for methyl and propyl acetates. Two recent publications provide vaporization enthalpies for both compounds (*3*, *10*), and corresponding values for *B* are presented in Table VIII. Those based on ref *10* are more posistive in all cases than those interpolated from the data of ref *3*. The former for methyl acetate are suspect, since *B* does not vary in the expected sense with temperature.

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Received for review February 4, 1980. Accepted July 21, 1980. The platinum resistance thermometer and G-2 Mueller bridge used for temperature measurement were purchased with funds from Research Corporation, for which we are most grateful.

Volumetric Behavior of Ethylene and Ethylene–Hydrogen Mixtures

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A Burnett apparatus designed and fabricated was used to collect volumetric data for ethylene and ethylene–hydrogen mixtures. Measurements were made in the temperature range 298.15–423.15 K at intervals of 25 K and in the pressure range 0.3–7.0 MPa. Virial coefficients derived from the compressibility data are tabulated. The data are fitted to different equations of state.

Introduction

The recent IUPAC (*1*) tabulation of *PVT* data for ethylene revealed that data are available above 1.5 MPa in the temperature range 298.15–423.15 K. The recent measurements of Douslin and Harrison (*2*) also do not extend below 1.5 MPa. The data of Charnley et al. (*3*) and Daniel and Stoltzenberg (*4*) do extend in the pressure range 0–20 MPa, but the temperatures are limited to 318.15 K. However, below 0.1 MPa, data are available (*5*–*7*) up to 473.15 K. In order to extract information about intermolecular forces, it is necessary to have information on *PVT* properties at reasonably low pressures over a sufficient temperature range. Further, as ethylene is a very important industrial chemical, it is better to have as much experimental data in all ranges of pressure and temperature as possible. For these reasons the work described in this paper was undertaken.

A detailed literature survey revealed that there are practically no *PVT* data on ethylene–hydrogen mixtures except for two points (*8*). Therefore, the *PVT* behavior of ethylene–hydrogen mixtures was studied in the range 298.15–423.15 K up to 7.0 MPa.

Experimental Apparatus and Procedure

The details of the experimental setup and procedure have been discussed by Prasad (*9*). A schematic diagram of the general arrangement is shown in Figure 1. The Burnett cell was fabricated from a 9-cm diameter AISI 304 stainless steel rod. The compartment dimensions were 13.0 and 6.5 cm in length and 5 cm in diameter. The compartments of the cell were

Table I. Properties of Chemicals Used (T = 303.15 K)

	refractive index		density	
	present work	lit.	present work	lit.
water	1.331 80	1.331 92 (*10*)	0.995 65	0.995 504 (*11*)
benzene	1.494 74	1.494 68 (*12*)	0.868 25	0.868 25 (*13*)
toluene	1.491 45	1.491 26 (*13*)	0.857 25	0.857 60 (*13*)

Table II. Analyses of Ethylene and Hydrogen

ethylene: ethylene, 99.5; carbon dioxide, 0.2; propane, nitrogen, and hydrogen, 0.1 each

hydrogen: hydrogen, 99.932; carbon dioxide, 0.029; oxygen, 0.029; nitrogen, 0.008

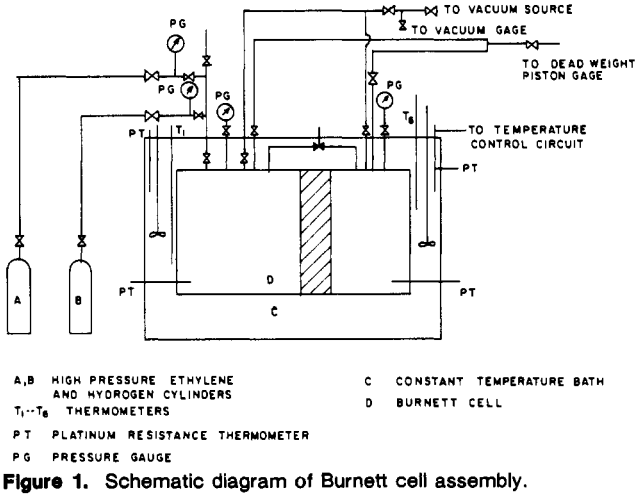

Figure 1. Schematic diagram of Burnett cell assembly.

