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THERMODYNAMIC PROPERTIES OF R134a (1,1,1,2-TETRAFLUOROETHANE)[1]

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ABSTRACT

The thermodynamic surface of R134a is expressed in terms of a 32-term modified Benedict-Webb-Rubin (MBWR) equation of state. Coefficients for this equation and for the ancillary equations representing the saturated liquid and vapor densities and the vapor pressure are presented. Temperatures are given on the new International Temperature Scale of 1990 (ITS 90). The MBWR coefficients were obtained using a multi-property fit that used experimental data for PVT properties, isochoric heat capacity, second virial coefficients, speed of sound, and coexistence properties, including new measurements not used in previous formulations. The formulation is applicable to both the liquid and vapor phases at pressures up to 70 MPa, and for a temperature range from the triple point to 450 K. The accuracy of the equation of state is estimated to be ±0.2 % in density, ±1.0 % in constant-volume heat capacities, and ±0.6 % in the sound velocity, except in the critical region, based on comparisons with experimental data. In addition, a comprehensive bibliography of experimental data for R134a is presented.

Keywords: equation of state; heat capacity; saturated liquid density; saturated vapor density; sound speed; refrigerants; R134a; vapor pressure

INTRODUCTION

Chlorofluorocarbons (CFC's) are a class of chemical compounds that have been implicated as a cause of the depletion of the ozone layer and as a potential source of global warming [1]. Due to their detrimental effects on the environment, an international treaty known as the Montreal Protocol dictates a schedule for phasing these substances out. In particular, the treaty specifies that R12 (dichlorodifluoromethane) is to be totally phased-out by the year 2000. In the United States, even stricter requirements have been set— R12 is to be phased-out by December 31, 1995. R12 has been widely used as a refrigerant, a foam blowing agent, and an aerosol propellant. R134a has emerged as the leading substitute for R12. It has the desirable properties of being chemically stable, has so far indicated very low acute and chronic toxicity, has zero ozone depletion potential and a small global warming potential [1]. There has been much interest in the measurement and the representation of the thermophysical property surfaces of R134a, since it is important to accurately know the thermophysical properties for proper equipment design. In this work, experimental data from the literature are used to develop a modified Benedict-Webb-Rubin [2] equation of state. The results of the correlation are compared with experimental data.

EXPERIMENTAL DATA

Table 1 lists the experimental pressure-volume-temperature (PVT) data sets for R134a. A subset of the available data, shown in *Table 1* with an asterisk, was used in the determination of the coefficients to represent the thermodynamic surface. The subset covers a broad range of temperature, 180-453 K, and pressures up to 70 MPa.

Table 1. Summary of experimental PVT data for R134a

Source	Number of data points	Pressure range MPa	Temperature range K
Zhu et al. [3]	42	0.14-1.27	283-353
Tillner-Roth and Baehr [4]*	411	0.09-16.4	293-453
Hou et al. [5]*	429	0.75-70.9	180-380
Weber [6]*	69	0.22-5.33	321-423
Wilson and Basu [7]	51	1.06-6.67	317-448
Piao et al. [8]	157	0.80-11.8	313-423
Magee [9]	150	2.58-34.9	187-343
Morrison and Ward [10]*	131	0.70-5.78	279-367
Maezawa et al. [11]	10	0.51-2.00	280-340
Baroncini et al. [12]	46	0.18-1.94	263-359
Fukushima [13]	63	0.55-5.73	294-424
Qian et al. [14]	21	0.13-1.93	320-340

Tables 2, 3 and *4* summarize the saturation boundary data for R134a. *Table 5* summarizes available data for other thermodynamic properties of R134a. The symbol w denotes the sound speed, and B is the second virial coefficient. The sets marked with an asterisk were used to develop the equation of state. Ideal gas heat capacities were obtained from the correlation developed by McLinden et al. [15], based on the sound speed data of Goodwin and Moldover [31].

