

NOTATION

- A = surface area, sq. ft.
 C_p = heat capacity, B.t.u./ (lb.) (°F.)
 D = diameter of cylinder, ft.
 g = acceleration due to gravity, (ft./hr.)/hr.
 Gr = $gD^3\beta(t_h - T_c)/\nu^2$ = Grashof number
 h = local heat transfer coefficient based on wall-temperature difference, B.t.u./ (hr.) (sq. ft.) (°F.)
 h_m = mean heat transfer coefficient over cold or hot wall based on wall-temperature difference, B.t.u./ (hr.) (sq. ft.) (°F.)
 k = thermal conductivity of gas, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 Nu = $h_m D/k$ = Nusselt number
 Pr = $C_p \mu/k$ = Prandtl number
 q = heat flux to wall, B.t.u./hr.
 r = radial distance from axis of cylinder, ft.
 r_o = radius of cylinder, ft.
 \bar{r} = r/D = dimensionless radial distance
 T = temperature, °R.
 T_c = temperature of cold wall, °R.
 T_h = temperature of hot wall, °R.
 T_o = $(T_h + T_c)/2$ = reference temperature, °R.
 \bar{T} = T/T_o = dimensionless temperature

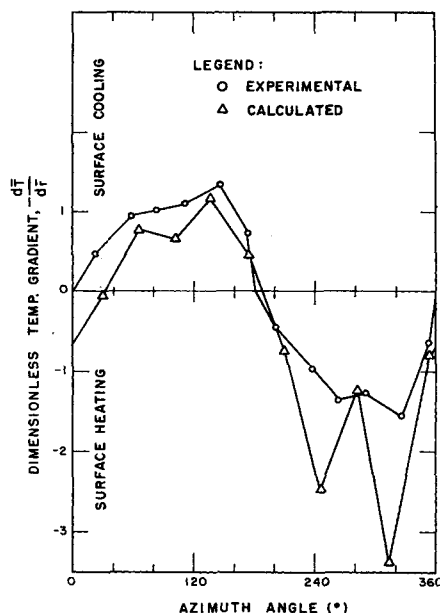


Fig. 16. Comparison of computed and measured temperature gradients, run 4, $T_h - T_c = 70^\circ\text{F}$.

w = distance from wall along a temperature traverse, ft.

Greek Letters

- β = volumetric coefficient of expansion, $^\circ\text{R}^{-1}$
 θ = azimuth angle from top of cylinder through cold side, deg.

- μ = viscosity, lb./ (ft.) (hr.)
 ν = kinematic viscosity, (sq. ft.)/hr.
 ϕ = angle between traverse line and radial line at cylinder wall, deg.

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The Compressibility of Carbon Dioxide-Argon Mixtures

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The compressibility factor has been measured at 50°C . and 50 to 1,000 atm. for each of seven mixtures of carbon dioxide and argon containing 12.9 to 83.1 mole % carbon dioxide. The compressibility factors are reported as smoothed values at even pressures and also in the form of empirical smoothing expressions which fit the experimental values with an average absolute deviation of 0.1%. Various methods of predicting gas-mixture compressibility are tested, and activity coefficients calculated from the experimental data are reported.

Despite more than 200 previous high-pressure P - V - T studies (5), it is still not possible to predict accurately the compressibility at high pressures of a gas for which experimental compressibility data are not available. The best methods of predicting gas-mixture compressibilities, for example, may be in

error by 5% at only a few hundred atmospheres of pressure. As a corollary the prediction of properties derived from the compressibility, such as the fugacity of the component of a gaseous mixture, is still more uncertain.

The present experimental study of the compressibility of mixtures of carbon dioxide and argon provides new information about gas-mixture compres-

sibility at temperatures and pressures approaching the critical region, where previous experimental information is rather limited. Seven different mixtures of carbon dioxide and argon were investigated, at 50°C . and pressures of 50 to 1,000 atm. The use of a large number of mixtures permits the calculation of accurate values of the fugacity of the components of the mixture.