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Table III. Compressibility Behavior of Ethylene

press., MPa	compressibility factor	press., MPa	compressibility factor
	temp: 298.15 K		
5.1719	0.618 79	1.2095	0.925 06
4.0419	0.727 57	0.8276	0.950 10
2.9952	0.807 73	0.5607	0.964 94
2.1465	0.863 32	0.3787	0.977 67
1.5032	0.912 11	0.2546	0.985 85
1.0330	0.940 89	3.6006	0.749 58
0.7035	0.960 37	2.6407	0.824 67
0.4752	0.973 22	1.8762	0.878 85
0.3201	0.983 32	1.3067	0.918 14
4.4762	0.676 24	0.8965	0.944 86
3.3889	0.767 97	0.6090	0.963 48
2.4650	0.837 91	0.4110	0.974 92
1.7446	0.889 50		
	temp: 323.15 K		
5.1733	0.740 35	0.7255	0.971 27
3.8267	0.821 47	0.4890	0.981 30
2.7257	0.877 66	0.3277	0.986 35
1.8969	0.916 21	0.2195	0.990 81
1.2998	0.941 74	3.4282	0.847 51
0.8827	0.960 01	2.4154	0.895 73
0.6007	0.979 27	1.6757	0.932 07
0.4022	0.983 37	1.1454	0.955 72
4.4624	0.786 11	0.7752	0.970 19
3.2386	0.855 78	0.5221	0.980 97
2.2754	0.901 90	0.3511	0.988 48
1.5729	0.935 15	0.2353	0.993 35
1.0703	0.956 32		
	temp: 348.15 K		
3.5117	0.868 18	0.8517	0.975 11
2.4554	0.910 56	0.5732	0.980 46
1.6928	0.941 66	0.3842	0.989 81
1.1516	0.960 90	0.2574	0.994 52
0.7779	0.973 64	5.6414	0.772 40
0.5242	0.984 12	4.1611	0.854 59
0.3518	0.990 79	2.9580	0.911 25
0.2353	0.993 99	2.0348	0.940 17
3.8074	0.861 07	1.3867	0.961 19
2.6650	0.904 05	0.9372	0.975 13
1.8480	0.940 33	0.6297	0.982 04
1.2619	0.963 20	0.4228	0.989 19
	temp: 373.15 K		
5.5173	0.829 89	1.0365	0.977 37
3.9391	0.886 55	0.6966	0.986 00
2.7408	0.925 29	0.4677	0.992 24
1.8783	0.951 16	0.3125	0.994 66
1.2743	0.967 97	4.9795	0.848 35
0.8586	0.978 98	3.5185	0.899 17
0.5766	0.985 45	2.4416	0.935 93
0.3870	0.992 10	1.6625	0.955 93
4.6348	0.863 31	1.1261	0.971 25
3.2634	0.911 81	0.7538	0.981 28
2.2506	0.943 24	0.5090	0.987 83
1.5343	0.964 52	0.3408	0.992 04
	temp: 398.15 K		
5.1519	0.886 74	0.5628	0.989 17
3.5854	0.925 69	0.3767	0.992 98
2.4554	0.950 90	0.2519	0.995 98
1.6687	0.969 36	4.6555	0.898 71
1.1247	0.980 04	3.2235	0.933 40
0.7552	0.987 77	2.2010	0.955 99
0.5056	0.991 23	1.4895	0.970 41
3.9391	0.911 70	1.0034	0.980 58
2.7243	0.945 79	0.6738	0.988 49
1.8521	0.964 50	0.4511	0.991 95
1.2502	0.976 58	0.3015	0.994 46
0.8400	0.984 20		
	temp: 423.15 K		
3.7936	0.937 92	0.8096	0.987 88
2.5926	0.961 46	0.5414	0.990 22
1.7521	0.973 24	0.3622	0.993 56
1.1764	0.981 24	0.2422	0.996 68
0.7896	0.987 94	3.3400	0.941 73
0.5283	0.992 36	2.2761	0.962 66
0.3532	0.994 49	1.5391	0.976 50
5.5366	0.915 52	1.0344	0.984 35
3.8612	0.929 93	0.6931	0.989 37
2.6429	0.954 93	0.4635	0.992 47
1.7907	0.970 38	0.3100	0.994 91
1.2054	0.979 80		

