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Simple correlations for saturated liquid and vapor densities of pure fluids

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Abstract

Two correlations for the saturated liquid and vapor densities of pure substances particularly those used as working fluids in refrigeration machines are proposed. They are shown to represent well the reduced density as a function of the reduced temperature ranging from 0.5 to 1. For about 30 pure substances with acentric factor and critical compressibility factor varying in a wide range, the correlations predict accurately the saturation densities.

Keywords: Saturated vapor density; Saturated liquid density; Pure fluids; Corresponding state correlation

1. Introduction

The knowledge of the physical properties of fluids is essential to the equipment design of the refrigeration machines. In particular, the quantitative description of vapor–liquid equilibrium (VLE) is required in such applications.

Equations of state (EOS) are widely used for VLE calculations of pure and mixtures of non-polar and slightly polar substances. Such equations inherently provide good information but, at present, their usefulness to polar substances is limited [1,2]. Many successful equations for densities at saturation of pure fluids had been developed following the general form [3,4]:

$$\rho_{\mathrm{r}} = \sum_{i=1}^{p} a_i T^i \tag{1}$$

$$\vec{a} = \{a_1, a_2, \ldots, a_{\mathrm{p}}\}$$

where $\vec{a} = \{a_i\}$ is a vector of adjustable parameters and i are real powers.

Although the general correlation (1) predicts with good accuracy the saturated densities data, it is a multiparameter model with power's values depending on the chosen compound that cannot be generalized to all considered fluids to obtain a simple and a common correlation.

The proposed research aims a principal objective: the elaboration with the help of the corresponding state principle of simple correlations for the saturated vapor and liquid densities of a set of pure fluids, especially of those used—or proposed for use—as working fluids in absorption refrigeration machines like water, ammonia, carbon dioxide, hydrocarbons, etc. Because the models will be integrated in a simulation software for refrigeration machines, the correlations do not need to describe with equal good accuracy the entire temperature range of saturated liquid and vapor densities, but would provide acceptable results in the working range of the previous machines.

About 30 fluids with an acentric factor, ω, and a critical compressibility factor, Z_{c}, ranging, respectively, from −0.2 to 0.35 and 0.23 to 0.3 are investigated. Their main characteristics are given in Table 1. The proposed models for the reduced saturated vapor and liquid densities are expressed in reduced variables: $T_{\mathrm{r}} = T/T_{\mathrm{c}}$ and $\rho_{\mathrm{r}} = \rho/\rho_{\mathrm{c}}$, where T_{c} and ρ_{c} are the critical data.

Adjustable parameters of the new models are obtained with the help of a nonlinear regression procedure [5] by minimizing as objective function the sum of the quadratic

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Nomenclature

P	pressure
R	universal gas constant
T	thermodynamic temperature
Z	compressibility factor

Subscripts

c	critical
r	reduced
f	liquid phase
v	vapor phase

Greek letters

ρ	saturated molar density
ω	acentric factor

average deviation between the density data [6] and its calculated values:

$$\Phi = \sum_{i=1}^{N}\left(1-\frac{\rho_{\mathrm{r,calculated}}}{\rho_{\mathrm{r}}}\right)_i^2 \quad (2)$$

The predictive quality of the proposed correlations is evaluated by means of the root standard deviations RMSD and RMSDr:

$$\mathrm{RMSD} = \left(\sum_{i}^{N}\frac{(\rho_{\mathrm{r}}-\rho_{\mathrm{r,calculated}})_i^2}{N}\right)^{1/2} \quad (3)$$

$$\mathrm{RMSDr\%} = 100\left(\frac{\sum_i^N (1-\rho_{\mathrm{r,calculated}}/\rho_{\mathrm{r}})_i^2}{N}\right)^{1/2} \quad (4)$$

with N being the number of used data.

2. Correlations

Pressure–volume–temperature (*PVT*) equation of state of pure gases and liquids are usually used to describe the volumetric behavior of the pure substances. Two-parameter cubic equations of state (CEOS) had been very successful for non-polar and slightly polar components. Their common shortcomings are inaccuracy of the predicted liquid density [7] and a unique value of the critical compressibility factor Z_c [2]. The virial equation can represent only

Table 1
Characteristics of the considered fluids [6]: critical data, acentric factor, normal boiling point temperature, temperature ranges of validity, T_{min}, T_{max} and number of used data

