Thermodynamic Properties of Compressed Gaseous Methane

Roland H. Harrison,[1] Richard T. Moore, and Donald R. Douslin
Bartlesville Energy Research Center, Bureau of Mines. U.S. Department of the Interior, Bartlesville. Okla. 74003

Real gas thermodynamic properties, $H - H°$, $S - S°$, and $G - G°$, of methane were determined as functions of temperature (0–350°C) and molal density (0.75–12.5 mol dm^{-3}) over a pressure range of 0–400 atm. These properties are recommended as standards for the development of equation of state relations, since the analytical and graphical correlating techniques produced results that reflect all the inherent accuracy of the basic data.

The common availability of high speed computing systems for generating thermodynamic quantities from equations of state has likewise generated a need for testing computational programs on definitive data systems (15). The previously published values of experimental P-V-T properties of methane (7), combined with the derived thermodynamic properties, represent a defined data system that is adequate for test purposes, since it includes an internally consistent, matched set of thermodynamic properties that reflect faithfully and consistently all of the trends that appear in a single source of experimental data of high quality and readily documented accuracy. The present work, which covers a temperature range of 0–350°C and a pressure range of 0–400 atm, complements previous results on pure tetrafluoromethane (8, 11). As in the case of tetrafluoromethane, the derived properties are based on unsmoothed experimental compressibility data (7) on a grid of temperatures and densities that forms a framework for the computational work as well as the tables. Thus, no inaccuracies that otherwise would have originated from cross plotting, interpolating, or smoothing of original data were introduced.

No literature data on methane were included in these calculations, because the variations in accuracy and precision that are found in any diverse collection of experimental results would have detracted from the meaning and objective for the present work.

Calculation Methods

Enthalpy, entropy, and Gibbs energy were evaluated from isometric and isothermal data by the following relations, in which T appears on the thermodynamic temperature scale:

$$H - H° = {}_T\!\!\int_0^\rho \frac{[P - T(\partial P/\partial T)_\rho]\,d\rho}{\rho^2} + P/\rho - RT \quad (1)$$

$$S - S° = {}_T\!\!\int_0^\rho \frac{[R\rho - (\partial P/\partial T)_\rho]\,d\rho}{\rho^2} - R \ln RT\rho \quad (2)$$

$$G - G° = {}_T\!\!\int_0^\rho \frac{[P - RT]\rho\,d\rho}{\rho^2} + P/\rho - RT + RT \ln RT\rho \quad (3)$$

The superscript degree mark as in $H°$, $S°$, or $G°$ indicates the ideal gas state at 1-atm pressure. Experimental values of P, T, and ρ were taken from Table I of ref. 7.

[1] To whom correspondence should be addressed.

The slopes, $(\partial P/\partial T)_\rho$, of the isometric lines were derived from the original data by combined analytical and graphical correlating techniques that reflect all of the inherent accuracy of the original measurements. Details of the method used and an analysis of the uncertainties involved in the calculation of the thermodynamic functions from P-V-T data have been fully described (11, 12). Briefly, the maximum uncertainty introduced from the correlational techniques is less than 0.2%, except above 300°C where the uncertainty increases and reaches 2.0% at 350°C.

Previously, gas density work on methane (7) conducted in this laboratory was based on the International Temperature Scale of 1948, 0°C. = 273.16K. The temperatures in the original work were converted to the thermodynamic scale according to a relationship derived by Beattie (3) and then used to calculate the thermodynamic properties. These values were reported in 1966 (13). Recently, however, the temperature scale and the liter were redefined, and recalculation of the values of the thermodynamic properties was necessary.

The present P-V-T data and thermodynamic properties of methane were corrected to the International Practical Temperature Scale of 1968 (6) by using accurate values of $(\partial P/\partial T)_\rho$ and the table of differences between the 1968 and 1948 scales. The pressures were also adjusted to densities in mol dm^{-3} by using accurate values of $dP/d\rho$.

In the determination of the original P-V-T relations for methane, the pressures were measured with a dead-weight gage calibrated against the vapor pressure of pure carbon dioxide, 26,144.7 mm of Hg at 0°, as determined by Bridgeman (4). However, a more recent measurement of the vapor pressure, 26,137.6 mm of Hg by Greig and Dadson (10), is supported by the most recent value, 26,139.7 mm of Hg by Sengers and Chen (17). We did not correct the methane data for this change in pressure calibration, however, because the methane data were originally adjusted by a factor of 1.00023, as previously discussed (7, 9), to take care, collectively, of small systematic errors in the pressure scale, gas constant, atomic weights, variations in C_{12}/C_{13} isotopic ratio, and sample weight.