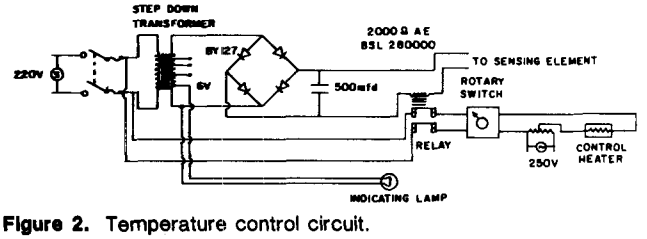

Figure 2. Temperature control circuit.

connected by a network of precision needle valves which could be operated to connect the compartments to (i) a vacuum pump for evacuation, (ii) pressure measuring devices, (iii) an expansion valve to conduct the desired expansion, and (iv) the feeding system. The entire cell assembly was thermostated.

The pressures were measured with Bourdon-type "standard test" gauges supplied by M/S Budenberg Gage Co., Broadheath, England. These were calibrated against a dead-weight tester of M/S Alfred J. Amsler and Co., Switzerland. A circular vernier was used for interpolation. Pressures measured were accurate to 1 part in 10 000 in most of the regions.

Temperatures were measured with mercury-in-glass thermometers and platinum resistance thermometers. These were calibrated against standard thermometers and also against the ice point and boiling point of high-purity water and the vapor pressures of benzene and toluene. The properties of these substances used for calibration are given in Table I. Besides these properties, water was tested for its electrical conductivity, and benzene and toluene were tested for their purity by va-

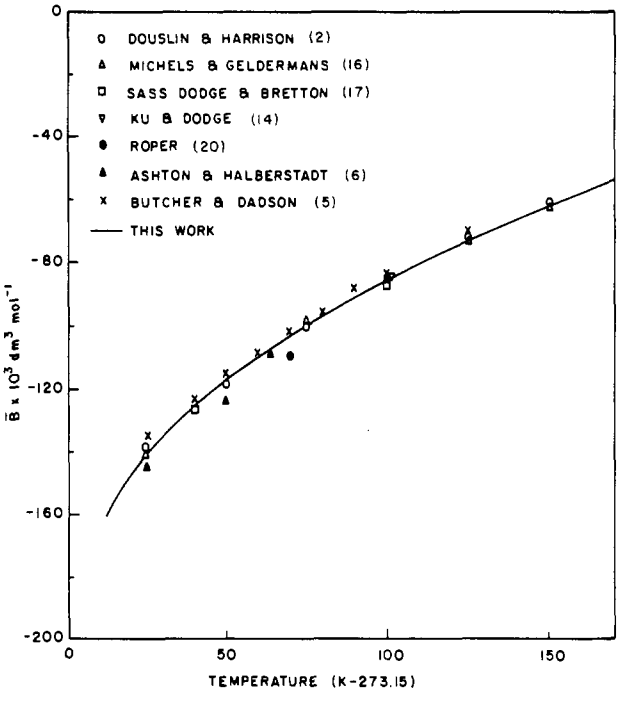

Figure 3. Second virial coefficients of ethylene.