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TABLE 1. COEFFICIENTS FOR SMOOTHING EQUATION (1)

Mole % CO ₂ in mixture	$A_1 \times 10^3$	$A_2 \times 10^6$	$A_3 \times 10^9$	$A_4 \times 10^{12}$	$A_5 \times 10^{15}$	$A_6 \times 10^{18}$
12.9	-0.807547	+2.349632	+3.337823	-14.969786	+40.005519	-24.913008
23.8	-1.144981	+3.995961	-9.656040	+43.430021	-76.748707	+61.698831
37.1	-1.510006	+2.976921	+4.676169	-13.777809	+20.459042	+5.0997600
46.4	-1.822606	+3.408848	+3.023837	-7.025959	+11.723338	+9.466274
62.4	-2.604265	+6.151487	-5.961679	+12.637514	-19.090902	+38.234449
72.1	-3.031094	+5.824879	+3.963679	-27.669235	+46.518088	+3.183392
83.1	-3.637742	+7.434813	+1.976118	-33.677804	+64.231621	-4.789510

The experimental fugacities in turn will assist the interpretation of diffusion studies to be made in the department of chemical engineering at Purdue University. It is planned to measure the rate of diffusion of argon and carbon dioxide in mixtures of the two at high pressures by a radioactive tracer technique currently being developed.

EXPERIMENTAL APPARATUS

A variety of methods has been used by previous investigators to measure the compressibility of gases and gas mixtures (1). One of the most successful investigators in the *P-V-T* field is Michels (15 to 22), who has reported *P-V-T* data to 3,000 atm. for a large number of pure gases. Michels estimates that densities measured in his laboratory are accurate to approximately 0.01% (21). He employs an elaborate and careful experimental approach, utilizing a glass cell supported from the outside by oil pressure equal to the gas pressure inside. However the equipment used in the present study resembles that used by Bartlett (2). An unknown quantity of gas of known composition is confined at a measured pressure in a high-pressure cell of fixed, known volume, which is immersed in a temperature-regulated bath. Then the quantity of gas in the cell is determined by expanding the high-pressure gas to a measured pressure near 1 atm. in a large, known volume of previously evacuated glassware immersed in a second constant-temperature bath. The density of the gas at the low pressure is calculated from the measured temperature and pressure.

Figure 1 is a schematic layout of the equipment, which consists of a supply of gas mixture, a high-pressure cell, a free piston gauge, low-pressure glassware, and ancillary equipment. The gas mixtures were purchased as mixtures and analyzed by Orsat analysis to 0.1% by volume. They are believed to contain less than 0.1% by weight of impurities. The gas mixtures are confined in standard carbon dioxide type of cylinders at sufficiently low pressure to be entirely gaseous at room temperature. The cylinder in use is enclosed in a steam-heated box maintained at approximately 50°C., and all the high-pressure gas lines are heated by steam in copper tubing taped to the lines in order to prevent condensation of liquid from the mixtures of carbon dioxide and argon. The critical temperature of carbon dioxide is 31°C., and preliminary investigation showed that

liquid is readily condensed from mixtures of carbon dioxide and argon at 25°C. However the experimental compressibility measurements confirm the expectation that mixtures of carbon dioxide and argon are incompressible at 50°C. and pressures up to 15,000 lb./sq. in.

The gas mixtures were dried by passage over analytical-grade magnesium perchlorate and compressed over mercury in a U-tube arrangement with high-pressure nitrogen as a source of pressure. The two vessels of the nitrogen-mercury-gas compressor are made from 4340 steel and provided with side-entering standard high-pressure fittings. The vessels are sealed at the top with O rings which swell only slightly when exposed to carbon dioxide at high pressure.

The high-pressure cell is made from a 60-in. length of stainless steel high-pressure tubing, 3/8-in. O.D. by 1/8-in. I.D., formed into the helical shape indicated in the layout. The valves of the high-pressure cell and other valves exposed to high-pressure gas mixture were made from Teflon-packed stainless steel. The volume of the high-pressure cell was determined by calibration with argon of known compressibility as reported by Michels (21). Complete details of the calibration procedure are reported in Reference 1. The calibration relies on the accuracy of Michels' data for argon in the pressure range from 50 to 1,000 atm. For eight pairs of calibration runs, each pair made at two argon densities in a ratio of two to one to each other, the average range of cell volumes in each pair was 0.04%, after computed corrections of less than 0.15% for elastic changes in cell volume with pressure. When a valve at the end of the cell was replaced, recalibration showed that the cell volume was increased by 0.8% or 0.09 cc.

