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ANALYSIS OF VAPOUR PRESSURES OF NITROGEN-CONTAINING COMPOUNDS USING AN EQUATION OF STATE

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ABSTRACT

Vapour pressures of organic nitrogen-containing compounds have been analysed using an equation of state allowing for association which was developed by Wenzel, Moorwood and Baumgartner, (Fluid Phase Equilibria, 9 (1982) 225). Besides vapour pressures, the equation accurately reproduces liquid and vapour densities at room temperature and critical coordinates of polar compounds. The equation is also able to predict vapour pressures in a wide temperature range from limited experimental input information.

INTRODUCTION

The knowledge of vapour pressures is essential for the investigation of the thermodynamic properties of pure components and for the modelling of separation processes. Therefore, accurate methods are needed for the correlation of vapour pressures in wide temperature ranges. The equation-of-state methods, when compared with correlation techniques (e.g. the so-called $1/T$ equations [1], the equations of Antoine, Abrams–Massaldi–Prausnitz, Wagner, Cox, Cragoe and Harlacher–Braun [2]) have the advantage of the possibility of correlating simultaneously properties other than vapour pressures, e.g. vapour and liquid densities and critical properties. Moreover, the equation-of-state parameters can be used directly for the computation of the properties of mixtures, and various thermodynamic functions (such as enthalpy, entropy and Gibbs energy of both phases) can be simultaneously evaluated.

Wenzel et al. [3] proposed an equation-of-state method particularly suitable for polar and associating compounds. This method makes it possible to correlate simultaneously vapour pressures, liquid and vapour densities and

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critical properties of pure polar compounds. Then the properties of mixtures can be calculated with only one binary parameter.

The purpose of the present paper is to perform a comprehensive analysis of the available vapour pressure data for nitrogen-containing compounds [4], i.e. pyridine bases, amines, nitrocompounds and nitriles. Besides the correlation of experimental data, the capability of the equation of state to predict vapour pressures beyond the temperature range of correlated data will be studied.

METHOD

The procedure has been explained in detail by Wenzel et al. [3]. Here, we summarise the main concepts underlying the method. The equation of state used has a van der Waals-type form [5]

$$P = RT/(V-b) - a(T)/\left(V^2 + (1+3\omega)bV - 3\omega b^2\right) \tag{1}$$

where R is the gas constant, T is the temperature, P is the pressure, V is the molar volume, ω is Pitzer's acentric factor, and $a(T)$ and b are parameters representing attractive and repulsive forces between molecules, respectively.

For a given substance, b is a given constant and the parameter a depends on temperature following a correlation that is Soave's well-known expression [2]. These parameters are determined from the critical data, T_c and P_c, and the acentric factor ω of the pure substance. In this model, a pure substance is thus characterised by three parameters, T_c, P_c and ω.

A pure polar substance is treated as a mixture of monomers and several associated species. The monomers and associated species are characterised by their hypothetical values of T_c, P_c and ω. Furthermore, each association reaction, $i\mathrm{A}_1 = \mathrm{A}_i$, is characterised by the enthalpy, δH, and entropy, δS, of forming an associated species. For a mixture of monomeric and associated species the following mixing rules are used

$$b = \sum x_i b_i \tag{2}$$

$$a = \sum\sum x_i x_j (1-\theta_{ij})(a_i a_j)^{0.5} \tag{3}$$

$$\omega = \sum x_i \omega_i z_i \tag{4}$$

with

$$z_i = b_i^{0.7} / \sum \left(x_i b_i^{0.7}\right) \tag{5}$$

where x_i is the mole fraction of component i in either the liquid or vapour phase and θ is the binary parameter. Then, chemical equilibria between the associated species and physical (phase) equilibria between all species present in the hypothetical mixture are calculated simultaneously. The stoichiometry of associates, i.e. the association model, is determined by trial-and-error to obtain the best fit of experimental data.

As input data the method requires the following pure component data: (1) vapour pressure versus temperature; (2) liquid and vapour density (at least at one, near ambient temperature); (3) critical coordinates, i.e. T_c, P_c; and (4) for mixtures, limited data relating to the binary phase equilibrium.

