imation that $F(\Delta C)$ was negligible. Since the value of ϵ for ClF_5 was determined by comparison with a curve, this deviation from ideality should not have affected its accuracy.

ACKNOWLEDGMENT

The authors express their appreciation to Walter Maya for aiding in the density measurements.

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RECEIVED for review August 4, 1967. Accepted March 25, 1968. Research sponsored by the Air Force Rocket Propulsion Laboratory, Research and Technology Division, Air Force Systems Command, Edwards, California, under Contract AF04(611)-9563, with G. Allen Beale acting as contract monitor.

Second Virial Coefficients of Binary Mixtures of Benzene with Methanol, Ethanol, Acetone, and Diethyl Ether

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Low pressure *PVT* data were obtained and second virial coefficients were derived for the benzene-methanol, benzene-ethanol, benzene-acetone, and benzene–diethyl ether at 40°, 60°, 80°, and 100° C. The standard deviation of the experimental second virial coefficients was 32 ml. per mole, and the standard deviation of the derived interaction coefficients was 52 ml. per mole. The Kihara potential represented the derived second virial coefficients of the pure materials within experimental error. The interaction second virial coefficients of benzene-methanol and benzene-ethanol were best represented by the Lennard-Jones potential when homomorph parameters were used as nonpolar estimates of the potential parameters, and the interaction coefficients of benzene-acetone and benzene-diethyl ether were best represented by the Kihara potential.

MODERN equations of state are based upon a microscopic view, which is dependent on intermolecular interactions. Through the statistical mechanical treatment of the forces between molecules in a gas, one can derive the virial equation of state. The second virial coefficient is dependent on only two body interactions. In formulating a potential energy function to describe the energy of interaction between molecules in a gas at low density, dispersion forces (due to definite electron configurations which cause instantaneous dipoles to be formed) and dipole forces (due to the interaction of the permanent dipoles of polar molecules) are the most important. In binary systems containing a polar and a nonpolar component, there is also an interaction energy arising from dipole-induced dipole forces.

These latter systems have not been thoroughly investigated. This study was undertaken to obtain experimental second virial coefficients for systems of this type and to examine the intermolecular potential energy functions which could be applied to them.

APPARATUS

A variable-volume piezometer was constructed of spherical glass bulbs connected with 2-mm. capillary tubing (*9*). The bulbs were sized such that, on each compression, the pressure would increase by approximately one half the initial pressure. The variable-volume piezometer is shown in Figure 1. The total volume of the piezometer was 318.77 ml. The pressure of the sample was measured by a constant leg mercury manometer. Mercury was also used to compress the sample in the piezometer bulbs. Air pressure or a secondary vacuum source was used to raise and lower the mercury in the piezometer and manometer.

The piezometer was placed in a constant temperature air bath (*10*). Two 140 cubic feet per minute blowers were used to circulate the air. Three 250-watt strip heaters and water cooling coils were used to heat and cool the bath. The heaters were controlled by a Fisher Model 44 thermistor probe temperature controller. The temperature was controlled within ±0.03° C. The bath had a double plate glass window in the front through which the mercury heights were measured using a cathetometer.

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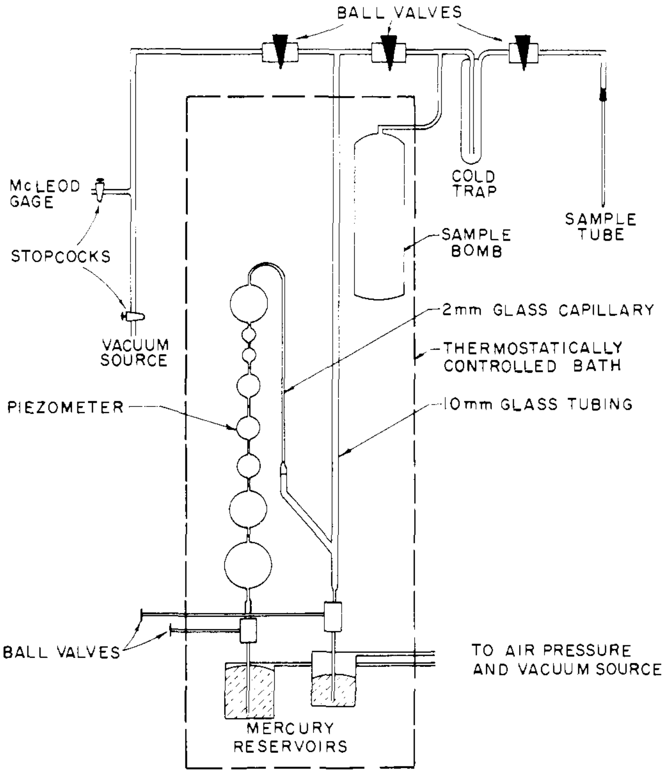