The cell volume extends from valve 9 at the top of the cell to the free surface of a column of mercury near the bottom of the cell. A U-shaped mercury seal, with valve 10 at the trough of the U, separates the high-pressure gas from the oil used with the piston gauge. The position of the mercury column in the cell is determined with a special transformer (Figure 2). The transformer consists of three coils mounted on a common form, two balanced output coils connected in electrical opposition, and one input coil. An A.C. voltage of 4 v. is applied to the input coil, and the net output of the two other coils is measured with a vacuum-tube voltmeter. As the five 7/64-in.-diameter steel ball bearings floating on the mercury enter the core of the transformer, the output voltage first rises to 20

mv. and then passes through a null reading of less than 0.5 mv. when the float is centered in the core of the transformer. The ball bearings have negligible friction; the null position, which was used for all measurements, is reproducible to within approximately 0.003 in. No change in the composition of the mixture was detected in three cases for which it was analyzed before and after compression. As a precaution, new O rings were seasoned before use by an exposure to pure carbon dioxide at high pressure.

The general construction of the heavily insulated constant-temperature bath containing the high-pressure cell has been described previously (9). The bath used by Hagenbach was modified for the present work. It is now equipped with a Lightnin' portable, propeller type of mixer mounted independently of the bath, a commercial mercury-in-glass thermoregulator, and a knife-blade immersion heater. The temperature is measured to 0.01°C. by a thermometer calibrated by the National Bureau of Standards. The bath fluid is a light oil. The bath temperature is maintained at 50.00 ± 0.01°C. and is uniform within ± 0.01°C. in the vicinity of the high-pressure cell.

The piston gauge, of a design credited to Keyes (11, 12), was calibrated for this study against the known vapor pressure of carbon dioxide at 0°C., 505.45 lb./sq. in. abs. This value is the average of those values determined by three different careful investigators (3, 14, 23).

The glassware consists of eight Pyrex glass bulbs in series and an integral mercury-in-glass manometer, interconnected with pressure types of stopcocks, as shown in Figure 1. The eight available glassware volumes range from 600 to 6,000 ml. The entire glassware assembly is mounted on an angle-iron frame and immersed in a constant-temperature water bath provided with a large Plexiglas window, through which the manometer is viewed with a cathetometer reading to 0.01 cm. The temperature of the bath is maintained at a uniform 30.00 ± 0.01°C. by a mercury-in-glass thermoregulator, two propeller types of mixers, and four knife-blade immersion heaters. The glassware volumes were measured by a method described by Scott (24) which involves the displacement of air by mercury.

A vacuum pump, rated at 0.3 μ ultimate pressure with a leak-free system, is attached to the glassware with vacuum-type rubber tubing at stopcock *j*, and a tilting type of Stokes-McCloud vacuum gauge is attached at stopcock *b*.

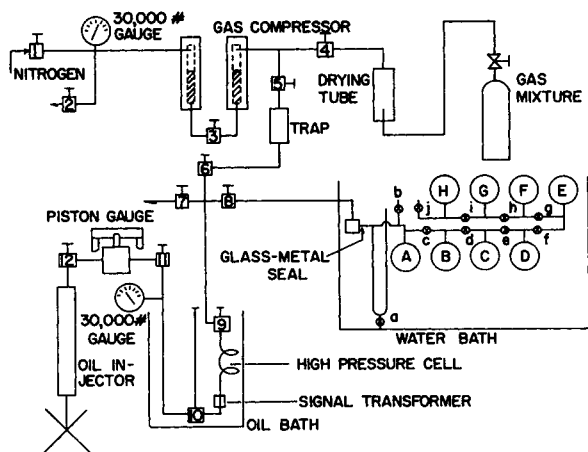


Fig. 1. Sketch of equipment.

PROCEDURE

A separate run is required for the measurement of each compressibility factor. Gas mixture of known composition is compressed over mercury and confined in the high-pressure cell for 10 to 15 min. The pressure of the gas is balanced against the oil pressure of the free piston gauge until the ball bearing float at the bottom of the cell is at the null position described earlier. Under the most unfavorable conditions, at the highest gas densities, a pressure imbalance of 0.01% can be detected, and the pressure required to bring the float to the null position appears to be reproducible within 0.02%. The temperature of the gas is assumed to be that of the constant-temperature bath in which the high-pressure cell is immersed. The high-pressure gas is then expanded through valves 9 and 8 into the low-pressure glassware, which has previously been evacuated to an absolute pressure less than 50 μ . The elevation of the mercury legs in the manometer is measured to 0.01 cm. with a cathetometer, and the barometric pressure is measured to 0.01 cm. with a conventional mercury-in-glass barometer.