RESULTS

Table 1 lists the parameters obtained for the equation of state. In some cases it was found that no association equilibrium has to be assumed and the data are accurately reproduced when the compound is treated as monomeric. This is the case for all pyridine bases, and for the majority of amines and higher nitriles. In most other cases it was sufficient to assume only one associate besides the monomer, i.e. the dimer or trimer. This indicates that the compounds studied are relatively weakly associated. In the case of methanamine it is possible to represent the data with similar accuracy using either the monomer–dimer–tetramer model or only the monomer.

Table 2 shows the quality of reproduction of the experimental vapour pressures, liquid densities and critical data which were used as input data for the fitting procedure. For the majority of compounds, the deviations obtained by correlating vapour pressures are low. Only in a few cases, i.e. for ethylpyridines and some of the nitriles and nitroaromatics, are considerable deviations noted. For these compounds, however, the experimental accuracy, as claimed by the authors, is relatively low [4,14,23]. The quality of reproduction of the liquid densities is, in general, satisfactory. Only in some cases do the deviations exceed 4%. Experimental critical data are available only for about half of the compounds studied. For the remaining compounds T_c and P_c were estimated using group contribution methods [19] and are denoted by asterisks in Table 2. If the selected group contribution method has been recommended by Simmrock et al. [13] the values in Table 2 are denoted by plus signs. In general, the deviations of calculated critical coordinates from experimental data are small and within experimental error. Only in the case of some compounds for which T_c and P_c were estimated from group contribution methods are the deviations higher.

To verify the predictive capability of the method, vapour pressures in the high pressure range were predicted using parameters determined in the low pressure range as given in Table 1. The high-pressure data are only known for three nitrogen-containing compounds [4,38]. The results are listed in Table 3. The deviations of the predicted values from experimental data are of the order of 1% and are considered to be very satisfactory.

TABLE 1

Parameters of the equation of state of Wenzel et al. [3]

Compound	Model	Multimer	T_c (K)	P_c (bar)	Ω	$-\delta S$ (J $\mathrm{mol}^{-1}\,\mathrm{deg}^{-1}$)	$-\delta H$ (kJ mol^{-1})
I. Pyridine bases							
Pyridine	monomer	1	617.07	55.92	0.2573		
2-Methylpyridine	monomer	1	620.32	45.54	0.3009		
3-Methylpyridine	monomer	1	644.53	46.26	0.2972		
4-Methylpyridine	monomer	1	647.57	46.76	0.2940		
2,3-Dimethylpyridine	monomer	1	656.35	40.80	0.3391		
2,4-Dimethylpyridine	monomer	1	651.14	39.75	0.3367		
2,5-Dimethylpyridine	monomer	1	647.87	39.18	0.3386		
2,6-Dimethylpyridine	monomer	1	625.41	38.60	0.3526		
3,4-Dimethylpyridine	monomer	1	684.54	41.12	0.3357		
3,5-Dimethylpyridine	monomer	1	672.30	40.40	0.3374		
2,4,6-Trimethylpyridine	monomer	1	638.29	30.74	0.4473		
2-Ethylpyridine	monomer	1	633.92	39.78	0.3507		
3-Ethylpyridine	monomer	1	671.67	40.61	0.2953		
4-Ethylpyridine	monomer	1	661.38	40.73	0.3522		
II. Amines							
Methylamine	monomer	1	429.92	76.77	0.3016		
Methylamine	monomer	1	426.33	80.97	0.2642		
	dimer	2	496.45	47.92	0.3100	115.51	21.63
	tetramer	4	514.63	19.31	0.4988	308.89	69.48
Propanamine	monomer	1	489.95	46.57	0.3384		
2-Propanamine	monomer	1	464.95	44.08	0.3310		
Butanamine	monomer	1	524.99	40.39	0.3698		