The molecular weight of the sample was calculated using the values of H = 1.008 and C = 12.011 (7); the value of R used was 82.0560 cm^3 atm deg^{-1} mol^{-1}.

Results

Values for the thermodynamic properties, P, $H - H°$, $S - S°$, and $G - G°$, are presented in Table I at 0.5-mol dm^{-3} intervals in density and 25°C intervals in temperature. Some values are given to an extra significant figure to minimize imprecisions in subsequent calculations rather than to indicate higher accuracy. Values of the properties, $(H - H°)/T$, $(S_P - S_P$, ideal), $(S_V - S_V$, ideal), $(G_P - G_P$, ideal), $(G_V - G_V$, ideal), $(G - G°)/T$, and activity coefficient, γ, were not included in Table I because they are derivable from those values by direct arithmetical computation. They will be sent to interested persons on request.

The effect of changes in the temperature scale and the

Table I. Gram-Molal Thermodynamic Properties[a]

Density, mol dm^{-3}	0.00°C				25.00°C				50.00°C			
	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal
0.75	16.1628	72.7	5.7192	1489.5	17.7850	67.9	5.8861	1687.0	19.3999	64.0	6.0415	1888.3
1.0	21.2765	•96.4	6.3281	1632.2	23.4762	90.0	6.4925	1845.7	25.6676	84.7	6.6463	2063.0
1.5	31.1269	142.8	7.2077	1825.9	34.5376	133.4	7.3676	2063.2	37.9272	125.6	7.5184	2304.0
2.0	40.5170	188.1	7.8529	1956.9	45.2077	175.8	8.0087	2212.0	49.8727	165.2	8.1564	2470.6
2.5	49.4849	232.3	8.3698	2053.9	55.5305	217.0	8.5214	2323.6	61.5317	203.9	8.6662	2596.6
3.0	58.0740	275.3	8.8053	2129.8	65.5511	257.2	8.9531	2412.2	72.9644	241.4	9.0950	2697.6
3.5	66.3449	317.2	9.1846	2191.6	75.3142	296.3	9.3288	2485.1	84.2144	278.0	9.4680	2781.6
4.0	74.3245	358.0	9.5229	2243.2	84.8728	334.3	9.6637	2546.9	95.3284	313.4	9.8002	2853.5
4.5	82.0705	397.6	9.8298	2287.4	94.2681	371.3	9.9674	2600.5	106.378	347.7	10.1013	2916.5
5.0	89.6163	436.2	10.1120	2325.9	103.546	407.2	10.2466	2647.9	117.366	381.1	10.3781	2972.6
5.5	97.0317	473.6	10.3743	2360.1	112.775	441.9	10.5061	2690.5	128.340	413.6	10.6354	3023.3
6.0	104.300	510.2	10.6203	2390.8	121.978	475.7	10.7494	2729.2	139.424	444.8	10.8767	3070.0
6.5	111.582	545.4	10.8526	2419.0	131.213	508.5	10.9794	2765.0	150.707	474.7	11.1046	3113.7
7.0	118.921	579.4	11.0735	2445.3	140.559	540.1	11.1979	2798.6	162.074	504.0	11.3213	3154.5
7.5	126.185	612.8	11.2847	2469.6	150.064	570.6	11.4068	2830.3	173.679	532.1	11.5285	3193.3
8.0	133.642	644.9	11.4875	2492.9	159.769	600.1	11.6075	2860.7	185.697	558.9	11.7275	3230.9
8.5	141.223	676.1	11.6832	2515.2	169.744	628.5	11.8011	2890.0	198.066	584.6	11.9196	3267.2
9.0	149.123	706.0	11.8727	2537.0	180.100	655.8	11.9886	2918.6	210.983	609.0	12.1058	3302.9
9.5	157.228	735.1	12.0568	2558.5	190.910	681.8	12.1710	2946.9	224.414	632.4	12.2868	3338.1
10.0	165.787	762.8	12.2363	2579.5	202.165	706.9	12.3488	2974.9	238.399	654.7	12.4634	3372.9
10.5	174.693	789.7	12.4119	2600.6	214.121	730.6	12.5229	3003.1	253.242	675.5	12.6363	3407.9
11.0	184.345	815.0	12.5840	2622.3	226.706	753.2	12.6938	3031.5	269.002	694.8	12.8061	3443.4
11.5	194.481	839.5	12.7534	2644.1	240.291	774.0	12.8618	3060.7	285.786	712.7	12.9732	3479.6
12.0	205.496	862.4	12.9203	2666.8	254.603	794.0	13.0276	3090.2	303.565	729.4	13.1382	3516.2
12.5	217.404	883.9	13.0853	2690.4	270.236	811.9	13.1915	3121.1	322.733	744.3	13.3015	3554.1

