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Specific Enthalpy Increments for Pentan-1-ol at Temperatures up to 623.2 K and 10.1 MPa

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Measurements of specific enthalpy increments for pentan-1-ol are reported. A countercurrent water-cooled flow calorimeter was used to measure 102 enthalpy increments over the temperature range 448.2 K to 623.2 K at pressures from 0.1 MPa up to 10.1 MPa. Extrapolation of the gas-phase measurements to zero pressure gave values in excellent agreement with pure component ideal gas enthalpies calculated by extrapolating data on lower alcohols. Values of the specific enthalpy of vaporization derived from the measurements are in agreement with other work and are well fitted by a modification of the Watson equation. A method for the calculation of the two-phase enthalpy−pressure envelope is described.

Introduction

Measurements of specific enthalpy increments of the alcohols methanol (Yerlett and Wormald, 1986), ethanol (Vine and Wormald, 1989), propan-1-ol (Vine and Wormald, 1999), propan-2-ol (Wormald and Vine, 1999), and butan-1-ol (Wormald and Fennell, 2000) have been reported in the literature, and measurements on pentan-1-ol are now described. Details of a countercurrent water-cooled heat-exchange flow calorimeter for the measurement of enthalpy increments at temperatures up to 700 K and pressures up to 15 MPa have been given (Wormald and Yerlett, 1985). The calorimeter operates by pumping liquid or a liquid mixture from a reservoir at atmospheric pressure through a flash vaporizer at a controlled temperature and pressure and allowing the hot vapor to condense in a water-cooled heat exchanger. The cooling water was supplied at a temperature of about 275 K, and the flow rate was adjusted so that the condensate leaving the calorimeter was at a temperature close to 298.15 K, the standard state temperature. Enthalpy increments were calculated from the flow rate of the cooling water, the flow rate of the fluid, and the temperatures of the incoming and outflowing water and fluid. The calorimeter was tested by making 90 measurements of enthalpy increments for steam over the range of temperature 423.2 K to 623.2 K at pressures up to 10.3 MPa. These agreed with steam tables to within ±0.5%. In addition to measurements on pure alcohols, the calorimeter has been used to measure specific enthalpy increments for hexane (Wormald and Yerlett, 1985) and benzene (Vine and Wormald, 1991) and molar enthalpy increments for the equimolar mixtures (propanone + hexane) (Wormald and Yerlett, 1987; Al-Bizreh et al., 1988), (propanone + benzene) (Wormald and Yerlett, 1992), (methanol + water) (Wormald and Yerlett, 1999), and (ethanol + water) (Wormald and Vine, 2000).

Experimental Section

Enthalpy increments were measured with the countercurrent heat-exchange calorimetric apparatus described previously (Wormald and Yerlett, 1985). The mole fraction purity of the pentan-1-ol was 0.995, and it was used as supplied. The alcohol was pumped at a rate of approximately 0.18 g s^{-1} through the apparatus. It was usually possible to adjust the flow rate of the cooling water so that the temperature of the alcohol leaving the calorimeter differed by little more than 2 or 3 K from the standard state temperature 298.15 K. At the temperatures $T = 598.2$ K and $T = 623.2$ K the fluid leaving the apparatus was analyzed by GLC to check for possible decomposition, but no evidence of this was found. Enthalpy increments measured over the temperature range $T = (448.2 \text{ to } 623.2)$ K at pressures from $p = (0.1 \text{ to } 10.1)$ MPa, and calculated as described below, are listed in Table 1 and plotted against pressure in Figure 1.

Calculation of Enthalpy Increments

At the pressure p of the experiment a stream of pentan-1-ol vapor at a high-temperature T_1 enters the water-cooled heat-exchange calorimeter and emerges as liquid at a temperature T_2, which is close to 298.15 K, and at the same pressure. Energy lost by the pentan-1-ol is gained by the cooling water. Measurement of the water flow rate and temperature rise allows the calculation of the quantity $\Delta h'$ where

$$\Delta h' = h(p,T_1) - h(p,T_2) \tag{1}$$

The desired quantity is the difference Δh between the specific enthalpy of the alcohol at (p, T_1) and the enthalpy at standard state conditions, 298.15 K and the saturation pressure p_s at this temperature.