Table 2. Summary of experimental saturated vapor densities for R134a

Source	Number of data points	Density range mol/dm^3	Temperature range K
Fukushima et al. [16]	9	3.15-4.91	371-374
Weber [6]*	5	0.60-2.33	320-365
Kabata et al. [17]	15	2.04-4.97	361-374
Morrison and Ward [10]	8	0.37-2.62	298-368
Niesen et al. [18]*	14	0.49-2.95	312-370
Fukushima [13]	6	0.28-3.07	293-371

Table 3. Summary of experimental saturated liquid densities for R134a

Source	Number of data points	Density range mol/dm^3	Temperature range K
Fukushima [19]*	7	12.08-13.58	244-292
Fukushima et al. [16]	8	5.04-7.31	369-374
Yokoyama and Takahashi [20]*	21	7.72-13.34	252-367
Hou et al. [5]*	10	8.49-15.29	180-360
Wilson and Basu [7]	9	6.86-13.73	239-372
Piao et al. [8]	7	6.79-11.22	313-372
Kabata et al. [17]	12	5.05-9.79	343-374
Morrison and Ward [10]*	27	5.05-12.80	268-374
Niesen et al. [18]*	14	7.18-11.26	312-370
Maezawa et al. [11]*	25	7.32-12.44	280-370
Fukushima [13]	3	8.79-10.80	323-357

Table 4. Summary of experimental vapor pressure data for R134a

Source	Number of data points	Pressure range MPa	Temperature range K
Goodwin et al. [21]*	79	0.02-3.97	214-373
Arita et al. [22]	3	0.29-1.32	273-323
Zhu et al. [23]	43	0.36-3.23	279-363
Baehr and Tillner-Roth [24]*	37	0.77-4.05	303-374
Weber [6]*	22	1.02-3.97	313-373
Wilson and Basu [7]	32	0.01-3.66	211-369
Piao et al. [8]	51	0.88-4.06	308-374
Kubota et al. [25]	25	0.13-3.97	253-373
Nishiumi and Yokoyama [26]	16	0.10-4.03	247-373
Magee and Howley [27]	19	0.001-2.46	180-350
Morrison and Ward [10]*	12	0.24-4.07	268-374
Niesen et al. [18]	14	0.99-3.77	312-370
Maezawa et al. [11]	14	0.38-2.46	280-350
Baroncini et al. [12]	64	0.08-2.98	243-359
Fukushima [13]	41	0.19-3.87	262-372

Table 5. Summary of single-phase sound speed, heat capacity, and virial coefficient experimental data for R134a

Source	Data type	Region	Temperature range, K	Pressure range, MPa
Magee [9]*	C_v	L	187-343	2.6-34.9
Saitoh et al. [28]	C_p	L	275-356	1.0-3.0
Nakagawa et al. [29]	C_p	L	273-356	sat-3.0
Guedes et al. [30]*	w	L	179-380	0.1-71
Goodwin and Moldover [31]*	w	V	231-340	0.01-0.6
Takagi [32]	w	L	290-379	sat-75
Goodwin and Moldover [31]*	B	V	235-440	
Weber [6]*	B	V	323-423	
Tillner-Roth and Baehr [4]*	B	V	293-453	

FORMULATION OF THE THERMODYNAMIC SURFACE

The modified Benedict-Webb-Rubin equation of state proposed by Jacobsen and Stewart [2] is used to represent the thermodynamic surface. The equation is wide-ranging in temperature and pressure and is applicable in both the liquid and vapor regions, except near the critical point. It has been applied successfully to hydrocarbons [33], common inorganics and cryogenic fluids [34]. It also has been used previously [15,35] to represent R134a; the availability of new, more accurate data makes it desirable to refit the MBWR equation of state. The functional form for this equation is essentially a polynomial in density and temperature:

$$P = \sum_{n=1}^{9} a_n(T)\rho^n + e^{-(\rho/\rho_c)^2} \sum_{n=10}^{15} a_n(T)\rho^{2n-17}. \qquad (1)$$

The temperature dependence of the $a_n(T)$ is summarized in *Table 6*.

