

## Prediction of halocarbon liquid densities by a modified Hard Sphere–De Santis equation of state

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### Abstract

The present paper describes a general scheme for the prediction of halocarbon liquid state densities based on a modified hard sphere De Santis equation of state. It is shown that it is possible to estimate the density of pure liquids solely from the critical constants and relative molecular mass to better than 1.5% for  $T^* < 0.9$  and from saturation vapour pressures up to 20 MPa.

**Keywords:** Theory; Equation of state; Corresponding states; Density; Liquid halocarbons

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### 1. Introduction

The needs for environmentally acceptable fluorocarbons for refrigeration, air conditioning, foam blowing, solvent cleaning and aerosol propellants have triggered a world wide research effort in the properties of substances with low ozone depletion and global warming potentials.

The increasing need for information on liquid density for the new refrigerants, has led the authors to try to implement a new scheme to correlate and estimate them from limited information. This scheme is not a replacement for the use of individual experimental data or equations of state of the halocarbon refrigerants. However the need of easy programable techniques of general use and moderate accuracy justifies the corresponding states approach.

As a starting point we have used the hard sphere De Santis (Sousa et al., 1992) equation to correlate the available experimental data of the density of liquid refrigerants (R114, R123, R142b, R152a, R22), in order to be able to obtain the parameters  $a(T)$  and  $b(T)$  needed to develop the model of the reduced equation of state.

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The new model has been used to predict the data available for other fluids (R32, R125, R134a, R134, R141b), with an error within  $\pm 1.5\%$  for  $T^*$  less or equal to 0.9. To the author's best knowledge there is no other density estimation method applicable to the halocarbons with such a high accuracy.

## 2. The model equation of state

The Carnahan–Starling De Santis (CSDS) equation of state (De Santis et al., 1976, see Morrison and McLinden, 1986) was used with reasonable success, by McLinden et al. (Morrison and McLinden, 1986), in correlating the saturation lines and vapour phase of the refrigerants,

$$Z = \frac{1 + y + y^2 - y^3}{(1 - y)^3} - \frac{a\rho}{RT(1 + b\rho)} \quad (1)$$

where  $y = b\rho/4$ ,  $Z$  the compressibility factor,  $\rho$  the density,  $R$  the gas constant,  $T$  the temperature, and  $a$  and  $b$  are van der Waals type parameters, usually considered as adjustable.

In order to develop an equation of state to be applied to the liquid phase, including the saturation boundary, we decided to make a fundamental modification to Eq. (1). The Carnahan–Starling term  $(1 + y + y^2 - y^3)/(1 - y)^3$  is a Padé approximant [3,3] to the six virial expansion of the hard-sphere virials (Carnahan and Starling, 1969). Since the publication of this equation, the virial coefficients of the hard-spheres have been calculated up to the tenth (Ree and Hoover, 1964, Erpenbeck and Wood, 1984). We have therefore replaced the first term of Eq. (1) (the Carnahan–Starling approximation for the hard-sphere state equation) with the first ten virial coefficients of the hard-sphere equation, in order to obtain the best available theoretical description of the repulsive contribution,

$$Z = \sum_{i=1}^{10} C_i (b\rho)^{i-1} - \frac{a\rho}{RT(1 + b\rho)} \quad (2)$$

where  $C_i$  are the virial constants obtained from Monte Carlo and Molecular Dynamics simulations (Ree and Hoover, 1964, Erpenbeck and Wood, 1984). The values of  $C_i$  are given in Table 1.

The parameters  $a$  and  $b$  were assumed as temperature dependent, and the authors have previously proposed (Sousa et al., 1992) the following relations:

$$a = a_0 + a_1 T + a_2 T^2 \quad (3)$$

$$b = b_0 + b_1 T + b_2 T^2 \quad (4)$$

The available data on the density of R114 (Wilson and Hules, 1981, Fukuizumi and Uematsu,

Table 1

Virial coefficients for the hard-spheres (Ree and Hoover, 1964, Erpenbeck and Wood, 1984)

$C_1$	1	$C_6$	$0.0386 \pm 0.0004$
$C_2$	1	$C_7$	$0.0137 \pm 0.006$
$C_3$	$5/8$	$C_8$	0.00421
$C_4$	0.28695	$C_9$	0.00131
$C_5$	$0.1103 \pm 0.0003$	$C_{10}$	0.00040

Table 2

Experimental range of the available data (reduced pressure and temperature) used in the correlation of Eq. (2)