$$\Delta h = h(p,T_1) - h(p_s,298.15\ \mathrm{K}) \tag{2}$$

Δh can be calculated from $\Delta h'$ by the addition of two terms:

$$\Delta h = \Delta h' + \int_{298\mathrm{K}}^{T_2} c_p(p,T)\,\mathrm{d}T + \int_{p_s}^{p} v(1 - \alpha T)\,\mathrm{d}p \tag{3}$$

where $c_{p,m}$ is the specific heat capacity of the alcohol, v is the specific volume, and α is the isobaric expansivity at 298.15 K calculated from density measurements listed by Wilhoit and Zwolinsky (1973) The cooling water flow rate was adjusted until T_2 was close to 298.15 K, so that the c_p integral was small and the pressure and temperature dependence of c_p could be neglected. Small corrections for heat leaks and for the rate at which kinetic energy enters

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Table 1. Specific Enthalpy Increments $\Delta h/(\mathrm{kJ\cdot kg^{-1}})$ for Pentan-1-ol Measured Relative to the Saturation Pressure p_s of the Liquid at the Standard Temperature 298.15 K

T/K	p/MPa	Δh/kJ·kg^{-1}	p/MPa	Δh/kJ·kg^{-1}	p/MPa	Δh/kJ·kg^{-1}	p/MPa	Δh/kJ·kg^{-1}	p/MPa	Δh/kJ·kg^{-1}
448.2	0.25	895	0.77	437	4.32	440	6.50	438	9.32	436
473.2	0.20	947	0.46	927	1.13	517	4.36	518	6.63	512
	0.35	937	0.64	527	2.94	519	5.61	517		
498.2	0.17	1005	0.47	995	0.74	970	2.10	600	7.15	604
	0.34	1000	0.57	991	1.20	600	2.78	598	10.1	595
523.2	0.15	1072	0.74	1044	1.35	1013	4.07	696	8.18	691
	0.49	1057	1.10	1022	2.33	703	5.38	690	10.1	685
548.2	0.12	1137	0.77	1123	1.88	1070	3.46	798	5.38	786
	0.52	1117	1.29	1103	2.52	800	4.03	790	8.38	785
573.2	0.11	1185	2.72	1112	3.96	905	5.67	885	7.63	872
	0.77	1175	3.50	918	5.12	889	5.98	883	8.31	871
	1.32	1162	3.75	910	5.23	888	6.63	881	9.50	870
586.2	0.11	1221	3.18	1127	4.50	942	5.67	937	8.31	924
	0.78	1210	3.50	1111	5.12	941	5.98	935	9.52	925
	1.33	1197	3.85	970	5.23	940	6.63	933		
	2.72	1152	3.96	957	5.48	938	7.63	924		
598.2	0.15	1259	4.13	1106	4.71	1015	5.96	987	10.1	966
	1.09	1232	4.32	1079	5.25	995	6.50	981		
	2.59	1200	4.51	1048	5.48	992	8.00	968		
	3.84	1135	4.66	1026	5.62	989	9.26	968		
623.2	0.40	1307	2.56	1274	5.35	1116	8.96	1075		
	1.33	1293	4.00	1222	6.63	1086	10.0	1070		

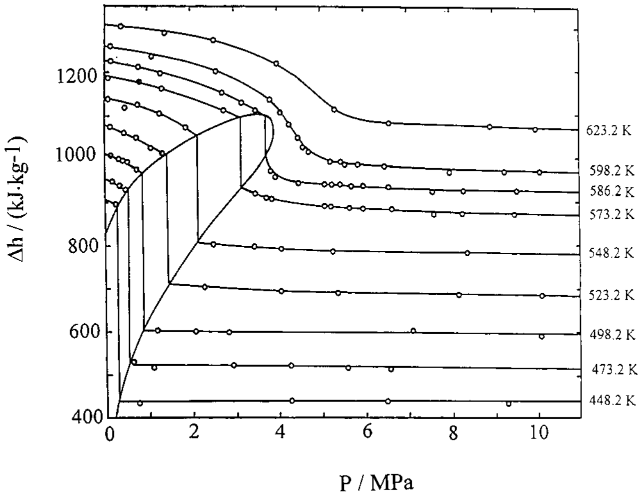

Figure 1. Specific enthalpy increments Δh for pentan-1-ol plotted as a function of the pressure p. ○, Table 1.

the calorimeter when the pressure is low were made, as previously described (Wormald and Yerlett, 1985). Systematic errors on the measurements are estimated to be no greater than ±1 kJ·kg^{-1}. Random errors arose mainly from fluctuations in the operation of the metering pump supplying the alcohol and were estimated to be ±3 kJ·kg^{-1}.