Fluid	T_c (K)	P_c (bar)	ρ_c (mol/l)	ω	T_{nbp} (K)	$T_{min}-T_{max}$ (K)	N
H_2	33.19	13.15	14.94	−0.2140	20.39	16.00–T_c	18
D_2	38.34	16.65	17.33	−0.1750	23.31	19.00–T_c	20
Ne	44.49	26.79	23.88	−0.0387	27.10	25.00–T_c	21
Ar	150.69	48.65	13.41	−0.00219	87.30	84.00–T_c	68
Kr	209.48	55.10	10.84	−0.0017	119.78	116.00–T_c	95
Xe	289.73	58.40	8.37	0.0036	165.03	162.00−T_c	28
CH_4	190.56	4.60	10.14	0.0114	111.68	92.00–T_c	50
O_2	154.58	50.43	13.63	0.0222	90.19	75.00–T_c	81
N_2	126.25	33.98	11.18	0.0372	77.36	63.15–T_c	23
F_2	144.41	51.72	15.60	0.0449	85.04	72.00–T_c	38
CO	132.80	34.93	10.85	0.0510	81.63	69.00–T_c	33
C_2H_4	282.35	50.42	7.64	0.0866	169.38	142.00–T_c	72
C_2H_6	305.33	48.72	6.87	0.0993	184.55	152.00–T_c	77
NF_3	234.00	44.61	7.92	0.1260	144.14	117.00–T_c	31
C_3H_6	365.57	46.65	5.31	0.1408	225.46	184.00–T_c	47
C_3H_8	369.85	42.48	4.96	0.1524	231.06	186.00–T_c	47
i-C_4H_{10}	407.85	36.40	3.86	0.1850	261.48	200.00–T_c	43
n-C_4H_{10}	425.16	37.96	3.92	0.2000	272.60	210.00–T_c	44
R22	369.29	49.90	6.06	0.2208	232.34	185.00–T_c	37
CO_2	304.21	73.84	10.62	0.2239	194.75	220.00–T_c	23
C_5H_{12}	469.70	33.66	3.22	0.2510	309.21	235.00–T_c	48
NH_3	405.40	113.33	13.21	0.2560	239.82	200.00–T_c	42
R143a	345.86	37.61	5.13	0.2615	225.91	172.00–T_c	36
R152a	386.41	45.17	5.57	0.2752	249.127	191.00–T_c	40
R32	351.26	57.82	8.15	0.2769	221.50	172.00–T_c	37
R123	456.83	36.62	3.60	0.2819	300.98	228.00–T_c	47
R124	395.42	36.24	4.10	0.2881	261.19	195.00–T_c	42
C_6H_{14}	504.82	30.18	2.71	0.2990	341.86	250.00–T_c	51
R125	339.33	36.29	4.78	0.3052	225.06	173.00–T_c	35
R134a	374.24	40.59	5.02	0.3268	247.08	185.00–T_c	39
H_2O	647.10	220.64	17.87	0.3443	373.12	325.00–T_c	66
C_7H_{16}	540.13	27.27	2.32	0.3490	371.53	270.00–T_c	54

Table 2
Parameters, RMSD and RMSDr for Rackett equation (7), 2-parameter equation (8) and 1-parameter equation (8) with correlation (11)

	H_2	D_2	Ne	Ar	Kr
Z_{RA}	0.325701	0.328306	0.302083	0.29498	0.288685
RMSD	0.02	0.01	0.02	0.01	0.01
RMSDr (%)	0.9	0.9	1.5	0.6	0.6
a	0.6749794	0.6839145	0.7440979	0.7489134	0.7592822
b	0.3153909	0.3337335	0.3421012	0.3317864	0.3288551
RMSD	0.01	0.01	0.01	0.00	0.00
RMSDr (%)	0.7	0.6	0.9	0.0	0.1
b	0.3187854	0.3299963	0.3256916	0.3234963	0.3170940
RMSD	0.01	0.01	0.02	0.01	0.01
RMSDr (%)	0.7	0.6	1.1	0.4	0.6
	Xe	CH_4	O_2	N_2	F_2
Z_{RA}	0.285851	0.293536	0.291739	0.290483	0.304937
RMSD	0.00	0.01	0.01	0.01	0.02
RMSDr (%)	0.3	0.4	0.6	0.6	1.6
a	0.7571569	0.7482718	0.7544534	0.7572631	0.7436589
b	0.3203486	0.3283942	0.3311013	0.3312663	0.3499347
RMSD	0.00	0.00	0.00	0.01	0.01
RMSDr (%)	0.1	0.0	0.1	0.3	0.4
b	0.3145823	0.3238401	0.3237516	0.3247854	0.3420198
RMSD	0.01	0.00	0.01	0.01	0.01
RMSDr (%)	0.3	0.2	0.4	0.3	0.6
	CO	C_2H_4	C_2H_6	NF_3	C_3H_6
Z_{RA}	0.285658	0.282755	0.283458	0.264823	0.267447
RMSD	0.01	0.01	0.00	0.01	0.01
RMSDr (%)	0.6	0.5	0.3	0.6	0.5
a	0.7683354	0.7713770	0.7674907	0.7924894	0.8019922
b	0.3320236	0.3285082	0.3259824	0.3082062	0.3249273
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.2	0.1	0.1	0.2	0.2
b	0.3219182	0.3241135	0.3267215	0.3093205	0.3153752
RMSD	0.01	0.00	0.00	0.00	0.01
RMSDr (%)	0.5	0.2	0.1	0.2	0.6
	C_3H_8	*i*-C_4H_{10}	*n*-C_4H_{10}	R22	CO_2
Z_{RA}	0.273584	0.271023	0.271929	0.268779	0.268308
RMSD	0.00	0.00	0.00	0.00	0.01
RMSDr (%)	0.1	0.3	0.2	0.1	0.5
a	0.7828663	0.7890920	0.7858455	0.7974639	0.8003152
b	0.3191917	0.3196161	0.3183360	0.3234936	0.3245757
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.1	0.2	0.1	0.1	0.0
b	0.3233674	0.3246415	0.3272815	0.3260549	0.3252737
RMSD	0.00	0.01	0.01	0.00	0.00
RMSDr (%)	0.2	0.3	0.5	0.2	0.0
	C_5H_{12}	NH_3	R143a	R152a	R32
Z_{RA}	0.268302	0.237171	0.259415	0.256534	0.246911
RMSD	0.01	0.02	0.00	0.00	0.00
RMSDr (%)	0.3	0.6	0.2	0.2	0.2
a	0.7912326	0.8598304	0.8144805	0.8210739	0.8447267
b	0.3154526	0.3096690	0.3185632	0.3183938	0.3189232
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.1	0.0	0.1	0.1	0.1
b	0.3268987	0.2911955	0.3174143	0.3143127	0.3033373
RMSD	0.01	0.03	0.00	0.01	0.02
RMSDr (%)	0.6	1.0	0.1	0.2	0.9