In order to avoid such systematic experimental errors as creep in the cell over a long period of time, the order of experimental runs was a modified random one. The runs were divided into eighteen groups, a high- and low-pressure group for

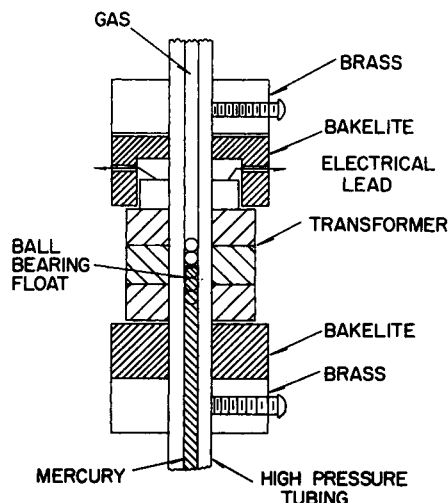


Fig. 2. Linear variable differential transformer.

each gas mixture. The order of groups and runs within a group was selected by use of a table of random numbers, although in a few cases the order of groups was changed for convenience.

CALCULATIONS

The calculation of results is straightforward. The independent variables of pressure, temperature, and composition are measured directly as explained earlier. Small corrections, amounting to less than 0.05% in all cases, are applied to the piston-gauge constant to correct for distortion of the piston gauge due to high pressures and changes in room temperature. The dependent variable, the high-pressure density, is calculated by a simple material balance, from the known volume of the high-pressure cell, the known volume of the glassware, and calculated values of the low-pressure density. Changes in volume of the cell, approximately 0.15%/1,000 atm., are computed according to a formula based on elastic theory developed by Love (13). Low-pressure densities are based on the known density of pure argon and pure carbon dioxide at low pressure (17, 18, 21) and the assumption of no volume change of mixing for these two gases. Data on the volume change which occurs on mixing two pure gases at atmospheric pressure and room temperature (14a) do not include the system argon-carbon dioxide; the system studied which is most comparable to argon-carbon dioxide was nitrogen-carbon dioxide. For this case the deviation from additive volumes, $\Delta PV/PV$, was 5×10^{-4} for a 50-50 mixture. Calculations based on the Lennard-Jones force law give a similar result for the argon-carbon dioxide system. The assumption of additive volume is therefore believed justified.

RESULTS

The general form of the experimental results is shown in Figure 3, a

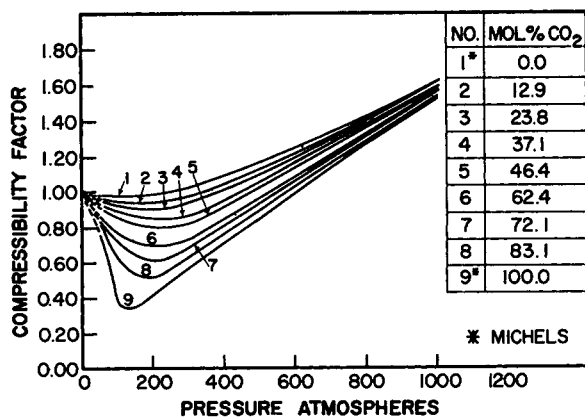


Fig. 3. The compressibility of carbon dioxide-argon mixtures at 50°C.

plot of compressibility factor vs. pressure at 50°C. for each of the seven experimental mixtures of carbon dioxide and argon. Plots for pure carbon dioxide and argon shown on the same plot are based on Michels' data. The general shapes of the curves closely resemble the familiar generalized compressibility charts (7). The lines for different gases do not cross; at any fixed pressure less than 1,000 atm., the compressibility factor increases continuously as the composition varies from pure carbon dioxide to pure argon.