Gaseous Region at Low Pressures

An important test of the accuracy of the measurements is to extrapolate them to zero pressure and to make comparison with ideal gas enthalpies obtained from spectroscopic measurements or gas-phase heat capacities. Figure 2a shows gas-phase enthalpies at pressures below 3 MPa extrapolated to zero pressure by fitting quadratic equations to the measurements. The single point at 448.2 K was extrapolated to zero pressure by drawing through it a line of the same slope as that at 473.2 K. The zero-pressure intercepts are shown in Figure 2b, where they are compared with perfect gas enthalpies obtained by Green (1961), who extrapolated data on the lower alcohols. To make comparison with our measurements which are based on a standard state of $T = 298.15$ K, it was necessary first to adjust these enthalpies to a scale on which the perfect gas enthalpy is zero at 298.15 K and second to add the enthalpy of vaporization of pentan-1-ol at this temperature. Wilhoit and Zwolinsky (1973) list few values; the only calorimetric measurement is that of Wadso (1966), who obtained 646 kJ·kg^{-1} at 298.15 K, and this we have used. With these adjustments the perfect gas enthalpy increments are given by the equation

$$\{h^{\mathrm{pg}}(T) - h^{\mathrm{pg}}(298.15\ \mathrm{K})\}/\mathrm{kJ\cdot kg^{-1}} = 319.4 + 0.6199(T/\mathrm{K}) + 1.5828 \times 10^{-3}\,(T/\mathrm{K})^2 \quad (4)$$

The curve shown in Figure 2b was calculated from eq 4. Agreement between the calculated enthalpy increments and those obtained by extrapolation of the Table 1 measurements to zero pressure is to within ±1%.

Enthalpies of Vaporization

The seven isotherms at temperatures between 448.2 K and 586.2 K start at low pressures in the gas-phase region, intersect the upper part of the two-phase boundary at the dew point pressure, cross the two-phase region, intersect the lower part of the boundary at the bubble point pressure, and continue to higher pressures in the liquid phase. The vertical lines which cross the two-phase region are at the saturation pressure of the alcohol. Saturation pressures were calculated from the equation

$$\ln(p/p_0) = a(1 + bT) + c/T + d\ln T \quad (5)$$

where T is the Kelvin temperature, $a = 378.892$, $b = 1.4181 \times 10^{-4}$, $c = -20535.9$, and $d = -58.323$. With these coefficients eq 5 fits the vapor pressure of pentan-1-ol (Thermodynamic Research Centre, 1999) from standard atmospheric pressure $p° = 0.101\,325$ MPa up to the critical point ($T_c = 588.1$ K, $p_c = 3.909$ MPa) to within ±0.25%, which is adequate for our purposes. The difference between the specific enthalpies of the saturated vapor Δh(g) and the saturated liquid Δh(l) is the specific enthalpy of vaporization $\Delta_{\mathrm{vap}}h$, and all three quantities are listed in Table 2.

Enthalpies of vaporization for pentan-1-ol over the temperature range $T = 431.15$ K to $T = 499.65$ K have

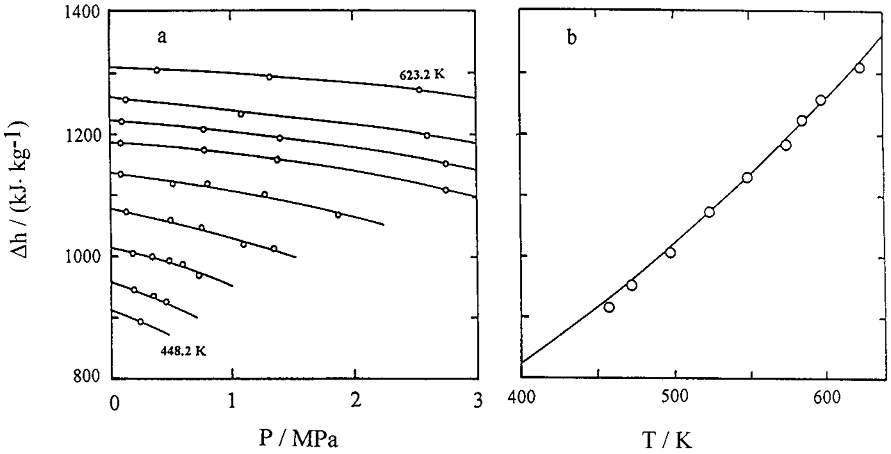

Figure 2. (a) Specific enthalpy increments Δh for pentan-1-ol at low pressures plotted as a function of pressure p, showing the extrapolation to zero pressure: ○, Table 1. (b) Zero-pressure values of the specific enthalpy of pentan-1-ol obtained from Figure 1 shown plotted against temperature. The curve was fitted to ideal gas enthalpies obtained by Green (1961) by extrapolating data on lower alcohols.