Table 2 (*Continued*)

	R123	R124	C_6H_{14}	R125	R134a
Z_{RA}	0.265406	0.267517	0.263335	0.265358	0.259582
RMSD	0.00	0.00	0.01	0.00	0.00
RMSDr (%)	0.2	0.4	0.5	0.2	0.2
a	0.7993085	0.7973294	0.7993375	0.7990249	0.8138468
b	0.3171035	0.3200154	0.3119135	0.3167709	0.3182686
RMSD	0.00	0.00	0.01	0.00	0.00
RMSDr (%)	0.2	0.2	0.2	0.1	0.1
b	0.3242385	0.3266135	0.3211916	0.3233557	0.3151565
RMSD	0.01	0.01	0.01	0.01	0.00
RMSDr (%)	0.4	0.4	0.5	0.4	0.2
	H_2O	C_7H_{16}			
Z_{RA}	0.240824	0.256519			
RMSD	0.06	0.02			
RMSDr (%)	2.2	0.7			
a	0.8289490	0.8148013			
b	0.2873880	0.3117860			
RMSD	0.02	0.01			
RMSDr (%)	0.9	0.3			
b	0.2896030	0.3083284			
RMSD	0.02	0.01			
RMSDr (%)	1.0	0.4			

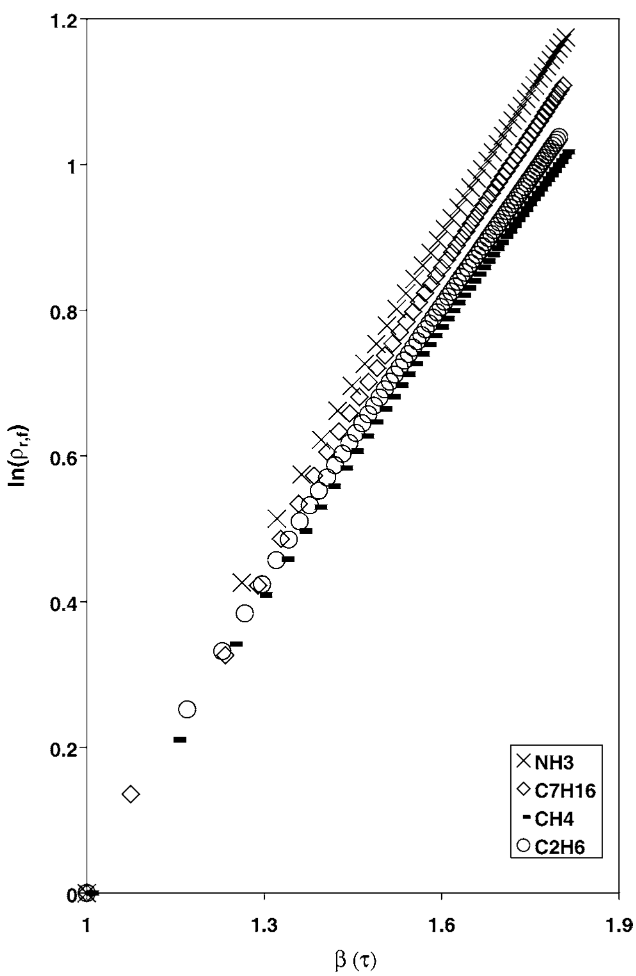

Fig. 1. Reduced saturation liquid density in the $(\beta(\tau), \ln(\rho_{r,f}))$ plane.