The experimental compressibility factors were fitted to smoothing expressions of the form

$$C = PV/RT = 1 + A_1D + A_2D^2 + A_3D^3 + A_4D^4 + A_5D^5 + A_6D^6 \quad (1)$$

The constants A_i were determined by least-squares analyses of the twelve to fourteen experimental compressibility factors for each mixture, with the assistance of a digital computer. Table 1 lists the constants. For the six mixtures containing 12.9 to 72.1 mole % carbon dioxide the average absolute deviation of smoothed and experimental compressibility factors is 0.10%. For the 83.1 mole % carbon dioxide mixture the series is satisfactory at and below 378 atm., giving an average absolute deviation of 0.14%. At higher pressures the series does not fit the experimental data adequately, deviating by nearly 1% in one case. Michels has

TABLE 2. EMPIRICAL CORRECTION Δ_2 FOR EQUATION (2)

Δ_2	Pressure, atm.
-0.004	378
-0.003	400
+0.002	500
+0.006	600
+0.009	700
+0.008	800
+0.004	900
-0.003	1,000

found that an improved series of the same general form as that used here similarly fails to reproduce experimental compressibility factors of pure carbon dioxide (17, 18). Hirschfelder, Curtiss, and Bird (10) suggest that even the actual virial series, containing an infinite number of terms, will begin to diverge as the density of the gas approaches that of a liquid. Accordingly it is believed that the high-pressure data for the 83.1 mole % carbon dioxide mixture are as reliable as the rest of the data, even though the smoothing expression fails to reproduce the data adequately. Above 378 atm., the compressibility factor of the 83.1 mole % carbon dioxide mixture is given by

$$C = (1.2990 \times 10^{-3} P) + 0.2514 + \Delta_c \quad (2)$$

Figure 4 is a plot of the residual compressibility factor vs. composition at various fixed pressures, where

$$\Delta C = C_M - X_A C_A - X_{CO_2} C_{CO_2} \quad (3)$$

The mixture data are taken from Table 3, whereas the compressibility factors for the pure gases are taken from the literature (17, 18, 21).

The maximum value of ΔC occurs at 125 atm. for a mixture containing 62.4 mole % carbon dioxide. Above 400 atm. $\Delta C/C_M$ is less than 2% in all cases.

Bartlett's rule of additive pressure (8) was also tested over the entire range of experimental pressures and compositions. The pressures predicted by Bartlett's rule are always low; for example the molar density experimentally measured with the 37.1 mole % carbon dioxide mixture at 585.81 atm. would be anticipated by Bartlett's rule to occur at 526 atm.

Kay's pseudocritical method (4) of predicting gas-mixture compressibilities works rather well for the mixtures of carbon dioxide and argon. The average deviation of the predicted compressibility factors from experimental values is only 2.2% for a representative selection of pressures and compositions.

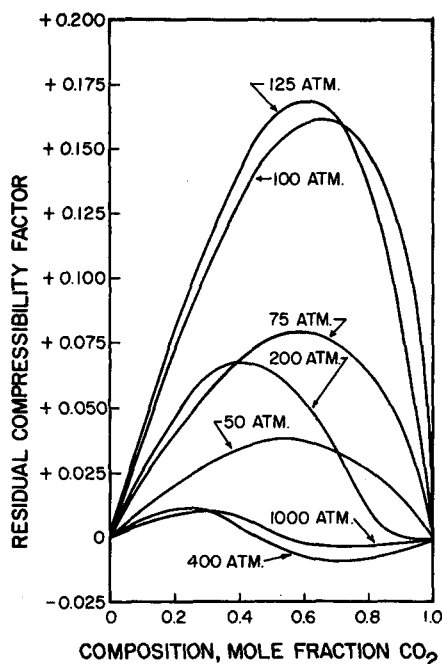


Fig. 4. Residual compressibility factors at 50°C. [Equation (3)].

As a check on the calibration of the equipment, measurements were made of the compressibility of carbon dioxide (99.95% by weight) at 50°C. and 50 to 1,000 atm. The results agree with values reported by Michels (17, 18), except in the pressure range 75 to 150 atm. Outside this range the average absolute deviation is 0.08%. The reason for disagreement in the pressure range of 75 to 150 atm., as discussed below, is not known. However the close agreement at other pressures was taken as evidence of the effectiveness of the equipment calibration. The experimental carbon dioxide measurements were not used in any calculations, since Michels' data (17, 18, 21) for pure carbon dioxide and argon are more precise than the present work.