Figure 1. Low pressure *PVT* apparatus

The piezometer was evacuated with an oil-diffusion pump connected in series with a two-stage mechanical vacuum pump. The sample injection system consisted of a sample tube, a cold trap, and a sample bomb. The sample bomb was placed in the constant temperature air bath.

EXPERIMENTAL

A sample was placed in the sample tube as a liquid. If a mixture was to be tested the components were accurately weighed into the sample tube. The sample was then frozen by placing a Dewar of liquid nitrogen around the sample tube. The space above the sample was evacuated, a Dewar of liquid nitrogen was placed around the cold trap, and the sample was distilled into the cold trap where it was refrozen. The sample was then melted and vaporized into the sample bomb.

To begin a test, a sample was introduced into the piezometer and allowed to come to temperature equilibrium. The sample was compressed through several bulbs and expanded through the first two bulbs to check the initial data. Pressure, volume, and temperature data were recorded on each compression or expansion. Sample pressures ranged from 5 to 45 cm. of Hg during a test.

The second virial coefficients were derived from the data by plotting PV *vs.* P at constant temperature. The following form of the virial equation of state was used to obtain the second virial coefficients:

$$PV = nRT = nBP \qquad (1)$$

The number of moles in the sample, n, and the second virial coefficient, B, were determined from a least squares analysis of the data. Data were obtained for benzene, methanol, ethanol, acetone, and diethyl ether and binary mixtures of benzene-methanol, benzene-ethanol, benzene-acetone, and benzene–diethyl ether at 40°, 60°, 80°, and 100° C. The resulting second virial coefficients for benzene, methanol, ethanol, acetone, and diethyl ether are compared with existing literature data in Figures 2, 3, and 4, respectively. The mixture data are presented in Figure 5.