Table 2. Specific Enthalpies h(l) and h(g) at the Saturation Temperature T_s and Pressure p_s, Specific Enthalpy of Vaporization $\Delta_{vap}h$, and Uncertainty $\delta\Delta_{vap}h$ on This Quantity, for Pentan-1-ol[a]

T_s K	p_s MPa	h(l) kJ·kg^{-1}	h(g) kJ·kg^{-1}	$\Delta_{vap}h$ kJ·kg^{-1}	$\delta\Delta_{vap}h$ kJ·kg^{-1}
298.15	0.0002			646	5
411.0	0.1013			504	5
448.2	0.299	440	895	455	15
473.2	0.544	520	930	410	10
498.2	0.914	600	960	360	10
523.2	1.442	710	1010	300	10
548.2	2.174	800	1050	250	10
573.2	3.179	930	1090	160	15
586.2	3.811	1010	1090	80	20

[a] The values at 298.15 K and the normal boiling temperature, 411.0 K, were obtained from the literature; all other values were obtained from the Table 1 measurements.

been measured by Radosz and Lydersen (1980), who fitted their measurements to an equation with four disposable parameters a_1 to a_4. The first term was $a_1(1 - T_r)^{1/3}$, and higher terms of similar form had the exponents 2/3, 5/3, and 6/3. Their measurements cannot be fitted with adequate accuracy by the Watson equation (Watson, 1943). Vine and Wormald (1989) found that the Watson equation was a poor fit to the enthalpies of vaporization of ethanol, and they suggested the equation

$$\Delta_{vap}h(T) = \Delta_{vap}h(T_b)\{ax + (1 - a)x^m\}^n \qquad (6)$$

where

$$x = (1 - T/T_c)(1 - T_b/T_c)^{-1} \qquad (7)$$

Here T_b is the normal boiling temperature and a, n, and m are disposable parameters. When $a = 1$, the equation reduces to that of Watson. The equation was tested by fitting 30 values of the enthalpy of vaporization of water from T_b to T_c obtained from the steam tables of Haar et al. (1984). The best fit parameters for water are $a = 1.1286$, $m = 2.9508$, and $n = 0.3967$. These fit the data with a standard deviation of ± 4 kJ·kg^{-1} whereas the standard deviation obtained with the Watson equation was ± 26 kJ·kg^{-1}.

To obtain an equation which best fits the specific enthalpy of vaporization of pentan-1-ol, we used $\Delta_{vap}h$(298.15 K) = 646 kJ·kg^{-1}, $\Delta_{vap}h(T_b)$ = 504 kJ·kg^{-1}, the specific enthalpies of vaporization obtained by Radosz and Lydersen (1980), and the values of $\Delta_{vap}h(T)$ in the temperature range 448.2 K to 548.2 K listed in Table 2. The parameter m in eq 6 was set at $m = 3$. The best fit parameters are $\Delta_{vap}h(T_b) = 500.4$ kJ·kg^{-1}, $a = 0.9631$, and $n = 0.4637$, and the standard deviation of the fit was $\sigma = 5$ kJ·kg^{-1}. The $\Delta_{vap}h(T)$ curve for pentan-1-ol is shown plotted against reduced temperature in Figure 3. It is so close to the curve fitted by Radosz and Lydersen (1980) to their own data that it is difficult to distinguish one from the other. Figure 3 shows the enthalpies of evaporation obtained from our measurements to be in good agreement with other work.