Isobutanamine	monomer	1	510.14	38.84	0.3596		
Dimethylamine	monomer	1	428.54	51.92	0.3726		
Diethylamine	monomer	1	491.53	36.72	0.3450		
Triethylamine	monomer	1	532.57	29.53	0.3294		
Cyclohexanamine	monomer	1	617.25	40.40	0.3208		
Benzenamine	monomer	1	681.26	48.98	0.3818		
	dimer	2	908.95	34.27	0.5158	90.27	15.31
III. Nitro compounds							
Nitromethane	monomer	1	523.95	45.76	0.2040		
	trimer	3	782.77	33.49	0.4618	167.61	43.45
Nitroethane	monomer	1	579.62	49.09	0.2888		
	dimer	2	663.03	34.24	0.5482	85.16	21.13
Nitropropane	monomer	1	576.86	34.49	0.2578		
	dimer	2	835.40	34.42	0.3445	92.72	19.86
2-Nitropropane	monomer	1	580.58	37.50	0.2353		
	dimer	2	787.05	33.13	0.3843	94.45	19.64
Nitrobutane	monomer	1	614.83	38.59	0.4346		
	dimer	2	728.99	25.35	0.5284	84.59	20.17
2-Nitrobutane	monomer	1	609.22	39.76	0.3699		
	dimer	2	717.54	26.35	0.4458	87.94	20.70
Nitrobenzene	monomer	1	686.49	33.26	0.3616		
	dimer	2	802.89	27.76	0.6247	81.93	25.41
1-Methyl-2-nitrobenzene	monomer	1	719.38	33.67	0.4066		
	dimer	2	872.14	26.77	0.7522	98.74	19.74
1-Methyl-4-nitrobenzene	monomer	1	734.08	30.49	0.3912		
	dimer	2	914.13	26.48	0.7634	103.23	19.67

TABLE 1 (continued)

Compound	Model	Multimer	T_c (K)	P_c (bar)	Ω	$-\delta S$ (J mol^{-1} deg^{-1})	$-\delta H$ (kJ mol^{-1})
IV. Nitriles							
Acetonitrile	monomer	1	515.20	42.13	0.3158		
	dimer		655.63	60.98	0.3415	77.72	19.94
Propanenitrile	monomer	1	549.85	41.54	0.3297		
	dimer	2	661.21	21.40	0.4557	93.08	19.96
Butanenitrile	monomer	1	541.60	39.24	0.3313		
	dimer	2	638.25	24.17	0.4629	83.60	25.61
Pentanenitrile	monomer	1	638.84	42.54	0.2798		
Hexanenitrile	monomer	1	644.65	33.08	0.3605		
Octanenitrile	monomer	1	696.43	31.36	0.4051		
Decanenitrile	monomer	1	716.88	24.21	0.5171		
Trimethylacetonitrile	monomer	1	574.82	37.69	0.2897		
Trichloroacetonitrile	monomer	1	553.04	42.51	0.2773		
Methacrylonitrile	monomer	1	578.65	53.20	0.2351		
IV. Miscellaneous							
Methylhydrazine	monomer	1	569.78	79.03	0.3763		
Pyrrole	monomer	1	640.01	69.57	0.3232		
1-Methylpyrrole	monomer	1	594.82	48.23	0.3188		
Pyrrolidine	monomer	1	566.15	55.55	0.2886		
Piperidine	monomer	1	592.03	45.00	0.2207		
	dimer	2	835.39	29.50	0.3977	119.67	17.39

TABLE 2

Quality of reproduction of experimental vapour pressure, liquid density and critical parameters

