressures ranging from 4.1 to 18.3 MPa were measured. The vapor–liquid equilibrium data for the carbon dioxide–2-methylanisole system were measured at 333.3, 363.2, and 393.2 K and pressures ranging from 6.4 to 19.2 MPa. For the carbon dioxide–3-methylanisole system, measurements were carried out at 333.2, 363.1, and 393.1 K and pressures ranging from 6.2 to 18.9 MPa. Phase equilibria for the carbon dioxide–4-methylanisole system were measured at 333.2, 363.2, and 393.1 K with pressures ranging from 6.3 to 18.7 MPa. The experimental measurements for the binaries are tabulated in Table I. In addition to measuring the temperature, pressure, and vapor- and liquid-phase compositions, the vapor- and liquid-phase densities are determined and also

Table I. Phase Equilibrium Data for Binary Systems

Carbon Dioxide (1)-Anisole (2)

pressure/ MPa	CO₂ mole fraction liquid	CO₂ mole fraction vapor	K_i factors $i = 1$	K_i factors $i = 2$	density/ (10^3 kg m⁻³) liquid	density/ (10^3 kg m⁻³) vapor
			T = 333.2 K			
6.200	0.4627	0.9952	2.1509	0.0089	0.8292	0.1462
8.300	0.5916	0.9953	1.6824	0.0115	0.8746	0.2291
10.80	0.8434	0.9883	1.1718	0.0747	0.7784	0.4345
11.18	0.8917	0.9836	1.1031	0.1514	0.7289	0.5051
			T = 363.3 K			
4.125	0.2330	0.9922	4.2584	0.0101	0.8174	0.0743
6.865	0.3632	0.9921	2.7316	0.0124	0.8545	0.1400
9.400	0.4766	0.9901	2.0774	0.0189	0.8915	0.2120
12.03	0.5844	0.9846	1.6848	0.0371	0.9258	0.3289
13.51	0.6641	0.9783	1.4731	0.0646	0.8838	0.3911
15.01	0.7398	0.9686	1.3093	0.1207	0.8591	0.5171
			T = 393.2 K			
6.275	0.2658	0.9818	3.6938	0.2048	0.8549	0.1082
9.275	0.3679	0.9823	2.6700	0.0280	0.8887	0.1747
12.34	0.4708	0.9781	2.0775	0.0414	0.8943	0.2607
15.28	0.5819	0.9674	1.6625	0.0780	0.8709	0.3683
18.33	0.7020	0.9511	1.3548	0.1641	0.8178	0.4979

Carbon Dioxide (1)-2-Methylanisole (2)

pressure/ MPa	CO₂ mole fraction liquid	CO₂ mole fraction vapor	K_i factors $i = 1$	K_i factors $i = 2$	density/ (10^3 kg m⁻³) liquid	density/ (10^3 kg m⁻³) vapor
			T = 333.3 K			
8.155	0.5357	0.9973	1.8617	0.0058	0.8452	0.1908
9.163	0.5892	0.9960	1.6904	0.0097	0.8827	0.2445
10.57	0.6739	0.9871	1.4648	0.0396	0.8677	0.3469
11.28	0.7380	0.9831	1.3321	0.0645	0.8353	0.4621
11.64	0.7801	0.9787	1.2546	0.0969	0.8000	0.4984
			T = 363.2 K			
6.425	0.3254	0.9952	3.0584	0.0071	0.7955	0.1146
9.380	0.4557	0.9935	2.1802	0.0119	0.8405	0.1966
12.33	0.5845	0.9889	1.6919	0.0267	0.8411	0.3011
15.09	0.7011	0.9758	1.3918	0.0810	0.8369	0.4429
16.15	0.7587	0.9597	1.2649	0.1670	0.7948	0.5398
			T = 393.2 K			
6.550	0.2699	0.9902	3.6688	0.0134	0.7677	0.1073
9.430	0.3686	0.9890	2.6831	0.0174	0.8063	0.1758
12.35	0.4603	0.9848	2.1395	0.0282	0.8414	0.2570
15.25	0.5717	0.9767	1.7084	0.0544	0.8344	0.3308
17.87	0.6638	0.9620	1.4492	0.1130	0.8236	0.4403
19.23	0.7152	0.9539	1.3364	0.1619	0.8143	0.5097