Density, mol dm^{-3}	75.00°C				100.00°C				125.00°C				150.00°C			
	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal	P, atm	$-(H - H°)$, cal	$-(S - S°)$, cal K^{-1}	$G - G°$, cal
0.75	21.0161	60.2	6.1857	2093.3	22.6268	56.7	6.3203	2301.7	24.2366	53.4	6.4463	2513.3	25.8434	50.2	6.5651	2727.8
0.80													27.5558	53.4	6.6992	2781.3
1.0	27.8535	79.8	6.7893	2283.9	30.0343	75.1	6.9229	2508.2	32.2099	70.7	7.0480	2735.5	34.3842	66.5	7.1661	2965.8
1.5	41.3069	118.2	7.6589	2548.3	44.6761	111.2	7.7905	2795.9	48.0390	104.5	7.9139	3046.4	51.3927	98.2	8.0306	3299.9
2.0	54.5031	155.5	8.2946	2732.3	59.1382	146.1	8.4242	2997.4	63.7373	137.4	8.5460	3265.2	68.3408	129.0	8.6614	3536.1
2.5	67.4995	191.7	8.8020	2872.8	73.4528	180.0	8.9299	3152.2	79.3594	169.3	9.0502	3434.1	85.2785	158.7	9.1643	3719.1
3.0	80.3311	226.9	9.2286	2986.1	87.6754	212.9	9.3547	3277.8	94.9706	200.0	9.4736	3571.9	102.256	187.5	9.5865	3869.0
3.5	93.0511	261.0	9.5994	3081.1	101.856	244.8	9.7239	3383.7	110.607	229.7	9.8413	3688.7	119.341	215.1	9.9531	3996.6
4.0	105.700	294.1	9.9296	3162.9	116.042	275.7	10.0524	3475.4	126.319	258.4	10.1686	3790.3	136.560	241.7	10.2792	4107.9
4.5	118.359	326.1	10.2288	3235.1	130.290	305.5	10.3502	3556.7	142.199	285.8	10.4650	3880.9	153.995	267.3	10.5746	4207.4
5.0	131.053	357.0	10.5038	3299.9	144.671	334.2	10.6237	3630.0	158.248	312.3	10.7373	3962.7	171.707	291.7	10.8459	4297.7
5.5	143.850	386.9	10.7593	3358.9	159.216	361.9	10.8778	3697.2	174.573	337.7	10.9903	4038.1	189.761	315.0	11.0980	4381.1
6.0	156.827	415.6	10.9988	3413.6	174.007	388.4	11.1161	3759.5	191.167	362.2	11.2274	4108.0	208.229	337.2	11.3342	4458.9
6.5	169.953	443.5	11.2252	3464.5	189.085	414.0	11.3411	3818.0	208.255	385.1	11.4514	4174.3	227.188	358.1	11.5574	4532.4
7.0	183.445	470.0	11.4404	3512.9	204.643	438.0	11.5551	3873.8	225.714	407.3	11.6644	4236.9	246.708	377.9	11.7696	4602.4
7.5	197.206	495.7	11.6462	3558.9	220.525	461.3	11.7597	3926.9	243.821	427.9	11.8681	4297.4	266.866	396.4	11.9725	4669.8

(continued — high-density rows)

ρ				
8.0	211.431	520.1	11.8440	3603.4
8.5	226.211	543.2	12.0350	3646.8
9.0	241.581	565.1	12.2201	3689.3
9.5	257.629	585.8	12.4001	3731.3
10.0	274.432	605.2	12.5759	3773.1
10.5	292.165	623.2	12.7479	3815.0
11.0	310.935	639.8	12.9169	3857.3
11.5	330.748	655.0	13.0832	3899.9
12.0	352.016	668.4	13.2475	3943.7
12.5	375.056	679.5	13.4103	3989.3

ρ				
8.0	287.822	413.4	12.1676	4735.3
8.5	309.522	429.4	12.3559	4799.0
9.0	332.248	443.8	12.5385	4861.9
9.5	356.014	456.7	12.7162	4924.1
10.0	381.112	467.8	12.8897	4986.5