Table 6. Temperature dependence of the MBWR coefficients.

$a_1 = RT$
$a_2 = b_1T + b_2T^{0.5} + b_3 + b_4/T + b_5/T^2$
$a_3 = b_6T + b_7 + b_8/T + b_9/T^2$
$a_4 = b_{10}T + b_{11} + b_{12}/T$
$a_5 = b_{13}$
$a_6 = b_{14}/T + b_{15}/T^2$
$a_7 = b_{16}/T$
$a_8 = b_{17}/T + b_{18}/T^2$
$a_9 = b_{19}/T^2$
$a_{10} = b_{20}/T^2 + b_{21}/T^3$
$a_{11} = b_{22}/T^2 + b_{23}/T^4$
$a_{12} = b_{24}/T^2 + b_{25}/T^3$
$a_{13} = b_{26}/T^2 + b_{27}/T^4$
$a_{14} = b_{28}/T^2 + b_{29}/T^3$
$a_{15} = b_{30}/T^2 + b_{31}/T^3 + b_{32}/T^4$

ANCILLARY EQUATIONS

In the development of the equation of state, it is desirable to ensure high accuracy in the region near the saturation boundary since many refrigeration systems operate in this regime. We accomplish this by heavily weighting the saturation boundary. To do this, we desire values of the saturated liquid and vapor densities, their temperature derivatives, and the vapor pressure at common temperatures. Since these usually are not available directly from experimental data, it is useful to fit saturation data to ancillary functions, and to use the ancillary functions in fitting the equation of state. These ancillary functions are used only to develop the final equation of state. The forms we have chosen to use for the vapor pressure P_σ, the saturated liquid density $\rho_{\sigma L}$, and the saturated vapor density $\rho_{\sigma V}$ are:

$$\ln\left[\frac{P_\sigma}{P_C}\right] = c_1\tau^{\varepsilon} + c_2\frac{\tau}{(1-\tau)} + c_3\tau + c_4\tau^3, \tag{2}$$

$$\rho_{\sigma L} = \rho_C\left[1 + \frac{d_1\tau^{\beta} + d_3\tau^2 + d_4\tau^3}{1 + d_2\tau^{1-\beta}}\right] \tag{3}$$

$$\rho_{\sigma V} = \frac{P_\sigma}{RT}\left[1 + \left(\frac{P_\sigma}{P_C}\right)\left(\frac{T_C}{T}\right)(Z_C - 1)\left(1 + f_1\tau^{\beta} + f_2\tau + f_3\tau^2 + f_4\tau^3\right)\right]^{-1}, \tag{4}$$

where $T_r = T/T_C$, $\tau = 1 - T/T_C$, $Z_C = P_C/(\rho_C R T_C)$, and $\varepsilon = 1.66$ and $\beta = 0.35$ are empirically determined exponents. For the critical point, we have used $T_C = 374.179$ K (ITS 90) [15], $\rho_C = 5.0308$ mol/dm^3, and $P_C = 4.056$ MPa. The critical pressure and density were determined by the fit.

The data marked by asterisks in *Tables 2-4* were used with a linear least-squares fitting routine, adapted from an algorithm developed at NIST [36]. In addition, at low temperatures (less than 260 K) vapor densities were generated by intersecting the virial equation using second virial coefficients given in Goodwin and Moldover [31] with the vapor pressure equation given in Eq.(2), since experimental values were not available at these temperatures. The resulting coefficients are in *Table 7*.