Refrigerant	$T/T_c$	$p/p_c$
R114 <sup>a</sup>	0.74–1.00	0.15–3.20
R123 <sup>b</sup>	0.61–1.00	0.03–3.25
R142b <sup>c</sup>	0.51–1.00	0.002–4.30
R152a <sup>d</sup>	0.83–1.00	0.33–2.21
R22 <sup>e</sup>	0.46–0.95	0.0002–6.12

<sup>a</sup> (Wilson and Hules, 1981; Fukuizumi and Uematsu, 1991a); <sup>b</sup> (Piao et al., 1991; Maezawa et al., 1990; Maezawa et al., 1991a); <sup>c</sup> (Sousa et al., 1992; Maezawa et al., 1991a); <sup>d</sup> (Iso and Uematsu, 1989); <sup>e</sup> (Blanke et al., 1988; Fukuizumi and Uematsu, 1991b; Defibaugh and Morrison, 1992).

1991a), R123 (Piao et al., 1991, Maezawa et al., 1990, Maezawa et al., 1991a), R142b (Sousa et al., 1992, Maezawa et al., 1991a), R152a (Iso and Uematsu, 1989) and R22 (Blanke et al., 1988, Fukuizumi and Uematsu, 1991b, Defibaugh and Morrison, 1992), and the respective ranges of temperature and pressure, in reduced units, are presented in Table 2. The data was correlated using Eq. (2) for each fluid. Using a non-linear adjustment (Press et al., 1986) (Levenberg–Marquadt method) it was possible to obtain the parameters for each refrigerant as presented in Table 3. Fig. 1 shows the comparison of the correlation with the experimental data for  $T^* = T/T_c$  less than or equal to 0.95. The deviations are less than  $\pm 0.5\%$  which is comparable with the experimental uncertainty (less than  $\pm 0.3\%$ ), and no systematic deviations are found for any fluid.

### 3. The general equation of state

To obtain a general equation of state, the authors have chosen to represent the parameters obtained for the different refrigerants as a function of  $T^*$  in order to observe their behaviour (Figs. 2 and 3).

As it can be seen, in the case of  $b$ , the temperature dependence is so small that one can assume that it is negligible ( $b \approx b_0$ ). As a second step, we reduced the parameters  $a$  and  $b$  with the critical constants according to the expressions:

$$a^* = \frac{a\rho_c^2}{p_c} \quad (5)$$

$$b^* = b\rho_c \quad (6)$$

Table 3

Parameters for the Eqs. (3) and (4)

	$a_0 \times 10^{-4}$	$a_1 \times 10^{-1}$	$a_2 \times 10^2$	$b_0 \times 10^4$	$b_1 \times 10^7$	$b^2 \times 10^{10}$	$\sigma/P_c$
R114	2.234954	−8.721153	7.932981	5.921175	−1.237742	3.156901	0.024
R123	2.386772	−8.725431	7.487230	5.471463	−0.344884	0.769728	0.077
R142b	1.347903	−5.227032	4.622546	4.804615	−3.553399	6.765317	0.041
R152a	0.811270	−3.126128	2.525539	−0.738200	23.742769	−34.308257	0.051
R22	0.621782	−2.222513	1.335290	2.794842	1.641533	−0.914907	0.027

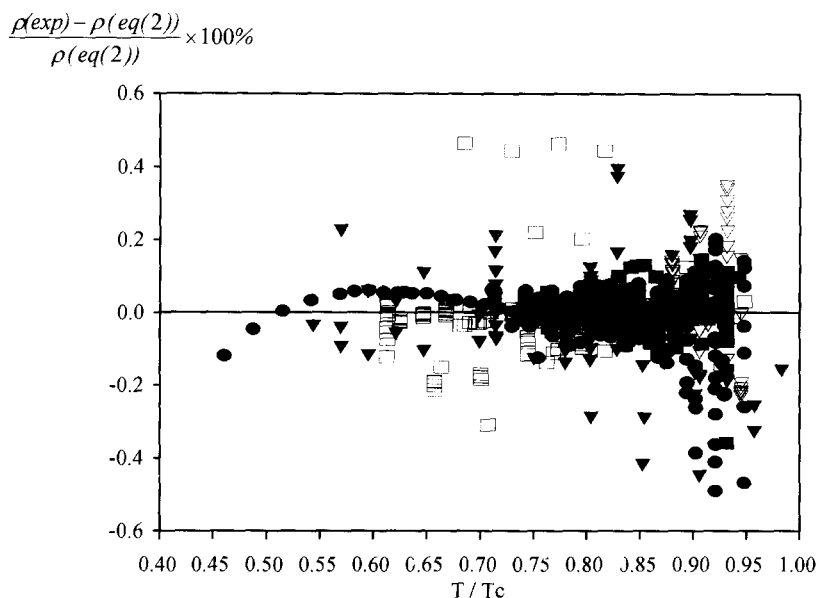


Fig. 1. Deviations between the experimental density data and the estimation using the correlation expressed by Eq. 2, using the parameters of Tables 1 and 3. ■, R114; □, R123; ▼, R142b; ▽, R152a; ●, R22.