Carbon Dioxide (1)-3-Methylanisole (2)

pressure/ MPa	CO₂ mole fraction liquid	CO₂ mole fraction vapor	K_i factors $i = 1$	K_i factors $i = 2$	density/ (10^3 kg m⁻³) liquid	density/ (10^3 kg m⁻³) vapor
			T = 333.2 K			
6.250	0.4687				0.7861	
8.230	0.5976	0.9970	1.6683	0.0075	0.8213	0.2082
9.775	0.7135	0.9897	1.3871	0.0360	0.8099	0.2892
10.63	0.7539	0.9844	1.3057	0.0634	0.8008	0.3199
11.17	0.8070	0.9789	1.2130	0.1093	0.7851	0.3622
			T = 363.1 K			
6.025	0.3381	0.9955	2.9444	0.0068	0.7985	0.1096
9.030	0.4724	0.9944	2.1050	0.0106	0.8269	0.1925
11.94	0.5944	0.9903	1.6660	0.0239	0.8360	0.3012
14.66	0.7263	0.9826	1.3529	0.0636	0.8135	0.4413
16.04	0.7935	0.9704	1.2226	0.1448	0.7750	0.5471
			T = 393.1 K			
6.275	0.2791	0.9897	3.5460	0.0143	0.7833	0.1020
9.100	0.3820	0.9897	2.5908	0.0167	0.8121	0.1573
12.30	0.4891	0.9856	2.0171	0.0282	0.8315	0.2368
15.28	0.5875	0.9781	1.6649	0.0531	0.8298	0.3268
18.88	0.7112	0.9563	1.3446	0.1513	0.7966	0.4363

Carbon Dioxide (1)-4-Methylanisole (2)

pressure/ MPa	CO₂ mole fraction liquid	CO₂ mole fraction vapor	K_i factors $i = 1$	K_i factors $i = 2$	density/ (10^3 kg m⁻³) liquid	density/ (10^3 kg m⁻³) vapor
			T = 333.2 K			
8.163	0.5867	0.9974	1.7000	0.0063	0.8040	0.1839
9.188	0.6492	0.9964	1.5348	0.0103	0.8269	0.2319
10.16	0.7232	0.9892	1.3678	0.0390	0.8070	0.2993
11.25	0.8043	0.9843	1.2238	0.0802	0.8076	0.4095
11.57	0.8768	0.9787	1.1162	0.1727	0.6306	0.4793
			T = 363.2 K			
6.550	0.3631	0.9957	2.7422	0.0068	0.7529	0.1117
9.325	0.4773	0.9942	2.0830	0.0111	0.8026	0.1764
12.46	0.6113	0.9891	1.6180	0.0280	0.8078	0.2769
14.51	0.6947	0.9732	1.4009	0.0878	0.7984	0.3690
16.00	0.7714	0.9645	1.2503	0.1553	0.7725	0.4547
			T = 393.1 K			
6.325	0.2912	0.9912	3.4038	0.0124	0.7271	0.0951
9.163	0.3861	0.9904	2.5651	0.0156	0.7729	0.1516
12.60	0.4942	0.9854	1.9879	0.0289	0.8143	0.2288
15.50	0.5948	0.9771	1.6427	0.0565	0.8146	0.3184
18.71	0.7035	0.9556	1.3584	0.1497	0.7921	0.4433

tabulated in Table I. Although density data are readily available for pure compounds, few density measurements are reported for mixtures. The phase density data and the phase compositions reported in this work can be used to rigorously test equations of state and mixing rules.

Shown in Figures 3 and 4 are a plot of vapor- and liquid-phase compositions and a plot of vapor- and liquid-phase densities as a function of pressure for the carbon dioxide–anisole system. As seen in Figure 3, as the temperature increases, the solubility of carbon dioxide in the liquid phase decreases and the solubility of anisole in the carbon dioxide phase increases at constant pressure. Figure 4 shows that both the vapor- and liquid-phase densities are smooth functions of pressure and approach the mixture critical density as the pressure is increased. As the pressure is increased, the vapor-phase density increases, while the liquid-phase density is constant, except near the critical point even though the liquid-phase composition depends strongly on pressure, as shown in Figure 3.