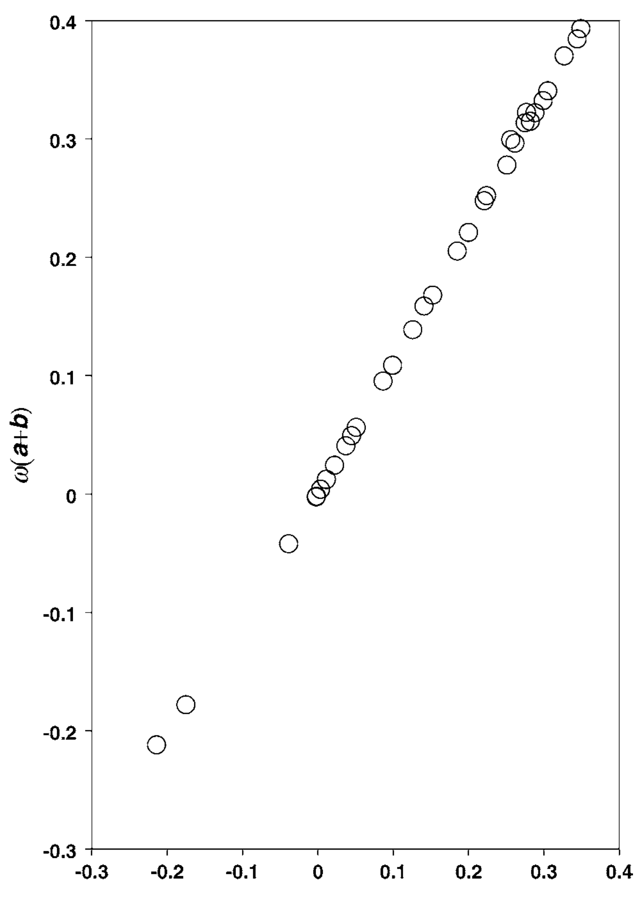

Fig. 2. $\omega(a + b)$ variation vs. acentric factor ω.

modest deviations from ideal gas behavior. Although the Lee–Kesler (LK) and the more elaborated EOSs (BACK, PC-SAFT, etc. [9]) are applicable over broader ranges of temperature and pressure and are capable of representing liquid-phase behavior, many specific computational problems are encountered when they are used [8].

2.1. Saturated liquid density

The standard equation to estimate saturated liquid density is the correlation of Rackett [1]:

$$\rho_f = \left(\frac{RT_c}{P_c}\right) Z_c^{-(1+\tau^{2/7})} \qquad (5)$$

where τ is given as

$$\tau = 1 - T_r \qquad (6)$$

To enhance the predictive capabilities of (5), Spencer and Danner proposed to replace the critical compressibility factor of the fluid, Z_c, by an adjustable parameter, Z_{RA}, the Rackett compressibility factor [1,8]

$$\rho_f = \left(\frac{RT_c}{P_c}\right) Z_{RA}^{-(1+\tau^{2/7})} \qquad (7)$$

where Z_{RA} is the only substance-specific constant in the equation. Sample values of Z_{RA} found by using (2) are listed in Table 2. Correlation (7) predicts accurately the saturated liquid density with an overall RMSDr of 0.55%, except for polar substances like water where it is of about 2.25% (cf. Table 2).

The models we proposed are intended to reproduce equally well the saturation data of all considered fluids polar as well as non-polar. The saturated liquid density is expressed as a function of the reduced temperature, T_r, for each fluid. Fig. 1 shows that $[\ln(\rho_{r,f})]$ can be expressed with the help of the intermediate function $\beta(\tau)$:

$$\ln(\rho_{r,f}) = a(\beta - \exp(1-\beta)) \qquad (8)$$

with

$$\beta(\tau) = 1 + \tau^b \qquad (9)$$

Besides its simplicity—only two adjustable fluid-specific parameters (a and b) are involved—the model fulfills the requirement of a steep decrease of the liquid density in the close vicinity of the critical point [3]:

$$\lim_{T\to T_c}\left(\frac{\partial \rho_f}{\partial T}\right) = -\infty \qquad (10)$$

This constraint is equivalent to the two conditions $ab > 0$ and $b < 1$.

2.1.1. Results and discussion

Correlation (8) can be considered either as a two or as a one adjustable parameter equation. If the constants a and b are treated separately, it is a two-parameter equation with an overall RMSDr of 0.23% (cf. Table 2). If, on the other hand, they are both correlated to the acentric factor ω (Fig. 2)—then they are dependent on one another—we found that

$$\frac{1}{\omega(a+b)} = a_1 + a_2\omega^3 + \frac{a_3}{\omega} \qquad (11)$$

where a_1, a_2 and a_3 are constants given in Table 3. In this case, the overall RMSDr increases to 0.45%.

Parameter b in Eq. (9) could not be correlated well with the acentric factor ω (Fig. 3) or the critical compressibility factor Z_c; if it is done, the inaccuracy of the predicted saturated density increases strongly.

2.2. Saturated vapor density

The same gases of Table 1 are considered here, except hydrogen and deuterium.