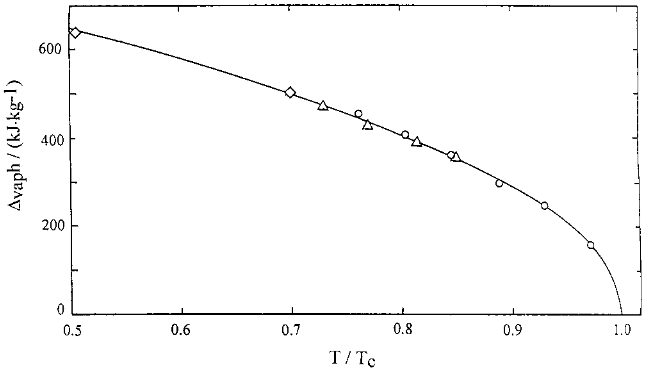

Figure 3. Specific enthalpies of vaporization $\Delta_{vap}h$ of pentan-1-ol plotted as a function of the reduced temperature T/T_c: ○, Table 2, derived from the Table 1 measurements; △, Radsoz and Lydersen (1980); ◇, Wadso (1966).

Constructing the Two-Phase Boundary

To construct the two-phase boundary on the enthalpy pressure diagram shown in Figure 1, it is convenient first of all to fit the specific enthalpies of vaporization to an equation in which the argument is a function of reduced pressure p_r rather than reduced temperature T_r. Equation 6 is again of suitable form, and it was soon established that the parameter $m = 1.5$ was a good choice.

$$\Delta_{vap}h(p) = \Delta_{vap}h(p°)(ay + (1 - a)y^{1.5})^n \qquad (8)$$

where y is given by

$$y = \{\ln(p/P_c)\}/\{\ln(p°/P_c)\} \qquad (9)$$

The specific enthalpy of vaporization $\Delta_{vap}h(p°)$ at the

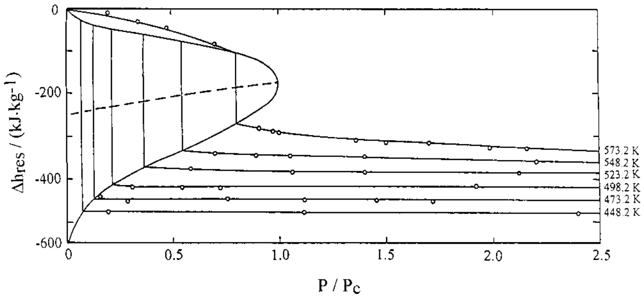

Figure 4. Residual specific enthalpy increments Δh_{res} for pentan-1-ol plotted against reduced pressure p_r. The length of the vertical lines is a measure of the specific enthalpy of vaporization. The broken curve is the locus of the midpoints of the vertical lines and was calculated from eq 12 as described in the text. The curve intersects the two-phase boundary at the critical point residual enthalpy $h_c = -175$ kJ·kg^{-1}. ○, calculated from Table 1 measurements.

standard pressure $p° = 0.101\,325$ MPa is of course the same quantity as $\Delta_{vap}h(T_b)$. As before $\Delta_{vap}h(p°)$ was treated as an adjustable parameter. Least squares optimization gave $\Delta_{vap}h(p°) = 500.7$ kJ·kg^{-1}, $a = 1.446$, and $n = 0.4324$. With these parameters eq 8 fits the vaporization enthalpies listed in Table 2 with a standard deviation of $\sigma = 5$ kJ·kg^{-1}, which is as good as the fit obtained using eq 6 and is adequate for our purposes. Again the curve is almost indistinguishable from that shown in Figure 3.

To construct the two-phase envelope, we first simplified Figure 1 by subtracting the ideal gas enthalpies calculated from eq 4 from the measurements listed in Table 1 and so obtained residual enthalpy increments

$$\Delta h^{res} = \Delta h(p) - \Delta h^{pg} \tag{10}$$

Residual enthalpy increments were then plotted against reduced pressure p_r, as shown in Figure 4. The use of reduced pressure facilitates comparison with similar diagrams for other fluids. The two-phase envelope is now more symmetric than before. The midpoint of each of the six vertical sections, which are the enthalpies of vaporization, was marked, and a curve through these midpoints was drawn and extrapolated to $p/P_c = 1$. In this way, a value of the residual enthalpy h_c at the critical point was obtained and found to be $h_c = (-175 \pm 10)$ kJ·kg^{-1}. At the pressure $p° = 0.101\,325$ MPa, which is a reduced pressure of $p_r = 0.026$, the enthalpy of vaporization calculated from eq 8 with the parameters given above is (504 ± 5) kJ·kg^{-1}. At this pressure the residual enthalpy of the liquid at the bubble point pressure is the sum of two terms, the residual enthalpy of the gas and the specific enthalpy of vaporization of the liquid.