Table 7. Coefficients for ancillary equations Eq. (2)-(4).

i	1	2	3	4
c_i	+3.946 984	-11.313 271	+3.693 108	+5.566 337
d_i	+2.081 196	-0.413 003	-1.177 335	+1.116 197
f_i	-0.902 329	+2.474 527	-2.762 865	+16.870 784

RESULTS

The coefficients obtained from multiproperty linear least-squares fitts [36,37] to the thermodynamic surface of R134a are given in *Table 8*. The coefficients are for use in Equation (1) with temperature in K, pressure in bar, density in mol/dm^3, and R=0.08314471 l•mol•bar^{-1}•K^{-1}. Comparisons of density values are shown in *Figure 1*, using the data sets used in the fits [4-6,10]. The average absolute percent deviation (AAD) is 0.10, the bias 0.05% and the root mean square error 0.188%. Figure 1 shows that, except in the critical region, all of the points are about 0.2% error or less. Comparisons of pressure show an AAD, bias and RMS error of 1.39%, -0.12%, and 3.77%. We found that for R134a the MBWR can be used successfully except when the temperature is within 10 degrees of the critical, or 364-384 K. When all of the available data sets for density in *Table 1* are considered, the overall average absolute percent deviation in density is 0.18%, and the overall average absolute percent deviation in pressure is 1.5%. The formulation is applicable to both the liquid and vapor phases at pressures up to 70 MPa, and for a temperature range from the triple point to 450 K. *Figure 2* shows the percent deviations between calculated and experimental isochoric heat capacities, using the data of Magee [9]. The AAD, bias and RMS errors are 0.37%, -0.18%, and 0.40%. The maximum deviations are almost always within 1%. *Figure 3* gives comparisons between calculated and experimental sound speeds, using the data of Goodwin and Moldover [31] and Guedes et al. [30]. The AAD is 0.2, the bias 0.01 and the RMS error 0.38 for these two data sets. Except in the critical region, the maximum deviations are less than approximately 0.6%. *Table 9* gives sample values along the saturation boundary as computed with the equation of state. Finally, in *Figure 4* we present a pressure-enthalpy diagram generated using the new MBWR equation of state. The reference states for enthalpy and entropy are 200 kJ/kg and 1.0 kJ/(kg·K) for the saturated liquid at 0° C.

Table 8. Coefficients for the MBWR equation of state.

b_1	+9.652 093 6222 E-02	b_2	-4.018 247 6889 E+00	b_3	+3.952 395 3286 E+01
b_4	+1.345 328 6896 E+03	b_5	-1.394 397 4135 E+06	b_6	-3.092 813 5518 E-03
b_7	+2.923 815 1228 E+00	b_8	-1.651 466 1356 E+03	b_9	+1.507 060 0312 E+06
b_{10}	+5.349 739 4831 E-05	b_{11}	+5.439 333 1762 E-01	b_{12}	-2.113 260 4976 E+02
b_{13}	-2.681 912 0385 E-02	b_{14}	-5.410 671 2595 E-01	b_{15}	-8.517 317 7940 E+02
b_{16}	+2.051 882 5365 E-01	b_{17}	-7.330 501 8809 E-03	b_{18}	+3.806 559 6386 E+00
b_{19}	-1.058 320 8759 E-01	b_{20}	-6.792 430 8442 E+05	b_{21}	-1.269 983 7860 E+08
b_{22}	-4.262 344 3183 E+04	b_{23}	+1.019 733 3823 E+09	b_{24}	-1.866 995 2678 E+02
b_{25}	-9.334 263 2342 E+04	b_{26}	-5.717 352 0896 E+00	b_{27}	-1.767 627 3879 E+05
b_{28}	-3.972 827 5231 E-02	b_{29}	+1.430 168 4480 E+01	b_{30}	+8.030 852 9426 E-05
b_{31}	-1.719 590 7355 E-01	b_{32}	+2.262 383 8566 E+00		

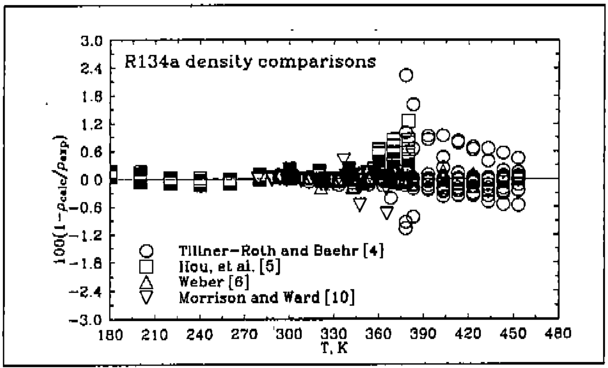

Figure 1 R134a density comparisons.