An equation for $\rho_{r,v}$ as a function of the temperature is established by plotting, for each fluid, $[\ln(\rho_{r,v})]$ versus $\alpha(\tau)$, a function of the reduced temperature

$$\alpha(\tau) = \exp(\tau^{1/3} + \tau^{1/2} + \tau + \tau^m) \qquad (12)$$

where $[\ln(\rho_{r,v})]$ is found to depend on $\alpha(\tau)$ as follows (Fig. 4):

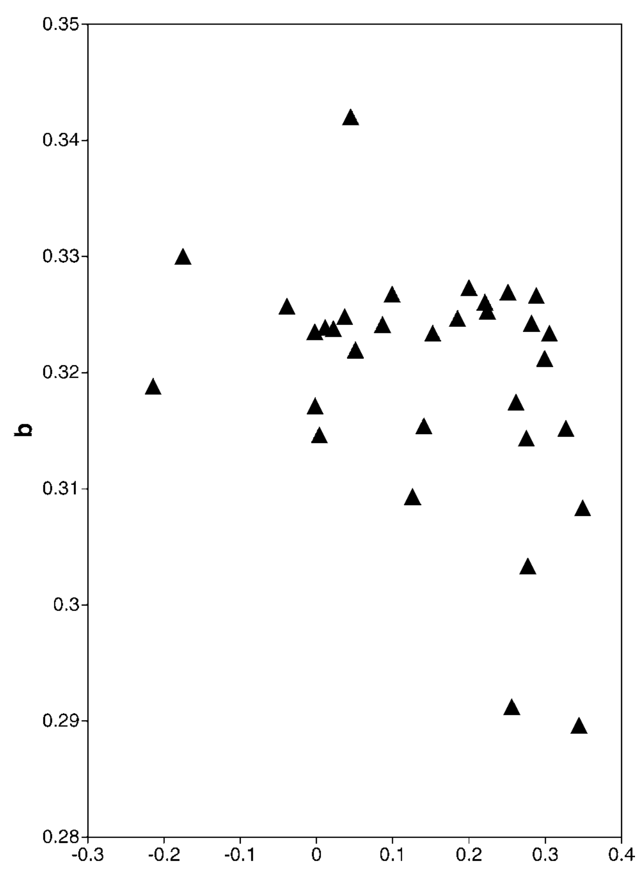

Fig. 3. b variation vs. acentric factor ω.

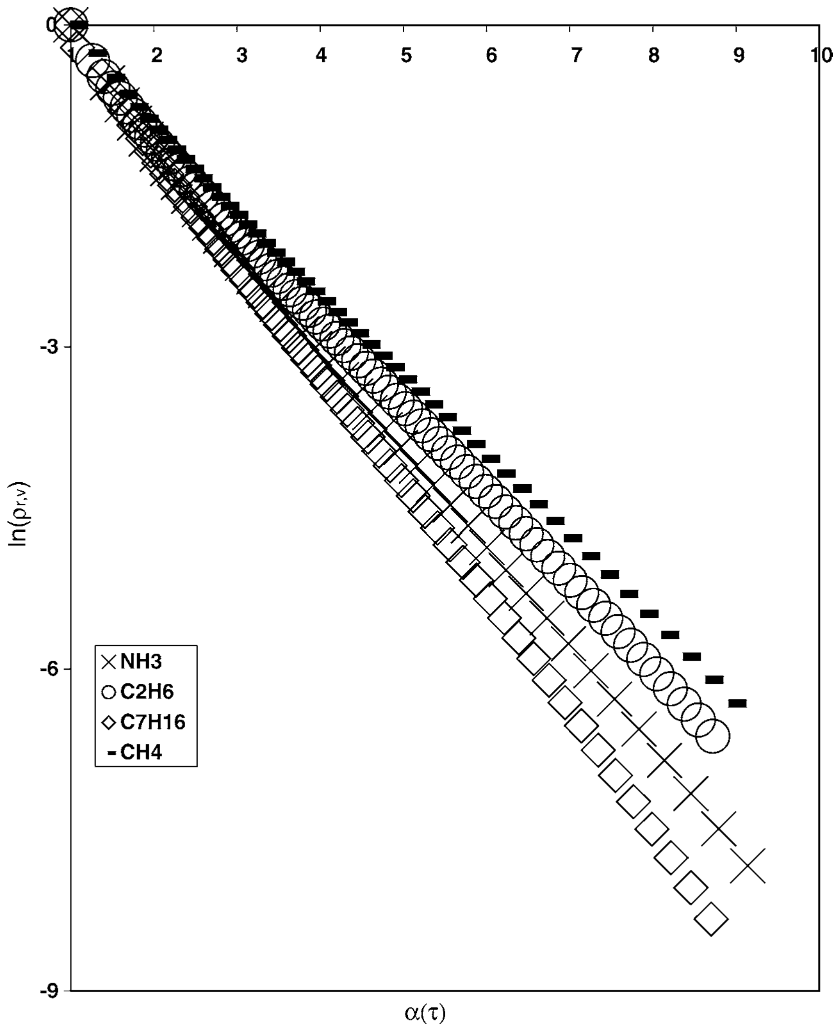

Fig. 4. Reduced saturation vapor density in the $(\alpha(\tau), \ln(\rho_{r,v}))$ plane.

$$\ln(\rho_{r,v}) = p[\alpha(\tau)^n - \exp(1 - \alpha(\tau))] \quad (13)$$

2.2.1. Results and discussion

Like Eq. (8), model (13) is made to fulfill the requirement of a steep increase of the density in the vicinity of the critical region:

$$\lim_{T \to T_c} \left(\frac{\partial \rho_v}{\partial T} \right) = +\infty \quad (14)$$

which implies if $m > 0$, then $(-p)(n+1)$ must also to be.