175.00°C

ρ				
0.75	27.4496	47.2	6.6773	2945.2
0.80	29.2741	50.2	6.8113	3002.2
1.0	36.5561	62.5	7.2776	3199.0
1.5	54.7406	92.2	8.1410	3556.1
2.0	72.9268	121.0	8.7706	3809.5
2.5	91.1652	148.8	9.2723	4006.6
3.0	109.514	175.5	9.6934	4168.7
3.5	128.032	201.1	10.0590	4306.9
4.0	146.786	225.5	10.3842	4428.2
4.5	165.812	249.0	10.6787	4536.7
5.0	185.134	271.6	10.9492	4635.3
5.5	204.991	292.5	11.2005	4726.9
6.0	225.293	312.5	11.4360	4812.5
6.5	246.179	331.3	11.6584	4893.4
7.0	267.679	348.9	11.8699	4970.6
7.5	289.972	365.1	12.0722	5045.1
8.0	313.184	379.7	12.2666	5117.6
8.5	337.273	393.1	12.4544	5188.3
9.0	362.434	405.0	12.6364	5258.0
9.5	388.946	415.0	12.8135	5327.4

200.00°C

ρ				
0.75	30.9899	47.1	6.9173	3225.8
1.0	38.7246	58.6	7.3833	3434.8
1.5	58.0819	86.4	8.2455	3814.9
2.0	77.5075	113.2	8.8741	4085.6
2.5	97.0515	138.9	9.3748	4296.8
3.0	116.766	163.9	9.7950	4470.8
3.5	136.704	187.4	10.1597	4619.7
4.0	156.942	210.0	10.4840	4750.5
4.5	177.546	231.4	10.7777	4868.1
5.0	198.558	251.8	11.0474	4975.3
5.5	220.124	270.8	11.2979	5074.8
6.0	242.191	288.9	11.5327	5167.8
6.5	265.041	305.3	11.7545	5256.3
7.0	288.536	320.8	11.9654	5340.7
7.5	312.903	334.8	12.1671	5422.1
8.0	338.357	347.0	12.3609	5501.6
8.5	364.629	358.4	12.5481	5578.7
9.0	392.351	367.5	12.7296	5655.5

225.00°C

ρ				
0.75	32.7054	44.1	7.0180	3452.0
1.0	40.8922	54.8	7.4836	3673.1
1.5	61.4192	80.8	8.3449	4076.2
2.0	82.0793	105.6	8.9725	4364.0
2.5	102.907	129.6	9.4724	4589.1
3.0	123.988	152.4	9.8917	4775.2
3.5	145.361	174.1	10.2556	4934.8
4.0	167.103	194.7	10.5792	5075.3
4.5	189.293	214.1	10.8722	5201.9
5.0	211.999	232.2	11.1412	5317.8
5.5	235.232	249.5	11.3911	5425.0
6.0	259.163	265.3	11.6253	5525.8
6.5	283.826	280.0	11.8466	5621.4
7.0	309.404	293.0	12.0570	5713.2
7.5	335.858	304.8	12.2582	5801.6
8.0	363.407	315.1	12.4516	5887.7
8.5	392.088	323.9	12.6383	5971.9

250.00°C

ρ				
0.75	34.4191	41.1	7.1139	3680.5
1.0	43.0580	51.1	7.5790	3913.9
1.5	64.7539	75.2	8.4395	4339.9
2.0	86.6447	98.2	9.0663	4644.9
2.5	108.762	120.3	9.5654	4883.8
3.0	131.189	141.2	9.9838	5081.8
3.5	153.994	160.9	10.3469	5252.1
4.0	177.233	179.6	10.6698	5402.3
4.5	200.982	197.0	10.9620	5537.8
5.0	225.336	213.2	11.2304	5662.0
5.5	250.303	228.3	11.4797	5777.3
6.0	276.064	242.0	11.7133	5885.8
6.5	302.611	254.6	11.9340	5988.7
7.0	330.154	265.5	12.1439	6087.6
7.5	358.720	275.1	12.3446	6183.0
8.0	388.438	283.1	12.5375	6275.9