$$\Delta h^{res} = \int_0^{p°} (\partial h/\partial p)_T \, dp + \Delta_{vap}h \tag{11}$$

Here $(\partial h/\partial p)_T$ can be calculated from the isothermal Joule–Thomson coefficient $\phi = B - T(dB/dT)$ of the vapor at the normal boiling temperature 411.1 K divided by the molar mass. The integration is carried out from $p = 0$ to $p = p°$. The second virial coefficients B for pentan-1-ol have been obtained from measurements of the heat of mixing of $(0.5N_2 + 0.5C_5H_{11}OH)$ and will be reported shortly (Wormald and Lacey, 2000). The value of ϕ derived from the measurements was $-(6500 \pm 1000)$ cm^3 mol^{-1}, and the value of the integral in eq 11 was found to be (8 ± 2) kJ·kg^{-1}. The specific enthalpy of vaporization at 411.1 K is (504 ± 5) kJ·kg^{-1}. The midpoint at 252 kJ·kg^{-1} of a vertical line drawn on the figure corresponding to an enthalpy of vaporization of 504 kJ·kg^{-1} fixes the low-pressure limit of the midpoint curve. As required by eq 11, this midpoint is plotted in Figure 4 at (252 ± 8) kJ·kg^{-1} and at $p°/P_c = 0.026$, which is almost zero reduced pressure.

An analysis of similar measurements on propan-1-ol (Vine and Wormald, 1999), propan-2-ol (Wormald and Vine, 1999), and butan-1-ol (Wormald and Fennell, 2000) showed that the locus of the midpoints of the two-phase region residual enthalpies for each of these fluids can be fitted to within experimental error by an equation of the form

$$h_{mid} = \{0.5\Delta_{vap}h(p°) - h_c\}(1 - p_r)^z \tag{12}$$

where p_r is the reduced pressure, h_c is the specific residual enthalpy at the critical point, and

$$z = 1 + s\{\ln(p_r^{\ -1})\} \tag{13}$$

When the adjustable parameter $s = 0$, the equation generates a linear midpoint line similar to the rectilinear diameter law for coexisting liquid and gas densities. The midpoints of the two-phase region liquid and gas residual enthalpies for pentan-1-ol can be fitted by a linear function only down to $p_r = 0.4$; to fit the midpoints at lower reduced pressures requires a function which is not far from linear at $p_r > 0.4$ and which changes more quickly as p_r approaches zero. The logarithmic functionality given by eq 13 fulfills this requirement. The best value of the parameter s was found to be 0.5, the same as that for propan-1-ol and butan-1-ol. Finally at each value of p_r the midpoint residual enthalpy was calculated from eqs 12 and 13, the dew point enthalpy h(gas) was obtained by subtracting half of the enthalpy of vaporization calculated from eq 7, and the bubble point enthalpy h(liq) was calculated by adding the same amount.

To calculate the two-phase envelope shown in Figure 1, it is only necessary to add perfect gas enthalpies calculated from eq 4. As the ordinate in Figure 1 is pressure rather than temperature, it is convenient to generate an equation which expresses the saturation temperature T_s as a function of the pressure. The saturation temperature of pentan-1-ol, obtained by fitting vapor pressure measurements as a function of $\ln(p/p°)$, where $p°$ is standard atmospheric pressure, is given by the equation

$$T_s/K = 410.4 + 32.30\{\ln(p/p°)\} + 2.3411\{\ln(p/p°)\}^{2.5} \tag{14}$$

The alternative procedure is to iteratively solve eq 5 to yield values of T_s at each chosen pressure, but it is less convenient. Ideal gas enthalpies can now be calculated at temperatures T_s, and the transformation of the two-phase envelope shown in Figure 4 back to that shown in Figure 1 is straightforward. The values of the midpoint curve h_{mid}(1) shown in Figure 1 were calculated from the values of h_{mid}(4) shown in Figure 4 from the equation

$$h_{mid}(1) = h^{pg} - h_c - h_{mid}(4) \tag{15}$$

where h^{pg} was calculated from eq 4 and h_{mid}(4) was calculated from eq 12. Addition or subtraction of half of the enthalpy of vaporization calculated from eq 8 generates the two-phase envelope shown in Figure 1.

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