Compound	Vapour pressure			Liquid density			Critical temperature			Critical pressure		
	AAD (P^{sat}) (%)	Experimental temperature range (K)	Ref.	AAD (V^l) (%)	Experimental temperature (K)	Ref.	T_c (K)	δT_c (%)	Ref.	P_c (bar)	δP_c (%)	Ref.
I. Pyridine bases												
Pyridine	0.03	320–389	6	3.11	293.15	7	620.0	0.47	7,8,9	54.9	−0.58	7,8,9
2-Methylpyridine	0.04	338–403	6	1.87	293.15	7	621.0	0.11	7,8,9	44.8	−0.32	7,8
3-Methylpyridine	0.06	354–418	6	4.87	293.15	7	645.0	0.07	7,8,9	45.3	−0.81	7
4-Methylpyridine	0.08	350–419	6	4.10	293.15	7	646.0	−0.24	8,9	45.4	−1.65	8
2,3-Dimethylpyridine	0.05	373–436	10	1.95	293.15	11	655.4	−0.14	7,8,9	40.0	−0.59	7
				1.81	298.15	11						
2,4-Dimethylpyridine	0.12	349–433	10	2.39	293.15	11	647.4	−0.58	7,8,9	38.5	−1.85	8
				2.24	298.15	11						
2,5-Dimethylpyridine	0.10	358–430	6	3.08	293.15	11	644.2	−0.50	7,8,9	37.7	−2.38	8
				2.93	298.15	11						
2,6-Dimethylpyridine	0.05	352–417	6	0.47	293.15	11	623.8	−0.26	7,8,9	38.0	−0.29	8
				0.34	298.15	11						
3,4-Dimethylpyridine	0.09	397–454	10	4.16	293.15	11	683.8	−0.38	7,8,9	40.2	−3.28	7
				4.00	298.15	11						
3,5-Dimethylpyridine	0.07	372–446	10	4.62	293.15	11	667.2	−0.77	7,9	38.5	−3.64	7
				4.47	298.15	11						
2,4,6-Trimethylpyridine	0.75	381–444	12	7.36	293.15	7	645.0	1.04	7,13	31.2	2.67	7,13
2-Ethylpyridine	0.54	324–372	14	0.36	293.15	7	634.0	0.01	7,13	39.9	1.69	7
3-Ethylpyridine	0.52	335–373	14	7.17	293.15	11	661.0	−1.61	7,13	39.0	−2.87	7
4-Ethylpyridine	1.22	334–372	14	1.70	293.15	7	663.0	0.24	7,13	39.9	−0.67	7,13
II. Amines												
Methylamine	0.36	190–267	15	1.07	293.15	16	430.7	0.09	8	74.2	−1.79	8
Methylamine	0.62	190–267	15	0.62	293.15	16	430.7	0.18	8	74.2	−2.17	8
				0.78	298.15	16						

TABLE 2 (continued)

Compound	Vapour pressure			Liquid density			Critical temperature			Critical pressure		
	AAD (P^{sat}) (%)	Experimental temperature range (K)	Ref.	AAD (V^l) (%)	Experimental temperature (K)	Ref.	T_c (K)	δT_c (%)	Ref.	P_c (bar)	δP_c (%)	Ref.
Propanamine	0.15	296–351	17	0.84	293.15	16	497.0	1.42	8,9	46.2	0.49	8,9
				0.90	298.15	16						
2-Propanamine	0.76	277–329	17	2.27	293.15	16	471.8	1.45	8,13	44.2	1.60	8,13
				2.26	298.15	16						
Butanamine	0.17	297–350	18	1.79	293.15	16	524.0	−0.19	8,9,13	40.5	0.18	8,9,13
				1.87	298.15	16						
Isobutanamine	0.04	293–341	18	0.01	293.15	16	500.0 *	−2.03	19	40.0 *	2.91	19
				0.07	298.15	16						
Dimethylamine	0.29	214–280	20	2.24	293.15	16	437.2	1.99	8	52.0	1.48	8
				2.14	298.15	16						
Diethylamine	0.14	308–334	21	0.08	293.15	16	496.6	1.02	8	36.1	−0.32	8
				0.03	298.15	16						
Triethylamine	0.30	333–368	21	1.57	293.15	16	535.6	0.57	8	29.5	1.29	8
				1.85	298.15	16						
Cyclohexanamine	0.41	334–402	22	2.18	293.15	22	600.0 *	−2.88	19	45.0 *	10.23	19
Benzenamine	0.67	309–458	23,24	0.27	293.15	25	699.0	0.08	7,8,9	51.7	0.50	7,8,9
			26	0.38	298.15	25						
III. Nitro compounds												
Nitromethane	0.06	329–410	27	0.31	293.15	16	588.0 $^{+}$	0.09	13	61.5 $^{+}$	1.55	13
				0.15	298.15	16						
Nitroethane	0.65	298–383	28	1.56	293.15	16	595.0 $^{+}$	−0.39	13	47.2 $^{+}$	−2.12	13
				1.63	298.15	16						
Nitropropane	0.32	332–405	23	0.06	293.15	16	606.0 $^{+}$	−0.10	13	39.0 $^{+}$	−0.16	13
				0.07	298.15	16						
2-Nitropropane	0.60	283–383	28	0.15	293.15	16	600.0 $^{+}$	0.12	13	40.4 $^{+}$	1.54	13
				0.01	298.15	16						
Nitrobutane	0.61	283–413	28	1.61	293.15	16	624.0 $^{+}$	−1.34	13	37.0 $^{+}$	−4.34	13
				1.30	298.15	16						
2-Nitrobutane	0.40	283–403	28	0.38	293.15	16	615.0 $^{+}$	−1.16	13	35.1 $^{+}$	−12.56	13
				0.11	298.15	16						