por-phase chromatograms and NMR spectra. Figure 2 shows

Table IV. Compressibility Behavior of Ethylene–Hydrogen Mixtures

press., MPa	compressibility factor	press., MPa	compressibility factor
temp: 298.15 K			
Mole Fraction of Ethylene = 0.742 26			
6.8893	0.643 81	1.3171	0.928 44
5.2077	0.729 02	0.9000	0.950 32
3.8123	0.799 43	0.6104	0.965 53
2.7208	0.854 71	0.4118	0.975 84
1.9052	0.896 54	0.2774	0.984 57
Mole Fraction of Ethylene = 0.123 84			
6.8183	1.010 18	1.3509	1.007 82
4.5486	1.009 51	0.9006	1.006 55
3.0352	1.009 10	0.6000	1.004 56
2.0252	1.009 10	0.4001	1.003 39
Mole Fraction of Ethylene = 0.397 88			
6.9962	0.821 66	1.0772	0.954 25
4.8471	0.852 76	0.7304	0.979 24
3.3593	0.885 31	0.4932	0.980 42
2.3182	0.915 24	0.3312	0.986 16
1.5839	0.936 71		
Mole Fraction of Ethylene = 0.224 01			
6.4963	0.954 98	1.3323	0.986 18
4.3900	0.966 73	0.8924	0.989 53
2.9559	0.975 09	0.5980	0.993 30
1.9872	0.982 01	0.4001	0.995 58
temp: 323.15 K			
Mole Fraction of Ethylene = 0.720 63			
6.7032	0.792 82	1.0841	0.967 18
4.7934	0.849 27	0.7324	0.978 87
3.3607	0.891 95	0.4925	0.985 98
2.3230	0.923 60	0.3298	0.989 01
1.5805	0.941 30		
Mole Fraction of Ethylene = 0.551 02			
7.0066	0.869 39	1.0496	0.982 39
4.8003	0.892 20	0.7048	0.988 27
3.3007	0.919 04	0.4172	0.990 96
2.2872	0.954 00	0.3153	0.992 02
1.5529	0.970 29		
Mole Fraction of Ethylene = 0.345 11			
6.8645	0.939 20	0.9599	0.990 72
4.6762	0.958 40	0.6428	0.993 77
3.1642	0.971 46	0.4297	0.995 27
2.1313	0.980 26	0.2870	0.995 77
1.4315	0.986 28		
Mole Fraction of Ethylene = 0.125 19			
6.7032	1.005 25	0.8841	1.000 12
4.4624	1.002 48	0.5904	1.000 47
2.9746	1.001 01	0.3939	0.999 89
1.9845	1.000 42	0.2629	0.999 69
1.3247	1.000 34		
temp: 348.15 K			
Mole Fraction of Ethylene = 0.895 33			
4.8899	0.843 20	1.1192	0.971 82
3.4703	0.896 40	0.7552	0.981 51
2.4064	0.931 16	0.5077	0.989 16
1.6480	0.955 28	0.3401	0.992 74
Mole Fraction of Ethylene = 0.683 42			
6.7928	0.845 78	1.0358	0.972 83
4.7051	0.877 58	0.6979	0.981 97
3.2497	0.907 96	0.4684	0.987 11
2.2423	0.938 53	0.3139	0.991 08
1.5288	0.958 51		
Mole Fraction of Ethylene = 0.548 49			
6.6136	0.893 61	0.9599	0.978 40
4.5417	0.919 27	0.6462	0.986 68
3.0973	0.939 11	0.4332	0.990 78
2.0104	0.955 86	0.2898	0.992 84
1.4233	0.968 37		
temp: 348.15 K			
Mole Fraction of Ethylene = 0.316 72			
6.6825	0.961 53	0.9144	0.992 51
4.4673	0.962 88	0.6118	0.994 66
2.9835	0.963 32	0.4118	0.996 30
2.0169	0.975 54	0.2732	0.996 90
1.3598	0.985 94		
Mole Fraction of Ethylene = 0.135 86			
6.5453	0.994 69	0.8682	0.995 30
4.3576	0.992 02	0.5807	0.997 25
2.9049	0.990 62	0.3877	0.997 28
1.9397	0.990 98	0.2588	0.997 11
1.2978	0.993 14		
temp: 373.15 K			
Mole Fraction of Ethylene = 0.836 59			
5.1864	0.890 08	0.7634	0.988 33
3.6247	0.931 86	0.5110	0.991 16
2.4830	0.956 22	0.3422	0.994 04
1.6846	0.971 84	0.2291	0.997 01
1.1358	0.981 53		
Mole Fraction of Ethylene = 0.671 51			
6.6204	0.904 11	0.9579	0.986 45
4.5693	0.934 91	0.6421	0.991 13
3.1193	0.955 91	0.4297	0.993 40
2.1169	0.971 77	0.2870	0.993 90
1.4253	0.980 15		
Mole Fraction of Ethylene = 0.567 39			
6.5929	0.940 53	0.9200	0.989 98
4.4845	0.958 35	0.6152	0.991 74
3.0325	0.970 78	0.4111	0.992 80
2.0424	0.979 45	0.2746	0.993 38
1.3716	0.985 29		
Mole Fraction of Ethylene = 0.370 21			
6.4495	0.978 93	0.8710	0.997 25
4.3369	0.986 10	0.5821	0.998 41
2.9096	0.990 84	0.3884	0.997 85
1.9486	0.994 25	0.2732	0.998 55
1.3033	0.996 14		
Mole Fraction of Ethylene = 0.193 36			
6.4274	0.985 31	0.8620	0.996 84
4.3080	0.989 29	0.5759	0.997 62
2.8835	0.991 95	0.3849	0.998 85
1.9286	0.993 92	0.2567	0.997 79
1.2895	0.995 43		
temp: 398.15 K			
Mole Fraction of Ethylene = 0.895 52			
4.3038	0.922 64	0.9158	0.987 69
2.9670	0.952 80	0.6138	0.992 62
2.0183	0.970 90	0.4104	0.994 25
1.3626	0.981 93	0.2746	0.996 51
Mole Fraction of Ethylene = 0.603 29			
6.6687	0.917 03	0.9565	0.990 02
4.5672	0.940 81	0.6393	0.993 45
3.1097	0.959 58	0.4277	0.995 49
2.1051	0.973 11	0.2856	0.996 01
1.4198	0.983 16		
Mole Fraction of Ethylene = 0.469 46			
7.5236	0.947 63	1.0413	0.989 34
5.1009	0.962 42	0.6973	0.992 37
3.4399	0.972 26	0.4663	0.994 13
2.3147	0.980 10	0.3118	0.995 96
1.5538	0.985 35		
Mole Fraction of Ethylene = 0.318 98			
6.6687	0.970 80	0.9062	0.995 07
4.4907	0.979 29	0.6056	0.996 12
3.0159	0.985 21	0.4049	0.997 79
2.0217	0.989 34	0.2705	0.998 42
1.3536	0.992 29		