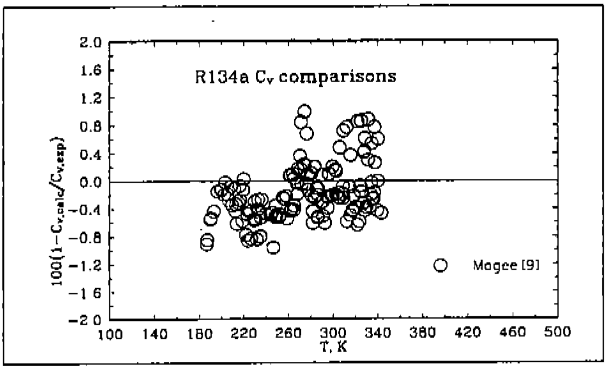

Figure 2 R134a C_v comparisons.

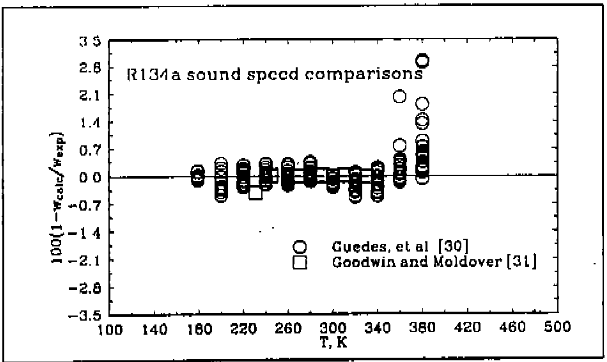

Figure 3 R134a sound speed comparisons.

Table 9. Properties of saturated liquid and saturated vapor calculated with the MBWR equation of state.