In the literature there are many independent measurements values for the critical constants of these compounds. It was not possible to identify for some of the values their internal consistence and uncertainty. In order to select the best available set of data to be used in the general correlation, we have looked to similarity in the properties of liquid refrigerants. It appears that the dependence of  $Z_c = p_c/R\rho_cT_c$  on the relative molecular mass is a good measure for the consistency of the available

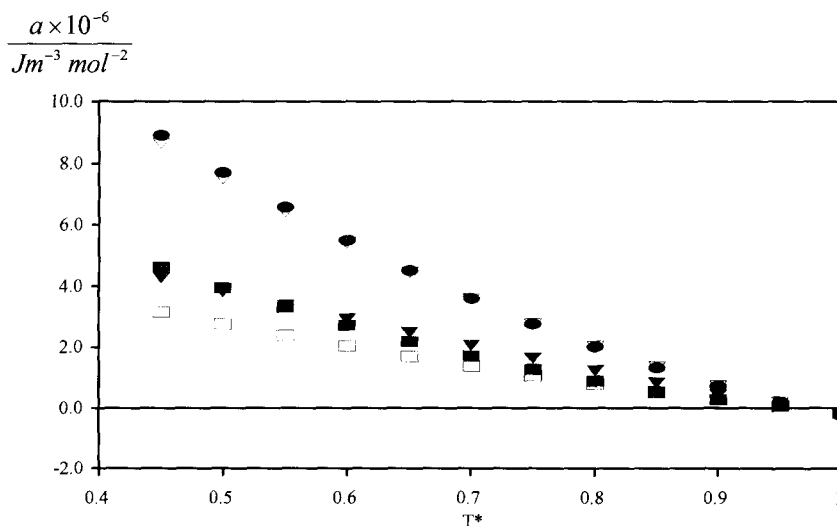


Fig. 2. Behaviour of  $a(T)$  for several refrigerants. ■, R152a; □, R22; ▼, R142b; ▽, R123; ●, R114.

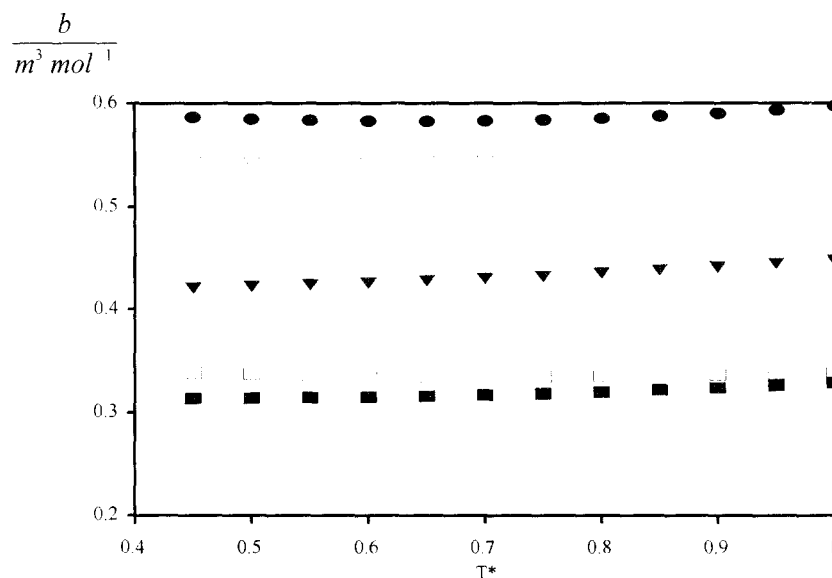


Fig. 3. Behaviour of  $b(T)$  for several refrigerants. ■, R152a; □, R22; ▼, R142b; ▽, R123; ●, R114.

data on the critical constants (Fig. 4). This fact associated with an analysis of the possible presence of impurities in the halocarbons studied by several authors led us to choose the values of the critical constants displayed in Table 4.