The vapor- and liquid-phase compositions of the carbon dioxide–anisole system are compared with the values of Kim et al. (2), and agreement is good. Comparisons also are made for densities of the vapor and liquid phases. There is reasonable agreement in vapor-phase densities; however, there are large differences in liquid-phase densities. We believe these differences are due to subtle differences in the procedures for
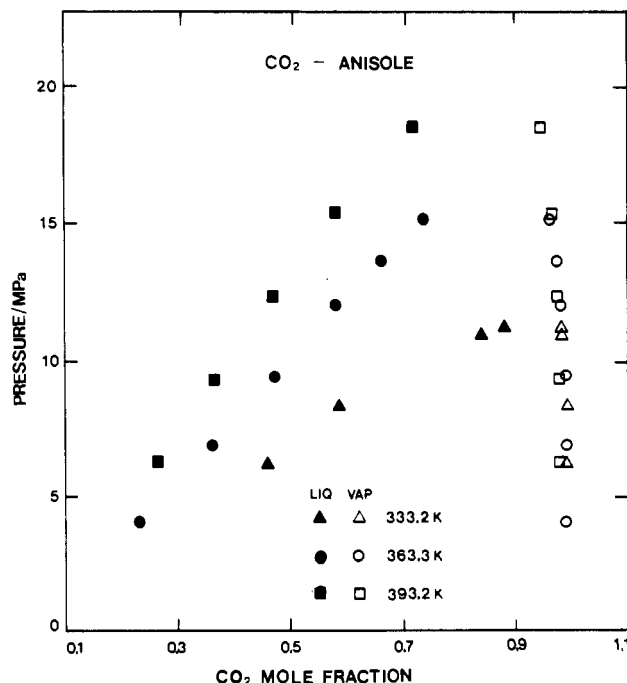
Figure 3. Liquid- and vapor-phase CO₂ mole fractions as a function of pressure for the carbon dioxide–anisole system.
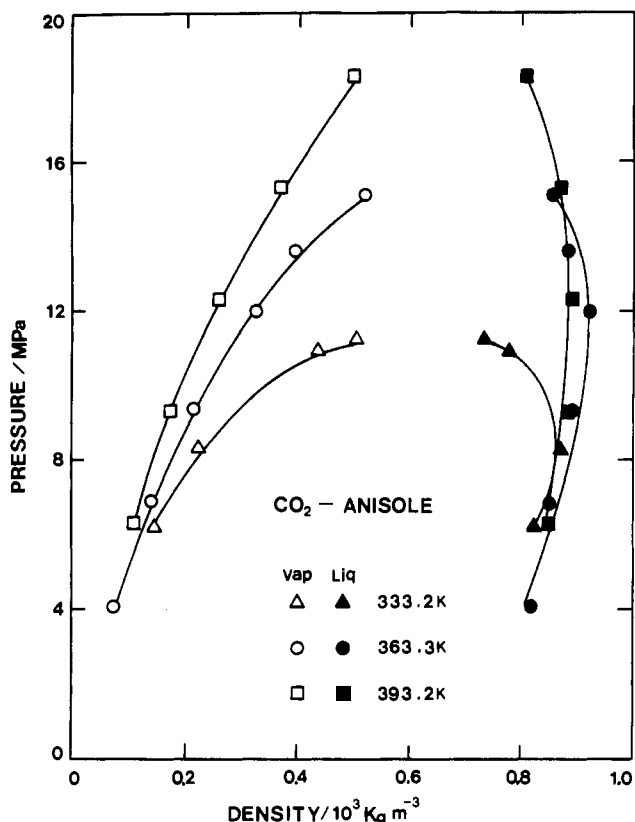
Figure 4. Measured vapor- and liquid-phase densities as a function of pressure for the carbon dioxide–anisole system.

calibrating the volumes of the sampling loops. While these differences in the calibration procedure do not affect the accuracy of the composition measurements or the accuracy of the vapor-phase densities, the differences in the results for liquid-phase densities suggest that there are inherent limitations in the accuracy that is obtainable for liquid-phase densities. In this work, the vapor- and liquid-phase densities of the mixture were calculated from known values of concentration, molecular weights of the components, and the sampling loop volumes. In this experiment, the vapor sampling loop volume was initially calibrated with the method of McHugh (*3*). Then, the liquid sampling loop volume and syringe volume were calibrated with this calibrated vapor sampling loop volume. Kim et al. (*2*) also calibrated their liquid sampling loop volume from their vapor sampling loop volume. However, this required extrapolation of data taken with the vapor sampling loop down to lower values of peak areas, which may have introduced errors in their calibration. In this work, we have extended the calibration curve for our system to lower pressures so that no extrapolation is needed. Therefore, these data should be more reliable than those of Kim et al. (*2*). To investigate the sensitivity to the calibration procedure, we have refit the calibration curve for Kim et al. (*2*), omitting their high-pressure points. We find that this causes a 7.5% difference in the calibration for the volume of their liquid sampling valve. This demonstrates the limitations inherent in determining liquid-phase densities by this technique. The overall estimated accuracies of our liquid- and vapor-phase densities are about 1% and 2%, respectively.