Temp* (°C)	Pressure (MPa)	Density (kg/m^3)		Enthalpy (kJ/kg)		Entropy (kJ/kg·K)		C_p (kJ/kg·K)		Sound Vel. (m/s)	
		liq	vap	liq	vap	liq	vap	liq	vap	liq	vap
–103.30^a	0.00039	1591.2	0.028	71.89	335.08	0.4143	1.9638	1.147	0.585	1135.	127.
–100.00	0.00056	1581.9	0.040	75.71	337.00	0.4366	1.9456	1.168	0.592	1111.	128.
–95.00	0.00094	1567.9	0.065	81.61	339.95	0.4701	1.9203	1.189	0.603	1079.	129.
–90.00	0.00153	1553.9	0.103	87.59	342.94	0.5032	1.8975	1.201	0.614	1051.	131.
–85.00	0.00241	1540.0	0.158	93.61	345.97	0.5357	1.8769	1.207	0.625	1025.	133.
–80.00	0.00369	1526.2	0.235	99.65	349.03	0.5674	1.8585	1.211	0.637	999.	134.
–75.00	0.00550	1512.4	0.342	105.71	352.12	0.5983	1.8419	1.213	0.648	975.	135.
–70.00	0.00801	1498.6	0.487	111.78	355.23	0.6286	1.8269	1.215	0.660	951.	137.
–65.00	0.01141	1484.8	0.679	117.86	358.36	0.6581	1.8136	1.217	0.672	928.	138.
–60.00	0.01594	1471.0	0.929	123.96	361.51	0.6871	1.8016	1.220	0.685	904.	139.
–55.00	0.02186	1457.1	1.248	130.07	364.67	0.7154	1.7908	1.224	0.698	881.	141.
–50.00	0.02948	1443.1	1.651	136.21	367.83	0.7432	1.7812	1.229	0.712	858.	142.
–45.00	0.03914	1429.0	2.153	142.37	371.00	0.7705	1.7726	1.235	0.726	835.	143.
–40.00	0.05122	1414.8	2.770	148.57	374.16	0.7973	1.7649	1.243	0.740	812.	144.
–35.00	0.06614	1400.4	3.521	154.81	377.31	0.8238	1.7580	1.251	0.756	789.	144.
–30.00	0.08436	1385.9	4.426	161.10	380.45	0.8498	1.7519	1.260	0.771	765.	145.
–26.07^b	0.10132	1374.3	5.259	166.07	382.90	0.8701	1.7476	1.268	0.784	747.	146.
–25.00	0.10637	1371.2	5.505	167.43	383.57	0.8755	1.7465	1.270	0.788	742.	146.
–20.00	0.13268	1356.2	6.782	173.82	386.66	0.9009	1.7417	1.282	0.805	719.	146.
–15.00	0.16387	1341.0	8.284	180.27	389.72	0.9261	1.7375	1.293	0.823	696.	147.
–10.00	0.20052	1325.6	10.037	186.78	392.75	0.9509	1.7337	1.306	0.842	672.	147.
–5.00	0.24324	1309.8	12.071	193.35	395.74	0.9756	1.7303	1.320	0.862	649.	147.
0.00	0.29269	1293.7	14.420	200.00	398.68	1.0000	1.7274	1.335	0.883	626.	147.
5.00	0.34954	1277.1	17.120	206.72	401.57	1.0242	1.7247	1.350	0.906	602.	147.
10.00	0.41449	1260.2	20.212	213.53	404.40	1.0483	1.7224	1.367	0.930	579.	146.
15.00	0.48825	1242.8	23.740	220.42	407.16	1.0722	1.7203	1.385	0.955	555.	146.
20.00	0.57159	1224.9	27.756	227.40	409.84	1.0960	1.7183	1.404	0.982	531.	145.
25.00	0.66526	1206.3	32.318	234.47	412.44	1.1197	1.7166	1.425	1.012	508.	144.
30.00	0.77008	1187.2	37.494	241.65	414.94	1.1432	1.7149	1.447	1.044	484.	143.
35.00	0.88685	1167.2	43.361	248.94	417.32	1.1668	1.7132	1.472	1.080	460.	142.
40.00	1.0165	1146.5	50.016	256.35	419.58	1.1903	1.7115	1.500	1.120	436.	140.
45.00	1.1598	1124.8	57.570	263.90	421.69	1.2138	1.7097	1.532	1.166	411.	139.
50.00	1.3177	1102.0	66.164	271.59	423.63	1.2373	1.7078	1.569	1.218	387.	137.
55.00	1.4913	1078.0	75.973	279.44	425.36	1.2609	1.7056	1.611	1.280	362.	134.
60.00	1.6815	1052.4	87.226	287.49	426.86	1.2847	1.7031	1.663	1.354	338.	132.
65.00	1.8895	1025.1	100.22	295.76	428.06	1.3088	1.7000	1.726	1.447	313.	129.
70.00	2.1165	995.6	115.38	304.29	428.89	1.3332	1.6963	1.806	1.567	287.	126.
75.00	2.3639	963.3	133.31	313.14	429.26	1.3580	1.6916	1.915	1.730	262.	122.
80.00	2.6331	927.4	154.91	322.41	429.02	1.3837	1.6855	2.069	1.967	235.	118.
85.00	2.9259	886.2	181.75	332.27	427.91	1.4105	1.6775	2.313	2.348	207.	113.
90.00	3.2445	836.9	216.80	343.01	425.48	1.4392	1.6663	2.766	3.064	178.	108.
95.00	3.5916	771.6	267.60	355.43	420.60	1.4720	1.6490	3.961	4.942	145.	102.
100.00	3.9721	646.7	377.3	374.03	407.08	1.5207	1.6093			105.	94.
101.03^c	4.0560	513.3	513.3	389.79	389.79	1.5593	1.5593	∞	∞	0.	0.