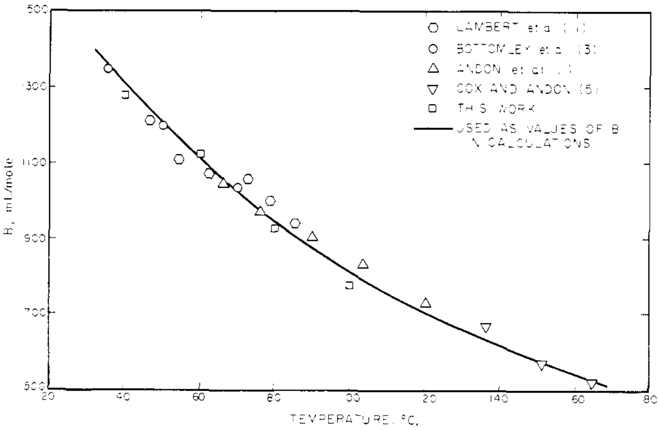


Figure 2. Second virial coefficient of benzene vs. temperature

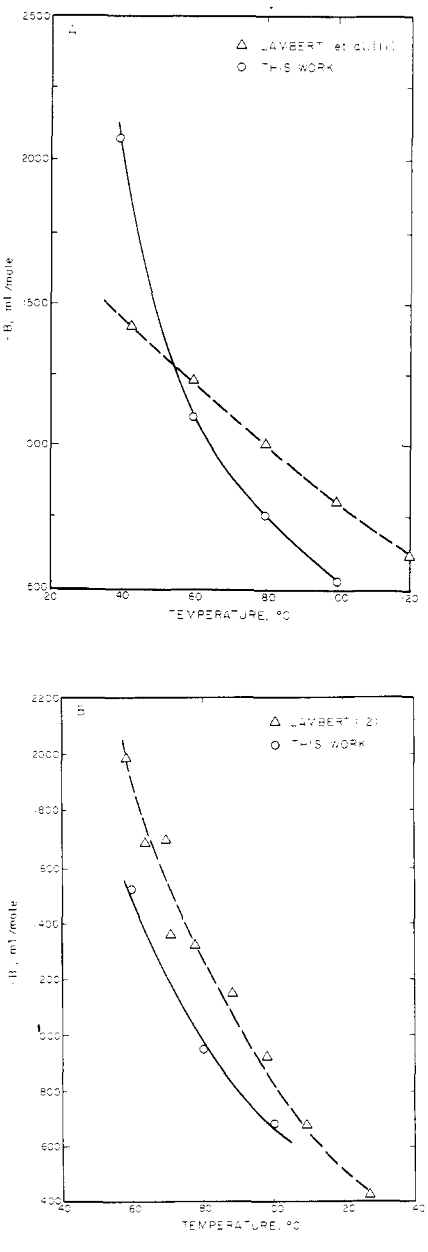


Figure 3. Second virial coefficients of *A.* methanol and *B.* ethanol as functions of temperature

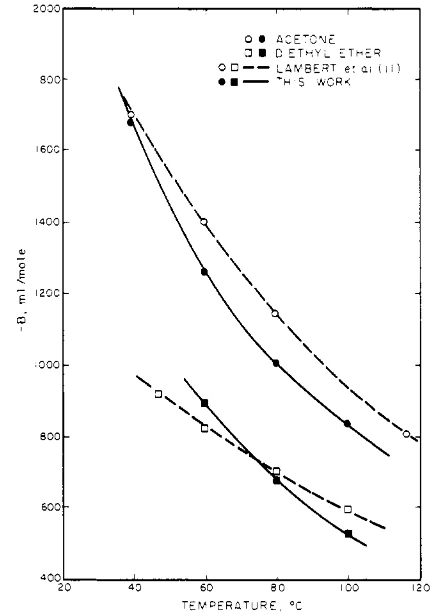


Figure 4. Second virial coefficients of acetone and diethyl ether as a function of temperature

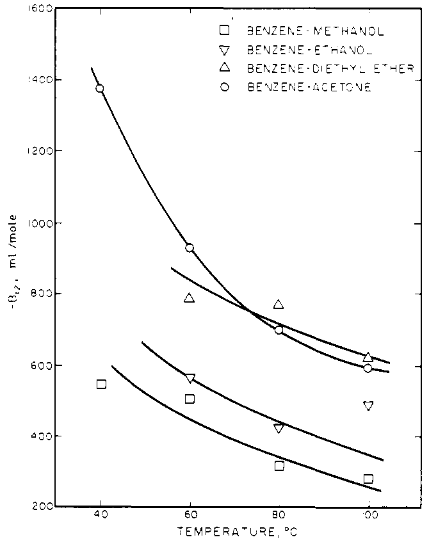
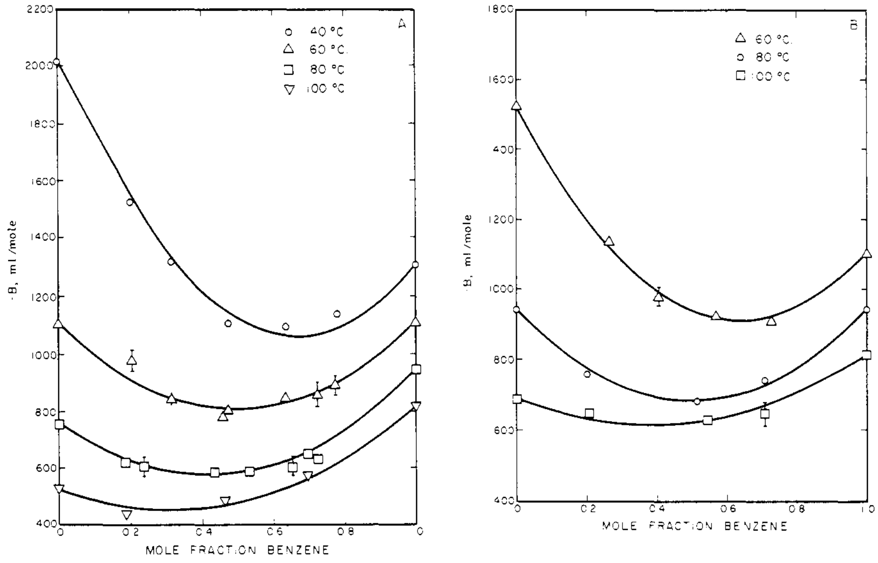