In Figure 5, a comparison of the liquid-phase mole fraction of carbon dioxide in anisole, 2-methylanisole, 3-methylanisole, and 4-methylanisole at 393 K indicates that carbon dioxide is most soluble in 4-methylanisole and least soluble in 2-methylanisole. The solubility of carbon dioxide changes in the order 4-methylanisole (most soluble) > 3-methylanisole > anisole > 2-methylanisole (least soluble). Among the methylanisole isomers, this follows the order of increasing molar volume:
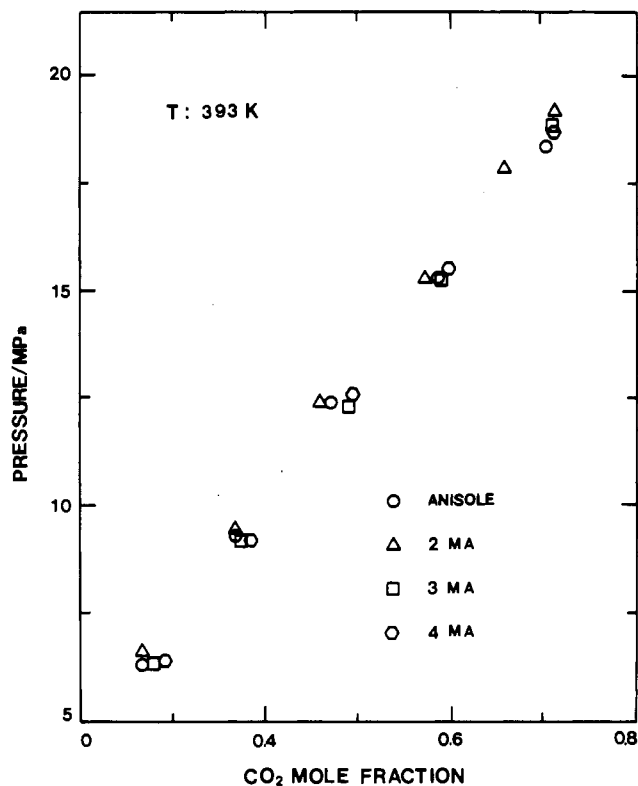

Figure 5. Comparison of liquid-phase CO_2 mole fractions as a function of pressure at 393 K for the binaries of carbon dioxide with the following: O, anisole; △, 2-methylanisole; □, 3-methylanisole; O, 4-methylanisole.
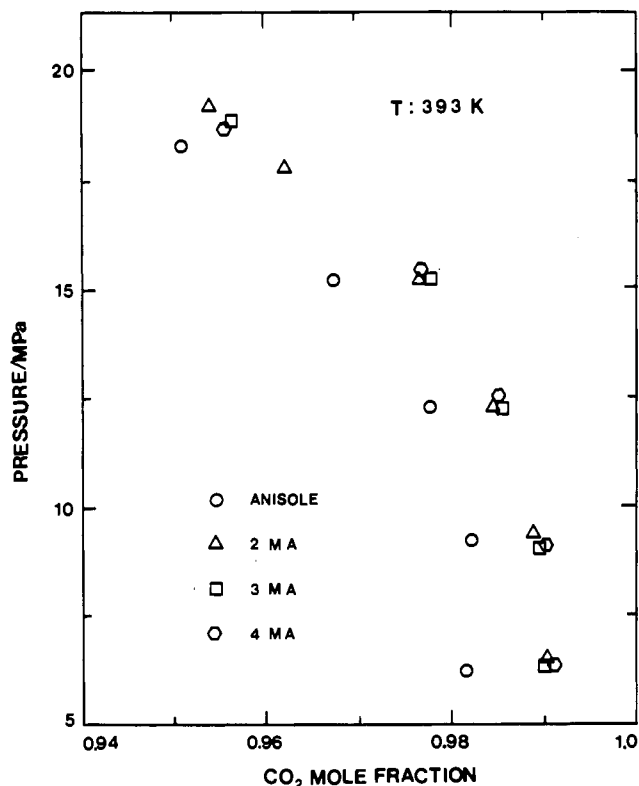

Figure 6. Comparison of vapor-phase CO_2 mole fractions as a function of pressure at 393 K for the binaries of carbon dioxide with the following: O, anisole; △, 2-methylanisole; □, 3-methylanisole; O, 4-methylanisole.

124.79, 125.82, and 126.21 cm^3/mol, respectively, for 2-, 3-, and 4-methylanisole at 293.2 K.

Competing effects between molecular structure and molecular size result in the solubility behavior of carbon dioxide in

these hydrocarbon systems. If one only considers the molecular packing of anisole and the methylanisole isomers, carbon dioxide would be more soluble in methylanisole isomers than in anisole since methylanisole isomers have more free volume and hence a lower cohesive energy density than those of anisole. On the other hand, if one considers only the entropic effect associated with the sizes of anisole and methylanisole isomers, the solubility of carbon dioxide would be more soluble in anisole than in methylanisole isomers because the size difference between the carbon dioxide and methylanisole isomers is greater than that between carbon dioxide and anisole molecules. It is believed that carbon dioxide is slightly more soluble in anisole than 2-methylanisole due to the competing effects of molecular structure and molecular size differences. Similar results are observed at 333 and 363 K.