*temperatures are on the ITS-90 scale

^{a}triple point; ^{b}normal boiling point; ^{c}critical point

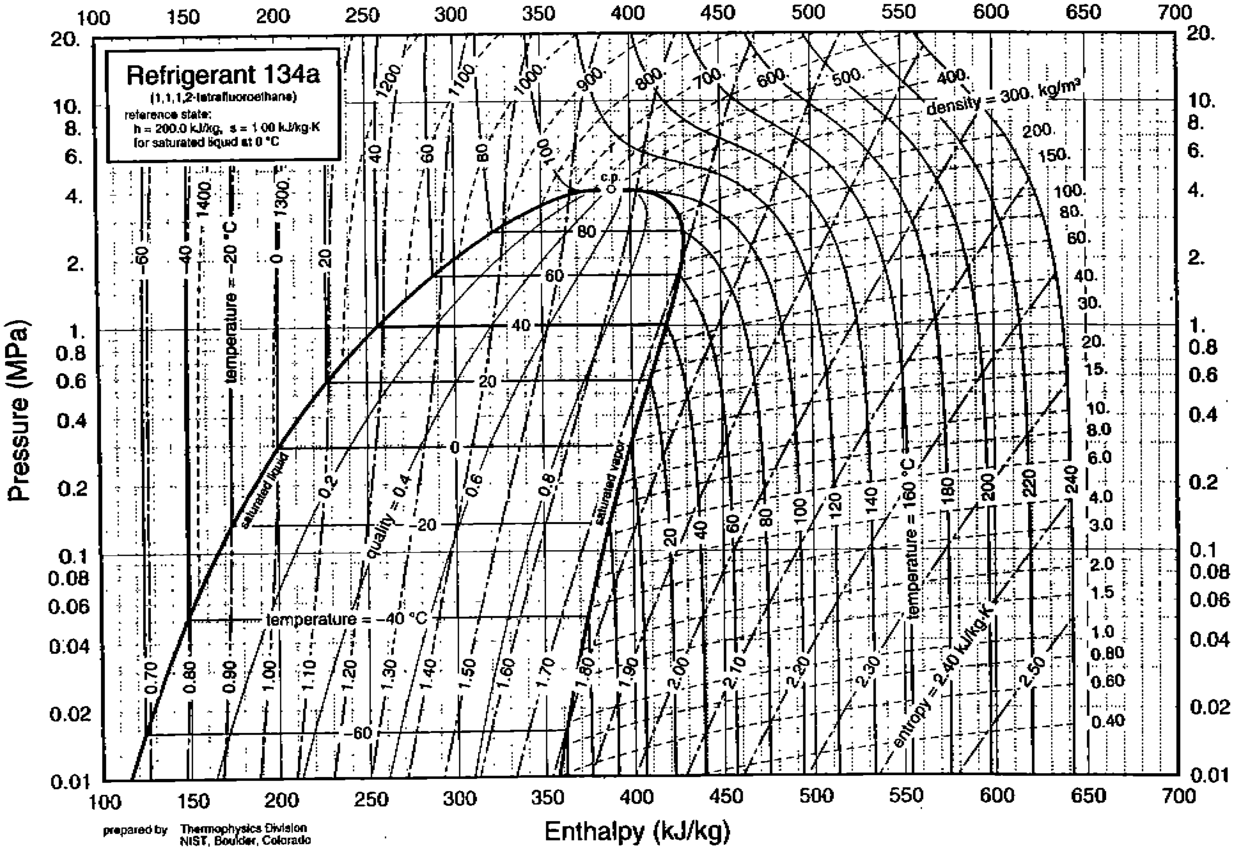

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Figure 4. Pressure-enthalpy chart for R134a.

CONCLUSIONS

We present the coefficients resulting from multiproperty fitting of experimental PVT, sound speed, second virial coefficient, and isochoric heat capacity data to an MBWR equation of state for R134a. We also present the coefficients of ancillary equations for vapor pressure, and for saturated liquid and vapor densities. Comparisons with experimental data are given. The equation represents the density of R134a to within 0.2%, the sound speed to within 0.6%, and C_V to within 1.0%, except in the critical region. A pressure-enthalpy diagram generated with the new equation of state is also presented.

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