Figure 6. Temperature dependence of the interaction second virial coefficients



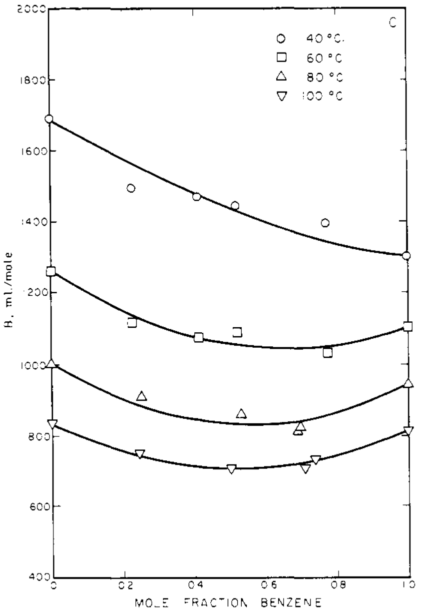
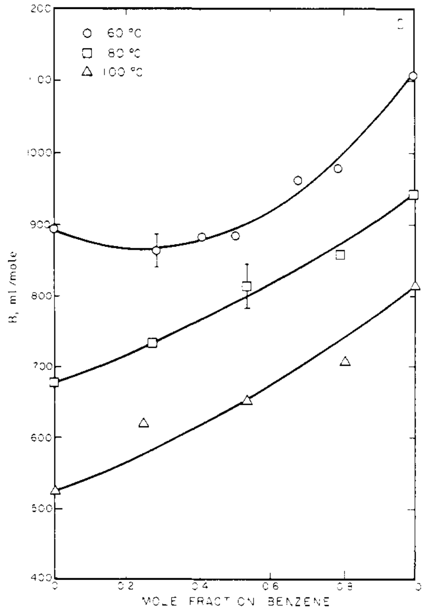


Figure 5. Second virial coefficients of binary systems vs. composition

A. Benzene-methanol
B. Benzene-ethanol
C. Benzene-acetone
D. Benzene-diethyl ether



The interaction coefficients were calculated from the mixture and pure component data using the following equation:

$$B_M = x^2B_{11} + 2x(1-x)B_{12} + (1-x)^2B_{22} \qquad (2)$$

The interaction second virial coefficients are plotted *vs.* temperature in Figure 6. The experimental second virial coefficients and calculated interaction coefficients are tabulated in Table I. A complete listing of all the results is available (*10*).