Shown in Figure 6 is a comparison of vapor-phase compositions of aromatic hydrocarbons at 393 K. Since anisole has the highest vapor pressure, it has the highest concentration in the vapor phase. However, there is not much solubility difference among 2-methylanisole, 3-methylanisole, and 4-methylanisole, because they have similar vapor pressures.

Conclusions

The newly built equilibrium apparatus was tested by comparing the measured vapor–liquid equilibrium data for the system carbon dioxide–toluene at 393 K with those of Ng and Robinson (1). The comparison showed quite good agreement.

Phase equilibrium measurements were made on binary systems containing carbon dioxide as one of the components and anisole, 2-methylanisole, 3-methylanisole, or 4-methylanisole as the other component. When the solubilities of anisole, 2-methylanisole, 3-methylanisole, and 4-methylanisole in dense carbon dioxide at 393 K are compared, anisole is the most soluble and methylanisole isomers show little differences.

The comparison of liquid-phase compositions of carbon dioxide showed that the solubility of carbon dioxide is on the order of 4-methylanisole (most soluble) > 3-methylanisole > anisole > 2-methylanisole (least soluble).

Literature Cited

(1) Ng, H. J.; Robinson, D. B. J. Chem. Eng. Data 1978, 23, 4.
(2) Kim, C. H.; Clark, A. B.; Vimalchand, P.; Donohue, M. D. J. Chem. Eng. Data 1989, 34, 391.
(3) McHugh, M. A., The Johns Hopkins University, Baltimore, MD. Personal communication, 1986.
(4) Vargaftik, N. B. Handbook of Physical Properties of Liquids and Gases, 2nd ed.; Hemisphere Publishing Corp.: Bristol, PA, 1975.

Viscosity of Three Binary Hydrocarbon Mixtures

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Viscosity measurements have been performed on three binary hydrocarbon mixtures at atmospheric pressure in the temperature range 15–70 °C. The binary mixtures reported are n-dodecane–n-hexane, n-dodecane–benzene, and n-dodecane–cyclohexane at n-dodecane mole fractions of 1.0, 0.75, 0.50, 0.25, and 0. The viscosities of the pure liquids are described by a modified Arrhenius equation and the mixtures by a modified Grundberg and Nissan equation with a standard deviation in fit of 0.4% or less. The accuracy is estimated to be 0.6% for the lowest viscosities reported, decreasing to 0.4% for the higher values reported. Literature data deviate by –2.4% to +3.0% from our results.

1. Introduction

A high-precision viscometer for studies of low-viscosity liquids has been utilized for studies of binary hydrocarbon mixtures. These measurements have been performed at atmospheric pressure. The present measurements of binary mixtures are part of a program on viscosity of oil reservoir related fluids, together with measurements of pure hydrocarbons, reported by Knapstad et al. (1) and Aasen et al. (2), and fluids pressurized up to 400 bar, reported by Knapstad et al. (3).

The aim of the study is to produce accurate viscosity data for pure hydrocarbons and their mixtures over a wide range in temperature and pressure for computer modeling of viscosity. The hydrocarbons and the pressure and temperature range in the study are chosen to be relevant to oil reservoir characterization and production, where viscosity is an important parameter.

2. Experimental Section

The oscillation viscometer employed measures the damping of a right cylinder in contact with the liquid under consideration. The cylinder may be a solid cylinder immersed in the liquid, or a hollow cup containing the liquid. In the present measurements the viscometer was equipped with a hollow cylinder that was completely filled with sample. Absolute viscosities are computed from the measured damping and the oscillating time of the cylinder. The working equations used are described by Brockner, Tørklep, and Øye (4).

The experimental setup to maintain a completely filled hollow cylinder during the measurements was described in a previous publication of viscosities of some pure hydrocarbons reported by Knapstad et al. (1). The hollow cylinder containing the sample has a capillary tube connected to its bottom part. In the opposite end the capillary is inserted into a sample reservoir at room temperature. The sample was thus sucked into the hollow cylinder when the temperature was decreased or transported from the hollow cylinder to the reservoir when the temperature was increased.

The temperature was controlled by means of a water bath circulator, and the temperature was measured with a calibrated Ni–NiCr thermocouple, as described by Knapstad et al. (1). The fully automated viscometer under control of a host computer

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