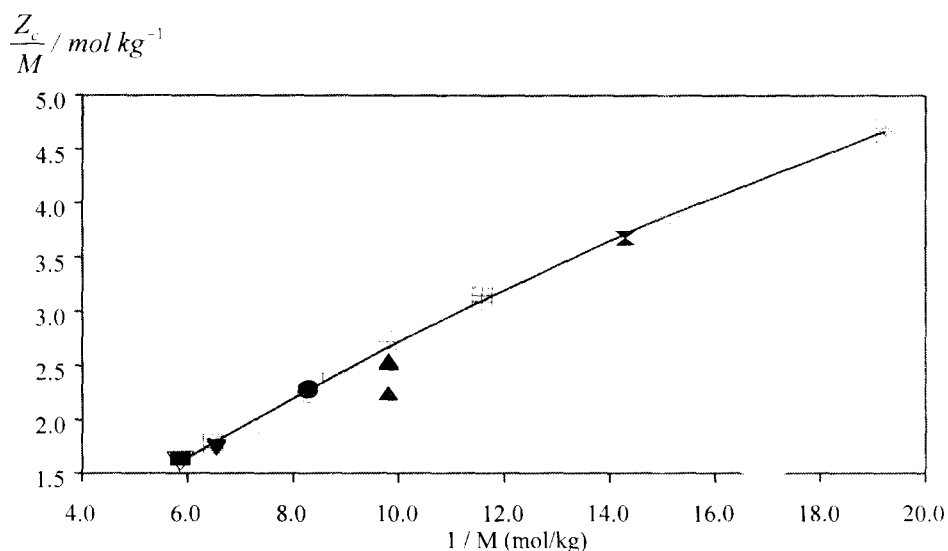


Fig. 4. Behaviour of the ratio of the critical compressibility constant over the relative molecular mass as a function of the inverse of the relative molecular mass. The line was obtained by the use of Eq. (7). □, R115; ●, R12; ▼, R123; ▲, R134a; ▽, R114; Δ, R134; +, R141b; ×, R142b; |, R124; ○, R125; ■, R114a; ⊠, R32; ⊞, R22; ⋈, R152a; ⚡, R23; —, Eq. (7).

Table 4

Critical constants selected from the literature based on Eq. (7)

Refrigerant	$M$ (kg mol <sup>-1</sup> )	$P_c$ (MPa)	$T_c$ (K)	$\rho_c$ (kg m <sup>-3</sup> )
R114a	0.170922	3.303 <sup>a</sup>	418.7 <sup>a</sup>	578 <sup>a</sup>
R114	0.170922	3.252 <sup>b</sup>	418.78 <sup>b</sup>	576 <sup>b</sup>
R115	0.154470	3.118 <sup>c</sup>	353.2 <sup>d</sup>	604 <sup>c</sup>
R123	0.152930	3.674 <sup>e</sup>	456.94 <sup>e</sup>	550 <sup>e</sup>
R124	0.136475	3.634 <sup>f</sup>	395.65 <sup>g</sup>	560 <sup>g</sup>
R12	0.120930	4.116 <sup>h</sup>	385.15 <sup>h</sup>	568 <sup>i</sup>
R125	0.120020	3.631 <sup>f</sup>	339.4 <sup>j</sup>	572 <sup>j</sup>
R141b	0.116950	4.46 <sup>l</sup>	480 <sup>l</sup>	460 <sup>l</sup>
R134a	0.102030	4.068 <sup>e</sup>	373.69 <sup>m</sup>	495.6 <sup>m</sup>
R134	0.102030	4.64 <sup>l</sup>	392 <sup>l</sup>	510 <sup>l</sup>
R142b	0.100495	4.041 <sup>n</sup>	410.25 <sup>a</sup>	446 <sup>n</sup>
R22	0.086468	4.98 <sup>o</sup>	369.32 <sup>i</sup>	515 <sup>i</sup>
R23	0.070013	4.82 <sup>p</sup>	298.98 <sup>p</sup>	526 <sup>p</sup>
R152a	0.066050	4.52 <sup>q</sup>	386.44 <sup>q</sup>	368 <sup>q</sup>
R32	0.052024	5.83 <sup>r</sup>	351.56 <sup>r</sup>	422.67 <sup>r</sup>