275.00°C

ρ				
0.80	36.1328	38.0	7.2050	3911.4
1.0	45.2226	47.2	7.6697	4157.0
1.5	68.0836	69.4	8.5292	4605.8
2.0	91.2081	90.4	9.1551	4928.0
2.5	114.616	110.5	9.6532	5180.9
3.0	138.398	129.5	10.0708	5390.8
3.5	162.609	147.3	10.4330	5571.6
4.0	187.346	163.8	10.7550	5731.5
4.5	212.676	179.1	11.0464	5876.0
5.0	238.638	193.3	11.3140	6008.4
5.5	265.337	206.3	11.5625	6131.7
6.0	292.934	217.7	11.7954	6247.9
6.5	321.362	228.1	12.0153	6358.1
7.0	350.882	236.8	12.2246	6464.1
7.5	381.501	244.2	12.4247	6566.4

300.00°C

ρ				
0.80	37.8421	35.1	7.2922	4144.4
1.0	47.3836	43.6	7.7567	4402.2
1.5	71.4083	64.1	8.6154	4873.8
2.0	95.7473	83.4	9.2406	5212.9
2.5	120.439	101.7	9.7381	5479.6
3.0	145.561	118.9	10.1550	5701.4
3.5	171.204	134.8	10.5166	5892.8
4.0	197.410	149.5	10.8380	6062.3
4.5	224.261	163.0	11.1287	6215.4
5.0	251.899	175.0	11.3956	6356.4
5.5	280.289	185.9	11.6433	6487.4
6.0	309.698	195.1	11.8754	6611.4
6.5	340.012	203.2	12.0947	6728.9
7.0	371.421	210.0	12.3032	6841.6

325.00°C

ρ				
0.80	39.5509	33.0	7.3769	4379.5
1.0	49.5458	40.8	7.8414	4649.5
1.5	74.7320	60.2	8.7003	5143.9
2.0	100.298	78.2	9.3258	5500.0
2.5	126.270	95.4	9.8237	5780.6
3.0	152.762	111.3	10.2411	6014.5
3.5	179.791	126.1	10.6033	6216.2
4.0	207.482	139.6	10.9252	6395.2
4.5	235.890	151.9	11.2165	6557.3
5.0	265.126	162.7	11.4839	6706.4
5.5	295.247	172.1	11.7321	6845.4
6.0	326.397	179.9	11.9645	6976.7
6.5	358.566	186.4	12.1838	7101.4
7.0	391.813	191.6	12.3921	7220.7

350.00°C

ρ				
0.80	41.2649	31.2	7.4589	4616.8
1.0	51.7083	38.6	7.9236	4899.0
1.5	78.0612	56.8	8.7828	5416.2
2.0	104.848	73.7	9.4088	5789.4
2.5	132.123	89.6	9.9071	6084.0
3.0	159.930	104.5	10.3249	6329.5
3.5	188.422	117.7	10.6875	6542.2
4.0	217.582	130.1	11.0098	6730.7
4.5	247.556	140.9	11.3016	6901.7
5.0	278.440	150.2	11.5694	7059.3
5.5	310.279	158.2	11.8180	7206.2
6.0	343.182	164.6	12.0508	7344.8
6.5	377.322	169.3	12.2707	7477.2

a Tabulated pressures were adjusted to 1968 IPTS and to even values of density in mol dm^{-3} from values previously published (7). $(S - S°)$ was determined by difference. 1 atm = 101 325 Pa.

Table II. Deviation^a of Published from Present Values of $(H - H^\circ)$, Cal Mol^{-1}

t, °C		Density, mol dm^{-3}						
		0.75	2.00	4.00	6.00	8.00	10.00	12.00
0	M&N (14)	−0.6	−0.1	−0.1	−2.9	−3.9	−3.8	
50	M&N	−0.7	0.8	0.2	−0.6	−0.3	1.1	
	S&L (16)	0.8	0.9	−0.4	−0.7	−0.5	1.9	2.8
100	M&N	0.8	1.9	1.0	1.6	2.2	2.2	
	S&L	2.5	5.3	8.7	10.5	11.8	12.6	12.7
150	M&N	2.5	5.8	8.3	9.4	9.0	11.0	
	S&L	1.2	3.0	5.0	6.9	9.5	9.9	

^{a} Deviation $= (H - H^\circ)_{\text{present}} - (H - H^\circ)_{\text{other}}$.

redefinition of the liter on the magnitude of the thermodynamic properties was minimal. The average and maximum percentage changes applied to the values of the functions were 0.05 and 0.22 for $(H - H^\circ)$; 0.003 and 0.01 for $(S - S^\circ)$; and 0.008 and 0.024 for $(G - G^\circ)$. These percentage variations are typical of all of the functions except $(G_P - G_P, \text{ideal})$ and $(G_V - G_V, \text{ideal})$ which change sign within the ranges of the variables.