Two series of carbon dioxide measurements were made, with carbon dioxide of the same grade (99.95% by weight) from two different cylinders. In both cases the experimental compressibility factors deviate smoothly

from the literature values in the pressure range of 75 to 150 atm. The experimental values are high in this range, exceeding the literature values by maxima of 4.5% and 1.2% at 100 atm. The reasons for these deviations have not been established. However the most plausible hypothesis seems to be that impurities in the carbon dioxide were responsible. In the case of the series showing a 4.5% deviation gross contamination of the carbon dioxide is suspected, as with a trace of oil for example. In the case of the 1.2% deviation it is at least conceivable that the anticipated impurities of 0.05% by weight of oxygen and nitrogen in the carbon dioxide are responsible for the observed deviations. Figure 4 shows that at 50°C. and 100 atm. the compressibility factor of carbon dioxide is very sensitive to small impurities of argon. A mixture containing only a small mole fraction of argon has a compressibility factor disproportionately larger than that of pure carbon dioxide. It seems reasonable that small amounts of nitrogen and oxygen should likewise raise the compressibility factor of carbon dioxide more than otherwise expected. Accordingly it is not surprising that the experimental values of the compressibility factor of carbon dioxide exceed the accepted values in a pressure range near 100 atm. Unfortunately additional experimental measurements with mixtures containing higher fractions of carbon dioxide would be required to prove that impurities of only 0.07 mole % do in fact cause the compressibility factor of carbon dioxide to increase by 1.2% at 50°C. and 100 atm.

Activity coefficients for carbon dioxide and argon in mixtures of the two have been calculated from the experimental data by a rigorous procedure described elsewhere (1) and are shown in Figures 5 and 6. The fugacity coefficients for the pure substances are given in Figure 7. The accuracy of the activity coefficients is difficult to estimate. It is thought that the activity coefficient for the major component of the

TABLE 3. SMOOTHED COMPRESSIBILITY FACTORS OF ARGON-CARBON DIOXIDE MIXTURES AT 50°C.

Mole % CO ₂ in mixture	Pressure in atm.											
	50	75	100	125	150	175	200	250	300	400	600	1,000
0*	0.9826	0.9768	0.9730	0.9715	0.9722	0.9753	0.9805	0.9973	1.0215	1.0867	1.2547	1.6317
12.9	0.9695	0.9576	0.9483	0.9420	0.9387	0.9384	0.9412	0.9549	0.9783	1.0464	1.2259	1.6229
23.8	0.9565	0.9387	0.9239	0.9124	0.9045	0.9006	0.9007	0.9125	0.9372	1.0112	1.2016	1.6181
37.1	0.9384	0.9106	0.8861	0.8661	0.8514	0.8424	0.8393	0.8486	0.8741	0.9540	1.1594	1.6007
46.4	0.9240	0.8882	0.8556	0.8278	0.8064	0.7928	0.7871	0.7967	0.8265	0.9169	1.1370	1.5927
62.4	0.8896	0.8356	0.7849	0.7408	0.7073	0.6871	0.6803	0.6972	0.7375	0.8454	1.0879	1.5724
72.1	0.8663	0.7967	0.7290	0.6698	0.6279	0.6078	0.6063	0.6365	0.6871	0.8078	1.0633	1.5614
83.1	0.8350	0.7437	0.6496	0.5665	0.5187	0.5093	0.5217	0.5725	0.6351	0.7679	1.0370	1.5471
100.0*	0.7710	0.6256	0.4135	0.3331	0.3530	0.3855	0.4212	0.4954	0.5700	0.7167	0.9992	1.5308

* Michels (17, 21).

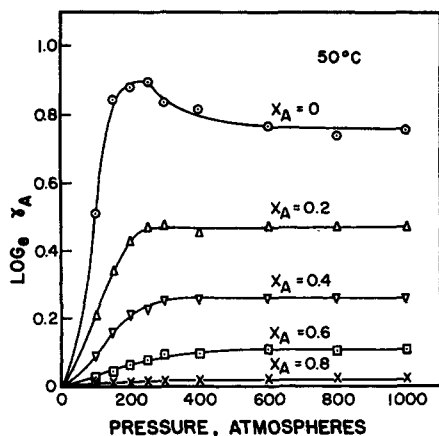


Fig. 5. Activity coefficients for argon at 50°C.