The average standard deviation of the experimental results was 32 ml. per mole. The standard deviations were determined from the statistical analysis of the *PV vs. P* plots. The analysis accounts for the error in pressure, volume, and temperature measurements. Any bias introduced into the data due to adsorption of the sample or systematic errors would not be reflected in the standard deviation calculated in this manner. However, the average deviations of 29.4 ml. per mole for multiple pure component tests and 13.4 ml. per mole for duplicate mixture tests compare favorably with the calculated average standard deviation. The standard deviation also agrees with a pre-

Table I. Summary of Experimental Results

Temp., °C.	40°		60°		80°		100°	
	x	B	x	B	x	B	x	B
Benzene-methanol	0.0000	−2079	0.0000	−1079	0.0000	−752	0.0000	−542
	0.2039	−1522	0.2039	−970	0.1843	−628	0.1903	−434
	0.3147	−1314	0.3147	−837	0.2380	−601	0.4638	−478
	0.4749	−1099	0.4617	−775	0.4349	−581	0.6961	−576
	0.6379	−1091	0.4749	−795	0.5334	−588	1.0000^a	−815
	0.7717	−1134	0.6379	−838	0.6521	−603		
	1.0000^a	−1303	0.7239	−854	0.6979	−649		
			0.7717	−882	0.7239	−731		
			1.0000^a	−1107	1.0000^a	−944		
B_{12}		−545		−507		−319		−281
Benzene-ethanol			0.0000	−1522	0.0000	−941	0.0000	−687
			0.2682	−1139	0.2033	−759	0.2071	−647
			0.4071	−979	0.5168	−683	0.5475	−629
			0.5712	−922	0.7112	−741	0.7067	−649
			0.7305	−913				
B_{12}				−567		−421		−490
Benzene-acetone	0.0000	−1690	0.0000	−1263	0.0000	−1005	0.0000	−834
	0.2277	−1493	0.2277	−1120	0.2530	−912	0.2438	−752
	0.4128	−1471	0.4128	−1077	0.5301	−861	0.5019	−714
	0.5198	−1446	0.5198	−1092	0.6947	−815	0.7122	−709
	0.7765	−1397	0.7765	−1032	0.6991	−823	0.7405	−734
B_{12}		−1376		−930		−769		−593
Benzene–diethyl ether			0.0000	−895	0.0000	−677	0.0000	−525
			0.2890	−865	0.2726	−734	0.2461	−619
			0.4124	−885	0.5365	−815	0.5342	−651
			0.5057	−885	0.7978	−860	0.8052	−706
			0.6800	−963				
			0.7915	−979				
B_{12}				−785		−698		−620

[a] Pure benzene values from Figure 2.

dicted value calculated from estimates of the measurement errors. The precision of the second virial coefficients agrees favorably with the bulk of the second virial coefficients presently available in the literature. Three experimental second virial coefficients are used to calculate the interaction coefficients. The estimated standard deviation in B_{12} is 52 ml. per mole. This deviation explains the behavior of the interaction coefficients when plotted *vs.* temperature in Figure 6.

The second virial coefficients calculated using Equation 1 are adequate for calculating thermodynamic quantities. The second virial coefficients calculated from the limiting slope of the density virial equation of state are required for interpretation of interactions between molecules. Both forms of the virial equation were used in the analysis of the data obtained in this study (*10*). However, the difference between the second virial coefficients calculated by the two techniques was less than the experimental error in the range of pressures studied. Therefore the coefficients derived using Equation 1 were used to make the theoretical comparisons.

THEORETICAL COMPARISONS

The statistical mechanical treatment of the forces between two molecules has been useful in studying second virial coefficients. This treatment requires a knowledge of the potential energy of interaction between pairs of molecules. Three potential functions were used for theoretical comparisons in this study.

The Lennard-Jones 6-12 potential function (*6*) has been used with success in representing the second virial coefficients of nonpolar molecules. This potential function assumes a soft shell molecular model which can be penetrated if the energies are sufficient. The potential energy for the Lennard-Jones potential is given by the following equation:

$$\psi = 4\epsilon\left[(\sigma/r)^{12} - (\sigma/r)^{6}\right] \qquad (3)$$

Second virial coefficients of polar molecules have been adequately represented by the Stockmayer potential (*14*). This potential is similar to the Lennard-Jones potential except that an additional term is required to account for the dipole-dipole interaction. The potential energy for the Stockmayer potential is given by the following equation:

$$\psi = 4\epsilon\left[(\sigma/r)^{12} - (\sigma/r)^{6}\right] - \frac{\mu^2}{r^3}\, g(\theta_1, \theta_2, \phi_2 - \phi_1) \qquad (4)$$