Nitrobenzene	0.10	407–484	29	0.11	298.15	29	712.0 +	0.09	13	42.9 *	20.71	19
1-Methyl-2-nitrobenzene	1.08	402–495	23	2.01	293.15	23	720.0 +	−0.36	13	33.2 +	−1.13	13
1-Methyl-4-nitrobenzene	0.19	420–506	23	1.63	298.15	23	735.0 +	−0.15	13	29.2 +	−3.67	13
IV. Nitriles												
Acetonitrile	0.04	315–355	30	0.01	293.15	16	547.9	−0.19	8,13	47.1	−2.97	8,9
				0.18	298.15	16						
Propanenitrile	0.88	309–371	23	3.30	298.15	16	564.4	0.07	8,9	40.8	0.09	8,9
Butanenitrile	0.25	333–401	31	6.12	293.15	16	582.2	0.06	8,9,13	36.9	0.62	8,9,13
				5.94	298.15	16						
Pentanenitrile	0.91	342–414	23	5.27	293.15	16	600.0 *	−3.64	19	30.0 *	−3.04	19
				5.07	298.15	16						
Hexanenitrile	0.52	371–442	31	6.85	293.15	16	622.0	−6.47	8,13	31.7	−41.78	8,13
				6.65	298.15	16						
Octanenitrile	0.39	374–461	30	0.21	293.15	16	600.0 *	−16.07	19	30.0 *	−4.54	19
				0.41	298.15	16						
Decanenitrile	0.93	381–517	30	4.48	293.15	16	700.0 *	−2.41	19	20.0 *	−21.04	19
				4.25	298.15	16						
Trimethyl-acetonitrile	0.12	313–370	32	0.10	293.15	16	570.0 *	−0.85	19	45.0 *	16.24	19
				0.12	298.15	16						
Trichloro-acetonitrile	0.44	293–357	33	0.07	293.15	33	550.0 *	−0.55	19	50.0 *	14.98	19
				0.15	298.15	33						
Methacrylonitrile	0.07	273–373	34	0.26	293.15	34	500.0 *	−15.73	19	40.0 *	−33.00	19
				0.13	303.15	34						
V. Miscellaneous												
Methylhydrazine	0.22	275–298	35	0.07	298.15	11	567.1 +	−0.47	13	78.0 +	0.03	13
Pyrrole	0.15	339–439	36	3.47	293.15	7	639.8	−0.03	7,8,9	70.6	2.77	7
				3.56	298.15	7						
1-Methylpyrrole	0.06	322–423	17	2.68	293.15	37	596.0 +	0.20	13	47.3 +	−0.69	13
Pyrrolidine	0.31	316–394	17	7.41	293.15	7	568.6	0.43	8,9	54.7	−0.27	8,9
Piperidine	0.28	316–417	17	0.06	293.15	37	594.0 +	−0.01	13	44.5 +	−0.70	13

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* Estimated using group contribution methods.

\+ Estimated using the selected group contribution method, as recommended by Simmrock et al. [13].

TABLE 3

Prediction of vapour pressures in the high-pressure region using parameters determined from the low and moderate pressure region

Compound	Temperature range (K)	Pressure range (bar)	AAD (P^{sat}) (%)	Ref.
Pyrrole	450–544	3.6–20.3	0.72	32
Pyrrolidine	422–566	5.3–54.6	1.12	32
Pyridine	450–616	4.6–54.3	0.99	32

CONCLUSIONS

The present results show that the equation of state used is an accurate tool for simultaneously correlating pure component vapour pressures, densities and critical properties of nitrogen-containing compounds. As well as yielding high quality correlated experimental data, the equation is able to predict pressures in temperature ranges for which experimental data are missing. The models found may not be expected to be suitable for the calculation of vapour–liquid equilibria in mixtures as data for mixtures were not considered.

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