Table IV (*Continued*)

press., MPa	compressibility factor	press., MPa	compressibility factor
temp: 398.15 K			
Mole Fraction of Ethylene = 0.157 82			
6.7032	0.987 73	0.8972	0.997 25
4.4900	0.991 09	0.5994	0.997 95
3.0042	0.991 33	0.4001	0.997 94
2.0086	0.994 93	0.2670	0.997 73
1.3426	0.996 21		
temp: 423.15 K			
Mole Fraction of Ethylene = 0.785 87			
5.2208	0.930 16	0.7393	0.993 59
3.5882	0.957 64	0.4946	0.995 64
2.4347	0.973 39	0.3305	0.996 60
1.6418	0.983 29	0.2208	0.997 66
1.1027	0.989 26		
Mole Fraction of Ethylene = 0.650 30			
6.7804	0.940 73	0.9482	0.992 38
4.6176	0.959 69	0.6345	0.994 76
3.1256	0.973 10	0.4242	0.996 30
2.1058	0.982 13	0.2836	0.997 64
1.4143	0.988 10		
temp: 423.15 K			
Mole Fraction of Ethylene = 0.313 76			
6.6342	0.984 24	0.8917	0.997 89
4.4521	0.989 43	0.5952	0.998 30
2.9821	0.992 80	0.3973	0.997 83
1.9962	0.995 54	0.2657	0.999 35
1.3343	0.996 83		
Mole Fraction of Ethylene = 0.530 31			
6.5170	0.960 30	0.8938	0.993 42
4.4073	0.972 83	0.5980	0.995 66
2.9677	0.981 28	0.3994	0.996 22
1.9928	0.987 08	0.2670	0.997 73
1.3350	0.990 59		
Mole Fraction of Ethylene = 0.122 56			
6.5584	1.028 09	1.2750	1.006 48
4.3397	1.019 08	0.8496	1.004 64
2.8801	1.011 31	0.2519	1.001 13
1.9149	1.009 05		

Table V. Comparison of Compressibility Factors of Ethylene with Literature Data

ref	range of comparison: temp, (T − 273.15 K)	range of comparison: press., MPa	no. of points compared	% av absolute deviation[a]
Ku and Dodge (*14*)	100	1.01–5.15	6	0.66
Lee and Edmister (*15*)	25–75	1.77–5.04	9	0.37
Michels and Geldermans (*16*)	25–150	1.86–5.49	45	0.57
Sass et al. (*17*)	40–100	0.72–5.48	15	0.28
Walters et al. (*18*)	26.66–37.77	0.35–2.03	69	0.49
Thomas and Zander (*19*)	30–50	0.10–2.03	15	0.28
Douslin and Harrison (*2*)	25–150	1.77–5.70	26	0.66
overall			185	0.50

[a] % average absolute deviation = (present − literature)/present × 100.