The Kihara potential (*8*) was derived for spherocylindrical molecules with impenetrable cores but has been applied to molecules of other shapes. The Kihara potential reduces to the Lennard-Jones potential when the impenetrable core becomes a point. The potential energy for the Kihara potential is given by the following equation:

$$\psi = \epsilon\left[(\rho_0/\rho)^{12} - 2(\rho_0/\rho)^{6}\right] \qquad (5)$$

The following two equations have been used to determine the potential parameters for binary mixtures (*6*):

$$\sigma_{12} = (\sigma_{11} + \sigma_{22})/2 \qquad (6)$$

$$\epsilon_{12} = (\epsilon_{11}\epsilon_{22})^{1/2} \qquad (7)$$

These equations are adequate for mixtures of nonpolar molecules. In the study of mixtures containing a polar and a nonpolar component, Blanks and Prausnitz (*2*) proposed a method using homomorphs to represent the nonpolar potential parameters of the polar components. Using these parameters and a correction factor to account for the dipole-induced dipole interaction, these authors were able to describe adequately the interaction second virial coefficients of several polar-nonpolar binary mixtures. The following equations were used to determine the mixture potential parameters:

$$\sigma'_{12} = \left[(\sigma_{11} + \sigma_{22,np})/2\right](1 + \xi_{12}^2)^{-1/6} \qquad (8)$$

$$\epsilon'_{12} = (\epsilon_{11}\epsilon_{22,np})^{1/2}(1 + \xi_{12})^2 \qquad (9)$$

where

$$\xi_{12} = \frac{\alpha_{11}\mu_{22}^{2}}{4\left(\frac{\sigma_{11}+\sigma_{22,np}}{2}\right)^{6}(\epsilon_{11}\epsilon_{22,np})^{1/2}} \tag{10}$$

However, Blanks and Prausnitz did find it necessary to multiply the correction factor for dipole-induced dipole interaction by an additional constant which was a function of $\sigma_{np} - \sigma_s$ for the polar component. It was not necessary to use this additional constant in the comparisons made in this study.

A nonlinear least squares IBM 1410 computer program was used to determine the potential parameters for the pure components. Some convergence problems were encountered, but by changing the initial estimates of the parameters, the least squares parameters were readily obtained. A spherical core molecular model was used with the Kihara potential. Tabulated potential functions (*4, 6*) were used in the calculations. The calculated Kihara and Lennard-Jones parameters are shown in Table II. The available literature data (*7*) were used to obtain the Lennard-Jones parameters for ethane, propane, isobutane, and pentane which were used as the homomorphs of methanol, ethanol, acetone, and diethyl ether, respectively. The benzene second virial coefficients were better represented by the Lennard-Jones potential here than by Sherwood and Prausnitz (*13*) because they used 30 experimental values, whereas in this study, 10 average values were used in the temperature range indicated.

The Stockmayer parameters were determined for methanol, ethanol, acetone, and diethyl ether. The tabulated Stockmayer potential function values were extrapolated to obtain a fit for methanol and ethanol. The Stockmayer parameters calculated are presented in Table III. The quantity t^* is the third parameter used to account for dipole interactions.

The calculated mixture parameters for the Kihara potential and the homomorph method proposed by Blanks and Prausnitz are shown in Table IV. The resulting interaction second virial coefficients are compared in Table V with the values determined from the experimental results of this work. The interaction coefficients for benzene-methanol and benzene-ethanol calculated using the Kihara potential over account for the dipole interaction and its effect upon the least squares value of the pure component potential parameters.