<sup>a</sup> (Mears et al., 1955); <sup>b</sup> (Higashi et al., 1985); <sup>c</sup> (Yada et al., 1989); <sup>d</sup> (Asahi, 1983, see Hongo et al., 1990); <sup>e</sup> (Weber and Sengers, 1990); <sup>f</sup> (McLinden, 1990); <sup>g</sup> (Kubota et al., 1988); <sup>h</sup> (Sukornick, 1989); <sup>i</sup> (Higashi et al., 1984); <sup>j</sup> (Shankland et al., 1989, McCain, 1989, see McLinden, 1990); <sup>l</sup> (Maezawa et al., 1991a); <sup>m</sup> (Du Pont, 1979, see Kabata et al., 1989); <sup>n</sup> (Tanikawa et al., 1991, see Maezawa et al., 1991b, Yada et al., 1991); <sup>o</sup> (HIR, 1982); <sup>p</sup> (Shavandrin, 1975, see McLinden, 1990); <sup>q</sup> (Higashi et al., 1987, see Maezawa et al., 1991b); <sup>r</sup> (Holcomb et al., 1992).

These values were used throughout the present study and are only shown as a consistent set for the correlation. For the case of R124 and R125 the value of  $p_c$  available in the literature was obtained by extrapolating the saturation line (McLinden, 1990).

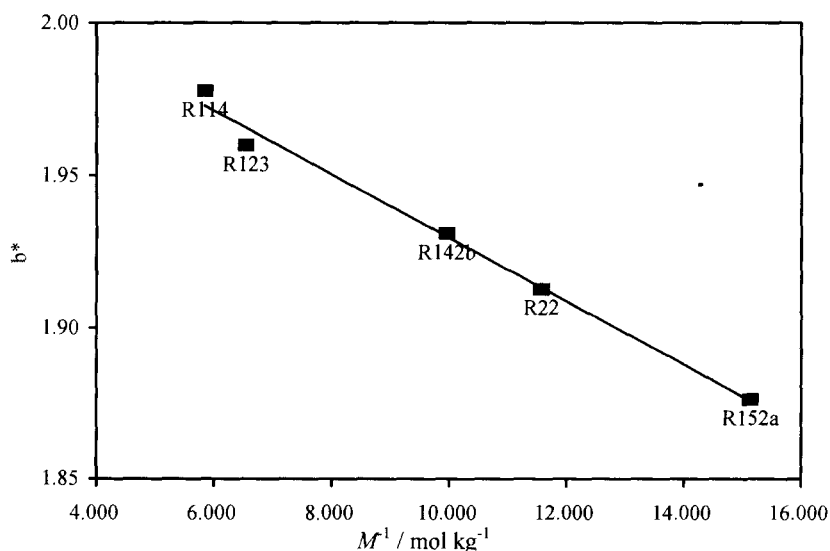


Fig. 5. Behaviour of the reduced parameter  $b^*$  as a function of the relative molecular mass for several refrigerants. The line was calculated by correlation (8).

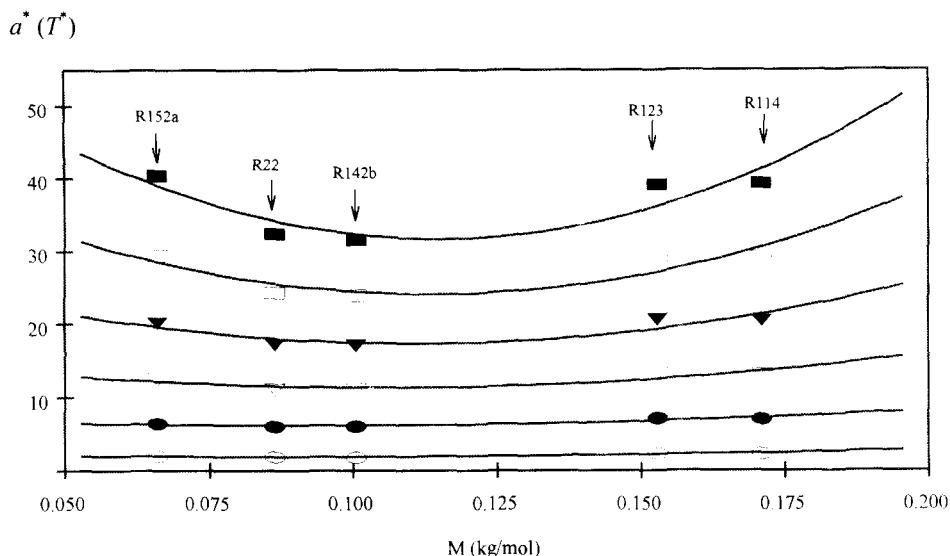


Fig. 6. Behaviour of the reduced parameter  $a^*$  as a function of the relative molecular mass and reduced temperature, for several refrigerants. The line was calculated by correlation (9). ■,  $T^* = 0.4$ ; □,  $T^* = 0.5$ ; ▼,  $T^* = 0.6$ ; ▽,  $T^* = 0.7$ ; ●,  $T^* = 0.8$ ; ○,  $T^* = 0.9$ .