Table VI. Virial Coefficients of Ethylene

temp, K	$10B$, MPa^{-1}	$10^3\bar{B}$, $dm^3\ mol^{-1}$	10^2C, MPa^{-2}	$\bar{C}$, $dm^6\ mol^{-2}$
298.15	−0.565 34	−140.2	−0.4313	−0.686
323.15	−0.441 04	−118.5	−0.1662	−0.205
348.15	−0.345 01	−99.9	−0.0842	−0.292
373.15	−0.272 02	−84.4	−0.0295	−0.428
398.15	−0.214 72	−71.1	0.0163	0.327
423.15	−0.173 95	−61.2	0.0421	0.895

the temperature control circuit used to achieve a temperature control of +0.005 K.

Materials. The analyses of ethylene and hydrogen used in the experiments are shown in Table II. Hydrogen was used to test the apparatus, to obtain the apparatus constant, and to get ethylene–hydrogen mixtures. Both of the gases were analyzed by means of an MS-10 mass spectrometer employing a Dempster's Type 2-in. radius permanent magnetic analyzer. Although ethylene was analyzed as 99.5%, the gas was used as such without further purification.

Data and Data Analysis. The experimental data have been used to determine compressibility factors and virial coefficients. The compressibility data thus obtained are tabulated in Tables III and IV. A comparison of ethylene data with literature values is given in Table V. Virial coefficients were extracted by using the relation

Table VII. Virial Coefficients of Ethylene–Hydrogen Mixtures

mole fraction of ethylene	$10B$, MPa^{-1}	$10^3\bar{B}$, $dm^3\ mol^{-1}$	10^3C, MPa^{-2}	$\bar{C}$, $dm^6\ mol^{-2}$
Temp: 298.15 K				
0.742 26	−0.564 24	−139.9	0.8246	0.025
0.397 88	−0.379 16	−94.0	1.6443	0.019
0.224 01	−0.114 30	−28.3	0.7912	0.006
0.123 84	−0.077 17	−19.1	1.0882	0.010
Temp: 323.15 K				
0.720 63	−0.316 35	−85.0	−0.3383	−0.006
0.551 02	−0.200 17	−53.8	−0.1226	−0.002
0.345 11	−0.114 01	−30.6	0.4660	0.004
0.225 19	−0.002 61	−7.0	0.1611	0.001
Temp: 348.15 K				
0.895 33	−0.219 17	−63.44	−2.2946	−0.015
0.683 42	−0.257 61	−74.57	0.1071	0.006
0.548 49	−0.230 63	−66.76	1.0817	0.014
0.316 72	−0.106 18	−30.74	0.4830	0.005
0.135 86	−0.073 83	−21.37	1.1641	0.010
Temp: 373.15 K				
0.836 59	−0.151 14	−46.9	−1.1044	−0.008
0.671 51	−0.157 81	−49.0	0.3639	0.006
0.562 79	−0.159 41	−49.0	1.4567	0.017
0.370 21	−0.036 84	−11.4	0.1224	0.003
0.193 36	−0.046 27	−14.4	0.0473	0.002
Temp: 398.15 K				
0.895 52	−0.151 87	−50.3	1.8625	0.023
0.603 29	−0.115 17	−38.1	−0.2295	−0.004
0.463 92	−0.116 04	−38.4	0.7674	0.010
0.318 98	−0.058 90	−19.5	0.2545	0.003
0.157 82	−0.047 28	−15.7	0.5669	0.006
Temp: 423.15 K				
0.785 87	−0.091 82	−32.3	−0.7443	−0.008
0.650 30	−0.083 26	−29.3	−0.0696	−0.001
0.530 31	−0.080 50	−28.3	0.3980	0.006
0.313 76	−0.032 93	−11.6	0.2017	0.003
0.122 56	−0.047 29	−15.0	−0.0789	−0.001

$$Z = 1 + BP + CP^2$$

and the values for the volume series were obtained by using the relations

$$\bar{B} = RTB$$

$$\bar{C} = R^2T^2(C + B^2)$$

Figures 3 and 4 show second virial coefficients plotted as a function of temperature for ethylene and ethylene–hydrogen