Table II. Parameters for the Kihara Potential

	ρ_0, A.	a, A.	ϵ/k, ° K.	Std. Dev., Ml./Mole
Benzene	3.37	0.00	1167.2	12.4
Ethane	2.472	0.00	729.4	8.2
Propane	5.94	0.00	256.79	1.9
Isobutane	5.89	0.00	325.1	5.6
Pentane	3.492	0.00	1046.2	2.7
Methanol	0.388	3.00	1724.7	26.1
Ethanol	0.331	3.20	1840.2	34.5
Acetone	2.166	0.200	1638.4	13.8
Diethyl ether	1.407	0.20	1940.2	7.7

Table III. Stockmayer Parameters

	ϵ/k, ° K.	σ, A.	t^*	Std. Dev., Ml./Mole
Methanol	762.0	2.54	0.565	140.0
Ethanol	624.2	2.44	0.804	14.1
Acetone	506.2	3.77	0.709	50.0
Diethyl ether	1112.8	2.39	0.228	32.5

Table IV. Mixture Potential Parameters[a]

Kihara Potential

	$(\rho_0)_{12}$, A.	$(\epsilon/k)_{12}$, ° K.	a_{11}, A.	a_{22}, A.
Benzene-methanol	1.879	1418.8	0.00	3.00
Benzene-ethanol	1.850	1465.0	0.00	3.20
Benzene-acetone	2.768	1382.9	0.00	0.20
Benzene–diethyl ether	2.388	1504.9	0.00	0.20

Lennard-Jones Potential (Using homomorph parameters for polar components)

	$(\rho_0)_{12}$, A.	$(\epsilon/k)_{12}$, ° K.	α_{11}, Cm.$^{-3}$	μ_{22}, Debyes
Benzene-methanol	2.896	1018.2	10.32	1.66
Benzene-ethanol	4.651	553.5	10.32	1.69
Benzene-acetone	4.620	632.1	10.32	2.74
Benzene–diethyl ether	3.425	1122.3	10.32	1.16

[a] $\sigma = (2)^{1/6}\rho_0$

Table V. Interaction Second Virial Coefficients

		Interaction Second Virial Coefficients, Ml./Mole		
Mixture	Temp., ° C.	Experimental	Kihara	Homomorphs
Benzene-methanol	40.00	−545	−2563	−565
	60.00	−507	−1998	−480
	80.00	−319	−1598	−415
	100.00	−281	−1304	−364
Benzene-ethanol	60.00	−567	−2375	−553
	80.00	−421	−1884	−498
	100.00	−490	−1527	−452
Benzene-acetone	40.00	−1372	−1457	−781
	60.00	−930	−1170	−693
	80.00	−698	−963	−621
	100.00	−593	−539	−561
Benzene–diethyl ether	60.00	−785	−1039	−1029
	80.00	−769	−839	−879
	100.00	−620	−695	−763

In applying a potential function to data covering such a limited temperature range, the resulting potential parameters have questionable significance. However, the approaches used herein do indicate which of the proposed methods yields the best results and would be most applicable in calculating interaction second virial coefficients when only pure component data are available. This analysis does show that the interaction between polar and nonpolar molecules in binary mixtures is very similar to the interaction of two unlike nonpolar molecules. That is, the polar molecules behave as if they were nonpolar molecules of the same size and shape in their interaction with other nonpolar molecules.

NOMENCLATURE

a = diameter of impenetrable core for Kihara potential, A.
B = second virial coefficient, ml./mole
g = function to account for dipole orientation in the Stockmayer potential
n = number of moles in sample, g.mole
P = pressure, cm. of Hg
r = distance of separation of two molecules in the Lennard-Jones potential, A.
R = universal gas constant, cm. of Hg-ml./g. mole ° C.
t^* = dimensionless Stockmayer parameter equal to $\mu^2/[8]^{1/2}\epsilon\sigma^3$
T = temperature, ° C.
V = volume, ml.
x = mole fraction of nonpolar component in binary mixture