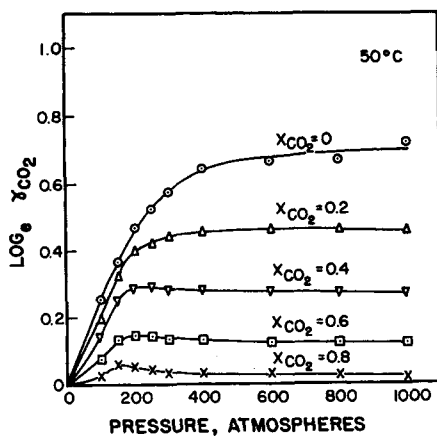


Fig. 6. Activity coefficients for carbon dioxide at 50°C.

mixture is accurate within 0.5%, but the activity coefficients for the minor component for the limiting cases of very low concentration of a component may be in error by as much as 5%.

The fugacity coefficients of argon and carbon dioxide were calculated from Michels' (17, 18, 21) data.

Further details of this study and the original experimental data may be found in reference 1.

SUMMARY

The compressibility factor has been measured experimentally at 50°C. and 50 to 1,000 atm. for each of seven mixtures of carbon dioxide and argon containing 12.9 to 83.1 mole % carbon dioxide.

Plots of compressibility factor vs. pressure resemble closely the familiar generalized compressibility factor charts. At a fixed pressure a mixture richer in argon always has a higher compressibility factor.

The experimental compressibility factors have been fitted to smoothing expressions of the form

$$PV/RT = 1 + A_1 D + A_2 D^2 + A_3 D^3 + A_4 D^4 + A_5 D^5 + A_6 D^6 \quad (1)$$

The smoothing equations (1) predict the experimental values of compressibility factor with an average absolute deviation of 0.1% for the six mixtures containing 12.9 to 83.1 mole %. For the 83.1 mole % carbon dioxide mixture the series (1) is unsatisfactory above 378 atm., and a different empirical expression is given.

Kay's pseudocritical method predicts a sample of fifteen experimental compressibility factors with an average absolute deviation of only 2.2%.

Amagat's law of additive volumes is in error by as much as 20% at intermediate pressures, but the molar volume agrees with that predicted by Amagat's law within 2% in all cases for pressures above 400 atm.

Activity coefficients for argon and carbon dioxide in mixtures of the two have been calculated from the experimental data.

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NOTATION

- A = coefficient in Equation (1)
- C = PV/RT compressibility factor, unity for an ideal gas
- D = density, the ratio of the gas density at the prevailing temperature, pressure, and composition to the density of an ideal gas at 0°C. and 1 atm.
- f_i = fugacity of a component of a gas mixture, atm.
- P = total absolute pressure, atm.
- X_i = mole fraction of a component in a gas mixture
- Δ_2 = graphical correction tabulated in Table 2 for even pressures
- ϕ_i = f_i/PX_i , fugacity coefficient
- γ_i = $f_i/f_i^\circ X_i$, activity coefficient

Subscripts

- A = argon
- M = mixture

Superscript

- o = pure substance

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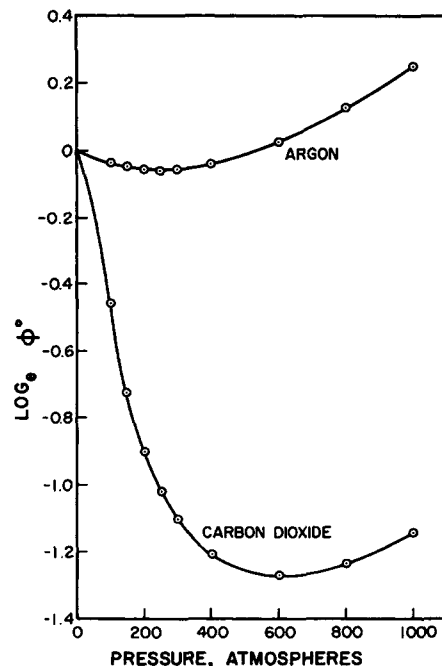


Fig. 7. Fugacity coefficients for pure argon and pure carbon dioxide at 50°C.

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