It is found that the parameters n and p are correlated to the acentric factor ω through the simple relation (Fig. 5):

$$\frac{1}{\omega(n-p)} = \frac{n_1 + n_2}{\omega} \quad (15)$$

The values of the parameters n_1 and n_2 are given in Table 4. Correlation (13) can be viewed as a two adjustable parameter equation (m and n or m and p) but can also be reduced to just a one parameter equation when the parameter p is expressed as function of the critical compressibility factor Z_c

$$\frac{Z_c}{p} = \frac{p_1 + p_2 Z_c \ln(Z_c) p_3}{Z_c} \quad (16)$$

where p_1, p_2 and p_3 are three adjustable constants as given in Table 4.

Thus, model (13) can be viewed either as a three-, a two- or just a one-parameter equation, depending on whether correlations (15) and (16) are considered or not. In either case, the proposed correlation reproduces accurately the vapor density at saturation. For instance, in the first case (three

Table 3
Constants a_1, a_2 and a_3 in Eq. (11)

a_1	a_2	a_3
−0.2675412	3.1738461	0.9395465

Table 4
Constants n_1, n_2, p_1, p_2 and p_3 in Eqs. (15) and (16)

n_1	n_2	p_1	p_2	p_3
−0.1497547	0.6006565	−19.348354	−41.060325	1.1878726

Table 5
Parameters RMSD and RMSDr for 3-parameter equation (13) and 1-parameter equations (13) with correlations (15) and (16)

	Ne	Ar	Kr	Xe	CH_4
m	4.4960428	3.1601166	3.4377347	3.2336283	2.9407241
n	1.1861523	1.1183779	1.1459094	1.1272991	1.1012273
p	−0.485720905	−0.5515808	−0.5312377	−0.5475391	−0.5697413
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.0	0.2	0.2	0.1	0.5
m	3.3849806	2.9759058	3.0146411	2.9689106	2.8902994
RMSD	0.01	0.00	0.00	0.00	0.00
RMSDr (%)	1.9	0.7	1.1	0.7	0.6
	O_2	N_2	F_2	CO	C_2H_4
m	2.9401387	2.8539512	2.5237557	2.1804625	2.6032071
n	1.1115040	1.1131452	1.0739354	1.0460487	1.1016984
p	−0.5649451	−0.5673304	−0.6052029	−0.6091577	−0.5971817
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.5	0.5	0.7	0.6	0.6
m	2.8687783	2.8518666	2.6082807	2.8356952	2.6708569
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.6	0.5	0.9	1.1	0.6
	C_2H_6	NF_3	C_3H_6	C_3H_8	*i*-C_4H_{10}
m	2.6379996	2.7962186	3.1010584	2.5491601	2.5860317
n	1.1048142	1.1483585	1.1872850	1.1169908	1.1380881
p	−0.6018817	−0.5712755	−0.5517088	−0.6110244	−0.6069818
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.5	0.5	0.8	0.7	1.1
m	2.6600497	2.7864527	2.7598233	2.6308038	2.6103362
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	0.7	0.7	1.1	1.1	1.3
	n-C_4H_{10}	R22	CO_2	C_5H_{12}	NH_3
m	2.4703930	2.4474190	2.4686277	2.4715148	2.9025748
n	1.1187258	1.1245385	1.1345838	1.1326580	1.1747326
p	−0.629651003	−0.6346948	−0.6240188	−0.6412635	−0.6213074
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.1	1.1	0.2	1.3	0.9
m	2.5923782	2.5118465	2.5301133	2.5474417	2.3891690
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.6	1.2	0.6	1.5	1.8
	R143a	R152a	R32	R123	R124
m	2.4390945	2.4036109	2.4973193	2.3683423	2.4354428
n	1.1210065	1.1186849	1.1207203	1.1374290	1.1506277
p	−0.6625750	−0.6709599	−0.6786065	−0.6444662	−0.6362176
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.4	1.5	1.3	1.8	1.7
m	2.3804779	2.3538883	2.2118258	2.5631044	2.5790251
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.4	1.5	1.7	1.9	1.7
	C_6H_{14}	R125	R134a	H_2O	C_7H_{16}
m	2.5036259	2.3507937	2.4203360	2.3609558	2.5477720
n	1.1549903	1.1406387	1.1516298	1.0916682	1.1770021
p	−0.6410813	−0.6493031	−0.6546052	−0.7452828	−0.6419602
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.6	1.4	1.8	1.6	1.7
m	2.5701781	2.6190442	2.5327802	2.0694497	2.6238695
RMSD	0.00	0.00	0.00	0.00	0.00
RMSDr (%)	1.6	1.6	1.9	1.7	1.7