Greek Letters

ψ = potential energy between two molecules, ergs
ϵ = minimum potential energy between two molecules, ergs
σ = distance of separation of two molecules in the Lennard-Jones potential when the potential energy equals zero, A.
μ = dipole moment of polar molecule, debyes
ρ_0 = distance between molecules in the Kihara potential when the potential energy is a minimum, A.
ρ = distance between molecules in the Kihara potential, A.
α = polarizability, cm.$^{-3}$
ξ = correction factor for dipole-induced interaction
θ_1,θ_2 = angles made by two molecules in relation to line joining their centers
ϕ_2-ϕ_1 = angle between the planes which pass through the line of their centers and contain the two axes of the two molecules

Subscripts and Superscripts

11 = nonpolar component
22 = polar component
12 = interaction parameter for polar and nonpolar component
M = mixture parameter
np = nonpolar contribution of parameter in polar component
s = Stockmayer parameter
$'$ = parameter corrected for dipole-induced dipole interaction

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RECEIVED for review April 10, 1967. Accepted February 21, 1968. D.H.K. was supported by a National Defense Education Act Fellowship and a Petroleum Research Fund Fellowship.

Lattice Parameter and Density in Bismuth-Antimony Alloys

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The hexagonal lattice parameters (*a* and *c*) and density (*d*) of chemically analyzed samples of homogeneous Bi-Sb alloy have been measured at 25° C. throughout the entire alloy system. Compositional dependences of *a* and *c* and *d* have an absolute accuracy of ±0.3 atomic % Sb. The excess volume of mixing, ΔV_m^{xs}, is close to zero in this system, indicating ideal thermodynamic behavior. Thus, it would be interesting to investigate ΔH_m^{xs} and ΔS_m^{xs} to determine whether the system is truly ideal.

IN recent years, there has been considerable interest in the electrical, thermal, and thermomagnetic properties of Bi-Sb alloys (*4, 12, 13, 16, 19*). The older investigations of the dependence of lattice parameter upon alloy composition in this system were either of insufficient accuracy (*2, 11*) or were made (*12*) on specimens not prepared under conditions of slow growth which were necessary to obtain homogeneous material (*3, 14, 20*). More recent measurements of lattice parameter on homogeneous specimens have been limited to Bi-rich alloys (*7, 10*). No measurements of the dependence of density upon alloy composition in the Bi-Sb alloy system have been reported. The purpose of this work is to establish accurately the dependences, respectively, of lattice parameter and density upon composition throughout the entire Bi-Sb alloy system.

EXPERIMENTAL PROCEDURES AND RESULTS

Homogeneous Bi-Sb alloy ingots were prepared from high-purity Bi and Sb by zone leveling using the procedure described by Yim and Dismukes (*20*). Typical mass spectrographic analyses of impurities in these materials are shown in Table I. The procedure for this study consisted of first

Table I. Mass Spectrographic Analysis of Starting Materials and Homogeneous Bi-Sb Alloys

Impurity Elements[a] Atomic, P.P.M.	Starting Materials		Homogeneous Alloys			
	Bi	Sb	$Bi_{95}Sb_5$	$Bi_{88}Sb_{12}$	$Bi_{50}Sb_{50}$	$Bi_{10}Sb_{90}$
Mg	0.3	0.6	0.2	2.1	0.2	0.3
Al	0.2	0.3	0.3	0.1	0.3	. . .
Si	1.0	2.4	3.9	1.4	5.4	5.4
Ca	0.4	0.7	0.4	0.1	0.6	. . .
Cr	0.1	0.7	0.4	0.5	. . .	0.6
Fe	0.5	2.1	0.9	0.9	1.0	1.0
Cu	0.5	0.2	0.3	0.1	0.3	. . .
Zn	1.9	0.3	. . .	0.8	. . .	. . .
Ga	0.5	. . .	. . .	. . .	0.2	0.9
As	0.1	2.7	. . .	. . .	1.1	3.0
Ag	0.2	. . .	0.8	. . .	. . .	0.1
Sn	. . .	0.1	. . .	. . .	. . .	0.5
Pb	0.1	0.6	. . .	. . .	0.3	0.4
Sb	6.0	MC	MC	MC	MC	MC
Bi	MC[b]	. . .	MC	MC	MC	MC

[a] Metallic impurities only. [b] Major components.