The present values of the enthalpy, $(H - H^\circ)$, are compared with the values of Michels and Nederbragt (14) and of Sage and Lacey (16) in Table II, which are the only other extensive sets that were derived from individual sources of P-V-T data. In general, the best agreement is with the results of Michels and Nederbragt. Although Table II is brief, it shows good agreement in the thermodynamic properties derived from primary sources of P-V-T data. In contrast, the use of averaged or correlated P-V-T values from several sources, according to the practice found in several literature correlations (1, 2, 5, 18, 19), generally produced significantly larger differences.

Nomenclature

G = molal Gibbs energy, cal mol^{-1}
H = molal enthalpy, cal mol^{-1}
P = pressure, atm
R = gas constant = 82.0560 cc atm mol^{-1} K
S = molal entropy, cal K^{-1} mol^{-1}
T = thermodynamic temperature, K = t + 273.15
t = International Practical Celsius temperature, 1968
V = molal volume, dm^3 mol^{-1}
ρ = molal density, mol dm^{-3}
γ = activity coefficient = exp. $[(G - G^\circ)/RT]/P$
atm = 101,325 Pa
cal = 4.184 J

Subscripts

ideal = ideal gas state
P = constant pressure along isotherms
V = constant volume along isotherms
ρ = constant density
T = constant temperature

Superscripts

$\circ$ = standard state

Literature Cited

(1) American Petroleum Institute, Division of Refining, Washington, D.C.,"Technical Data Book" (loose-leaf sheets, extant April 1971).
(2) American Petroleum Institute Research Project 44, Chemical Thermodynamics Properties Center, Texas A&M University, College Station, Tex., "Selected Values of Properties of Hydrocarbons and Related Compounds" (loose-leaf sheets, extant Dec. 31, 1971).
(3) Beattie, J. A., "Temperature, Its Measurement and Control in Science and Industry," Vol. II, Chap. 5, Reinhold, New York, N.Y., 1955.
(4) Bridgeman, O. C., J. Amer. Chem. Soc., **49**, 1174 (1927).
(5) Canjar, L. N., Manning, F. S., "Thermodynamic Properties and Reduced Correlations for Gases," Gulf Publishing Co., Houston, Tex., 1967.
(6) Comité International des Poids et Mesures, Metrologia, **5** (2), 35 (1969).
(7) Douslin, D. R., Harrison, R. H., Moore, R. T., McCullough, J. P., J. Chem. Eng. Data, **9**, 358 (1964).
(8) Douslin, D. R., Harrison, R. H., Moore, R. T., McCullough, J. P., J. Chem. Phys., **35** (4), 1357 (1961).
(9) Douslin, D. R., Harrison, R. H., Moore, R. T., J. Phys. Chem., **71**, 3477 (1967).
(10) Greig, R. G. P., Dadson, R. S., Brit. J. Appl. Phys., **17**, 1633 (1966).
(11) Harrison, R. H., Douslin, D. R., J. Chem. Eng. Data, **11**, 383 (1966).
(12) Harrison, R. H., Douslin, D. R., "Perfluorocyclobutane Thermodynamic Properties of the Real Gas," U.S. Bur. Mines Rept. Invest. **6475** (1964).
(13) Harrison, R. H., Moore, R. T., Douslin, D. R., "Thermodynamic Properties of Compressed Gases," AFOSR Final Technical Summary Report, AD-647-893, 1966.
(14) Michels, A., Nederbragt, G. W., Physica, **3**, 569 (1936).
(15) Morsy, T. E., J. Chem. Eng. Data, **15**, 256 (1970).
(16) Sage, B. H., Lacey, W. N., "Thermodynamic Properties of the Lighter Paraffin Hydrocarbons and Nitrogen," API Research Project 37, American Petroleum Institute, New York, N.Y., 1950.
(17) Sengers, J. M. H. L., Chen, W. T., J. Chem. Phys., **56** (1), 595 (1972).
(18) Tester, H. E., "Thermodynamic Functions of Gases," F. Din, Ed., Vol. III, pp. 1–71, Butterworths, London, 1961.
(19) Zagoruchenko, V. A., Zhuravlev, A. M., "Thermophysical Properties of Gaseous and Liquid Methane" (Teplofizicheskie svoistva gazoobraznogo i zhidkogo metana), Moscow, Izdatel'stvo Komiteta Standartov, Mer i Izmeritel' nykh Priborov pri Sovete Ministrov SSSR, 1969 (in English, translated from Russian through Israel Program for Scientific Translations, Jerusalem 1970).