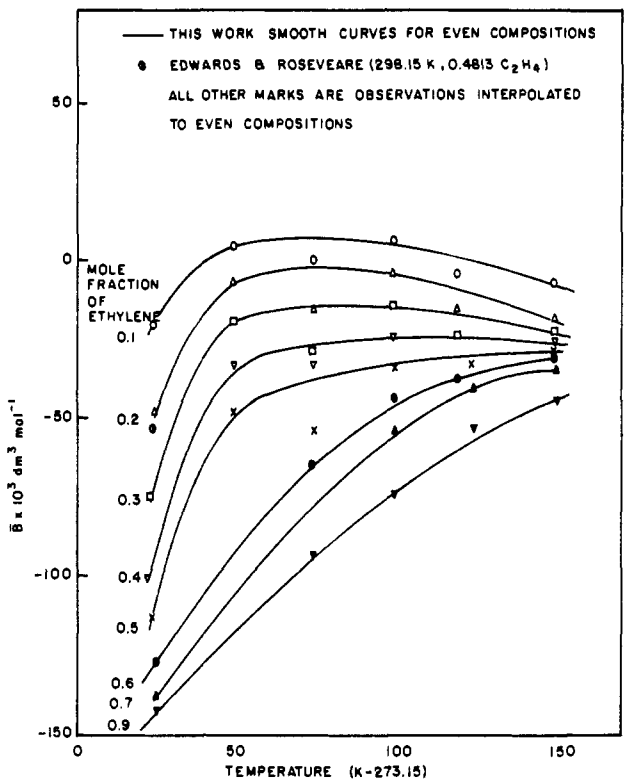

Figure 4. Second virial coefficients of ethylene–hydrogen mixtures.

mixtures. The figures also include the literature data. The values of the virial coefficients are presented in Tables VI and VII.

Three different equations of state, the virial equation as pressure series (up to P^2), the simple two-constant Redlich–Kwong equation, and the complicated Angus et al. equation (with 32 constants), were used to predict compressibility factors for ethylene. In the temperature range 298.15–423.15 K and the pressure range 0.2–5.6 MPa, a total of 145 points were compared with experimental data. The three equations (in the order cited) gave percent average absolute deviations of 0.56, 0.54, and 0.44 respectively.

In the case of ethylene–hydrogen mixtures, only the Redlich–Kwong equation was used. However, the two constants a_{mix} and b_{mix} for the mixtures were calculated by using the following mixing rules:

1. Redlich–Kwong rule

$$a_{mix} = a_1x_1^2 + a_2x_2^2 + 2(a_1a_2)^{1/2}x_1x_2$$

$$b_{mix} = b_1x_1 + b_2x_2$$

2. Joffe–Zudkevitch rule

$$a_{mix} = a_1x_1^2 + a_2x_2^2 + 2(a_1a_2)^{1/2}x_1x_2$$

$$b_{mix} = b_1x_1^2 + b_2x_2^2 + (b_1 + b_2)x_1x_2$$

3.

$$a_{mix}^{1/3} = \frac{x_1a_1^{1/3}M_1^{1/3} + x_2a_2^{1/3}M_2^{1/3}}{x_1M_1^{1/3} + x_2M_2^{1/3}}$$

$$b_{mix} = x_1b_1 + x_2b_2$$

4.

$$a_{mix} = \frac{0.4278R^2Tc_{mix}^{2.5}}{Pc_{mix}}$$

$$b_{mix} = \frac{0.0867RTc_{mix}}{Pc_{mix}}$$

$$Tc_{mix} = x_1Tc_1 + x_2Tc_2$$

$$Pc_{mix} = x_1Pc_1 + x_2Pc_2$$

For 249 points compared, the above four relations gave average absolute deviations of 1.82, 1.82, 2.36, and 1.54%, respectively.

Acknowledgment

It is my (D.S.V.) pleasure to thank B. Kay McNew for typing this manuscript.

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Received for review March 28, 1980. Accepted July 25, 1980.