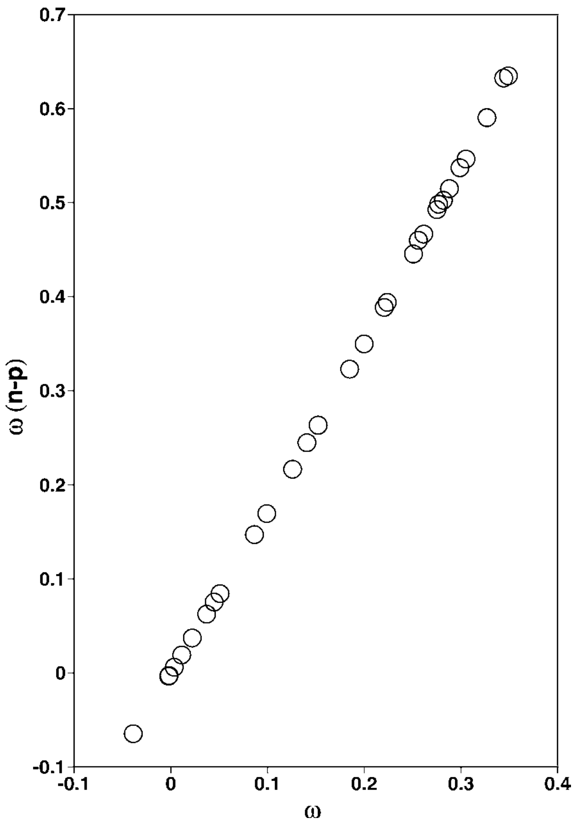

Fig. 5. $\omega(n-p)$ variation vs. acentric factor ω.

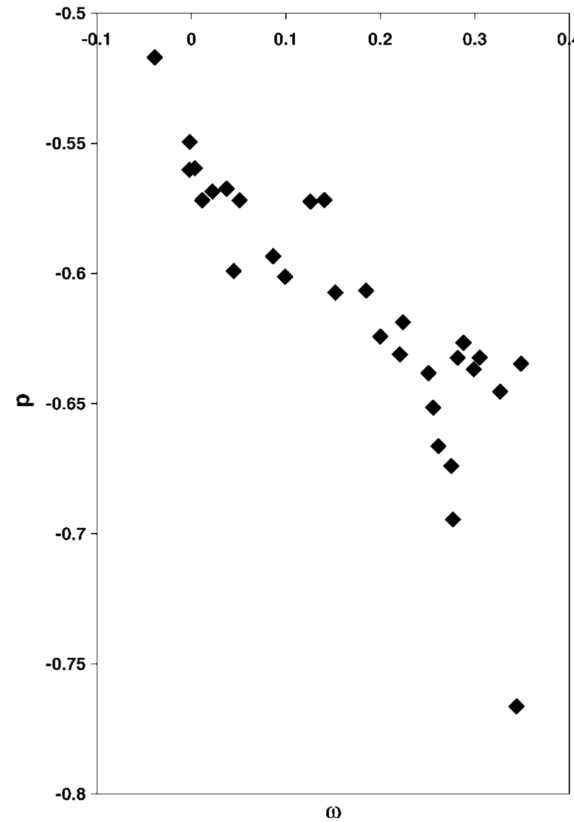

Fig. 7. p variation vs. acentric factor ω.

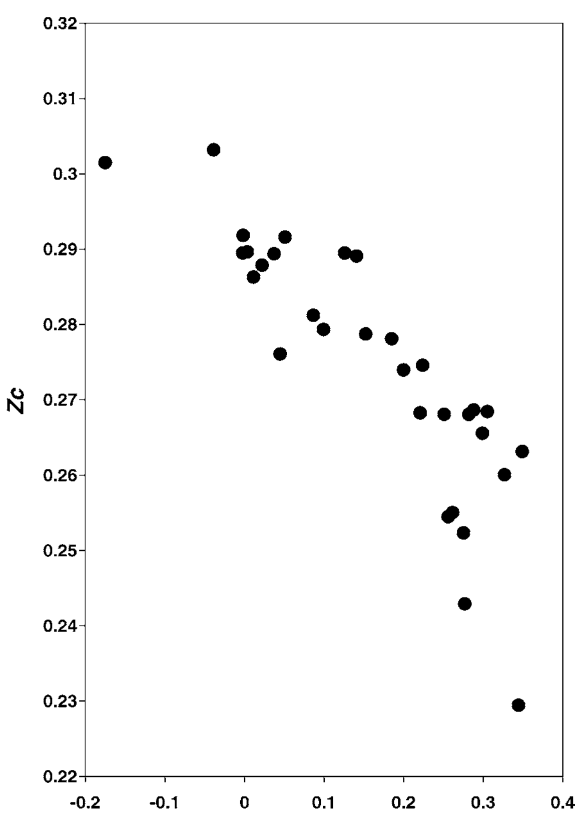

Fig. 6. Critical compressibility factor Z_c vs. acentric factor ω.

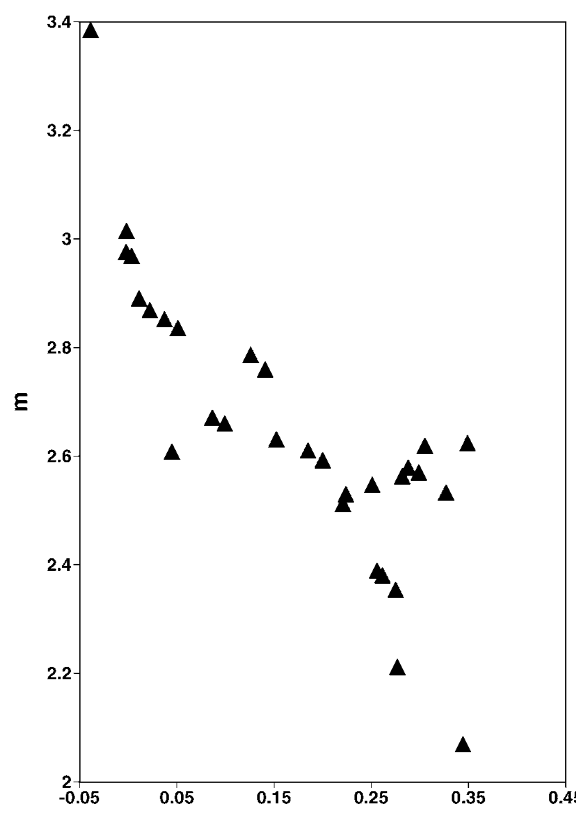

Fig. 8. m variation vs. acentric factor ω.

parameters) the overall RMSDr is under 1% (0.98%) (cf. Table 5). Its value is 1.25% (cf. Table 5) in the one-parameter case.

In view of the obtained results some general remarks can be formulated:

- Because of the non-regular behavior of Z_c on ω (Fig. 6) for the considered fluids, parameter p could not be expressed as a simple function of the acentric factor ω (Fig. 7).
- Although a correlation between m and the acentric factor ω (Fig. 8) gives a three-parameter corresponding state equation, the accuracy of the model decreases this way.
- For each fluid, the inaccuracy of the predicted vapor density at saturation increases substantially as the critical region approaches.

3. Conclusion

The vapor–liquid envelope of water and ammonia are shown in Figs. 9 and 10, respectively, in comparison with the saturated data. The good agreement observed illustrates the quality of the proposed analytical expressions for the liquid and vapor density. Thus the proposed models—developed for the use in the refrigeration machines's simulation software—are accurate, simple and can be used for polar as well as non-polar fluids.

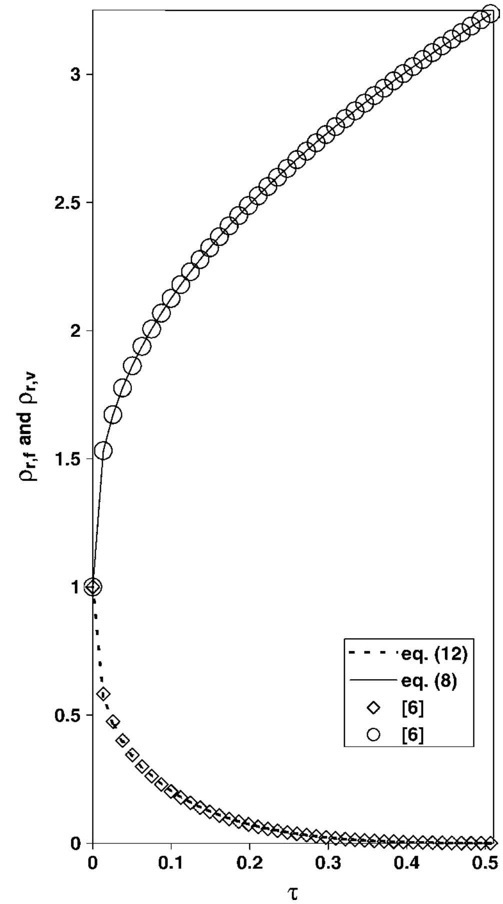

Fig. 10. $\rho_{r,f}$ and $\rho_{r,v}$ vs. τ for NH_3.

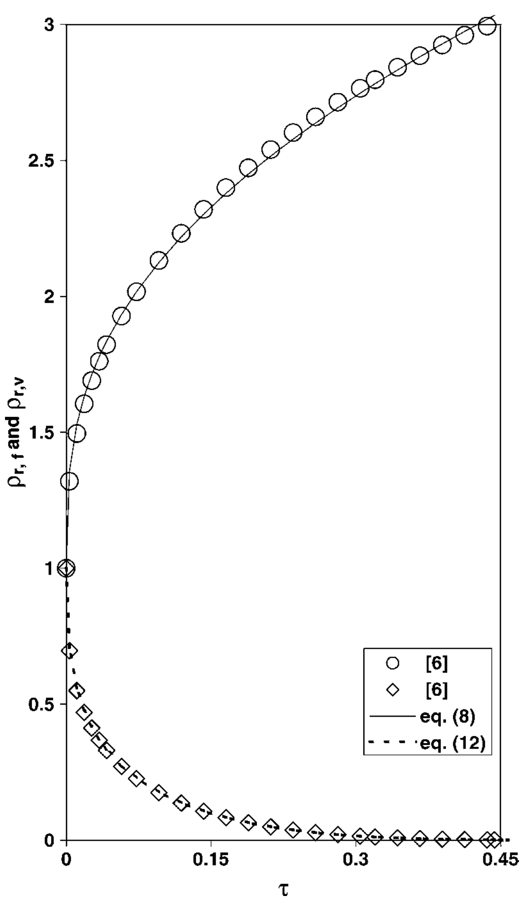

Fig. 9. $\rho_{r,f}$, $\rho_{r,v}$ vs. τ for H_2O.

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