The correlation obtained was:

$$Z_c = 0.317 - \frac{3.41 \times 10^{-3}}{M} - 0.138M \quad (7)$$

Most of the available critical compressibility factors can be represented by this equation within  $\pm 2\%$ . The values of  $b^*$  were found to be temperature independent and a linear function of  $M^{-1}$ , as demonstrated in Fig. 5, the line being given by Eq. 8, and the values of the parameter  $a^*$  depend on both temperature and relative molecular mass through Eq. (9):

$$b^* = (2.0335 + 0.01039M) \pm 0.0044 \quad (8)$$

$$a^* = \sum_{i=0}^2 \left( \sum_{j=0}^2 a_{ij}^* T^{*j} \right) M^i \quad (9)$$

where  $M$  is the relative molecular mass in  $\text{kg mol}^{-1}$ .

Fig. 6 shows this behaviour and the parameters  $a_{ij}^*$  are presented in Table 5.

Table 5  
Parameters used in Eq. (9)

$a_{ij}^*$	$i = 0$	$i = 1$	$i = 2$
$j = 0$	$2.069249 \times 10^5$	$-2.332869 \times 10^6$	$9.949903 \times 10^6$
$j = 1$	$-4.245370 \times 10^5$	$5.268112 \times 10^6$	$-2.222515 \times 10^7$
$j = 2$	$2.194730 \times 10^5$	$-2.991949 \times 10^6$	$1.251112 \times 10^7$

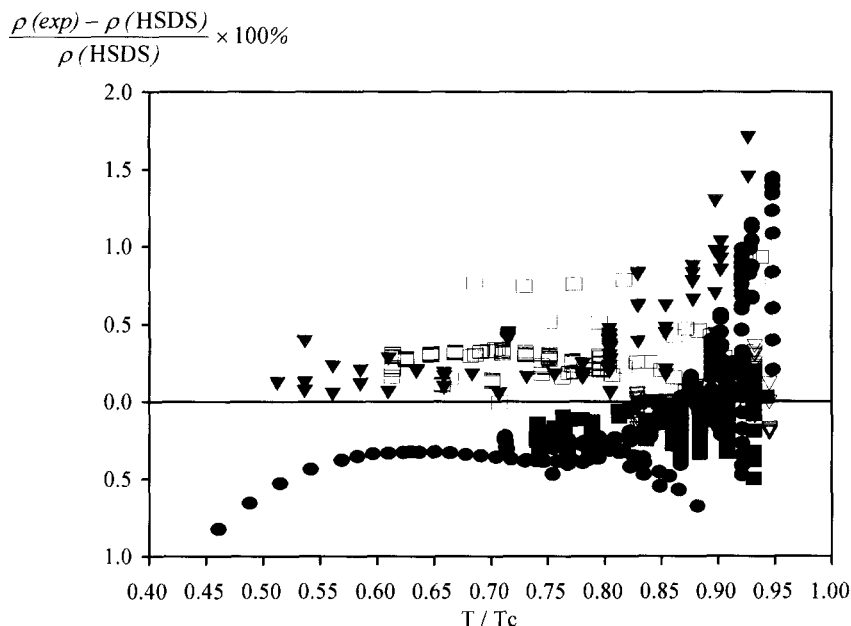


Fig. 7. Deviation plot from the experimental data, used in the development of the reduced equation, and the calculations obtained by the model.  $\square$ , R123;  $\bullet$ , R22;  $\blacktriangledown$ , R142b;  $\nabla$ , R152a;  $\blacksquare$ , R144.

In order to test the consistency of the reduced equation we have calculated the densities of the refrigerants used in the development of the general correlating scheme.

Fig. 7 shows the deviation between the experimental data of the refrigerants used in the scheme and the values obtained using Eqs. (2) and (5)–(9). As can be seen no point deviates by more than 1.6% and most of the data deviate by no more than  $\pm 1\%$  for  $T^* \leq 0.9$ , although some of the fluids show systematic trends, impossible to avoid with this type of averaged correlations.

#### 4. Discussion

The general equation developed here was tested by calculating the densities of other refrigerants not used in its development, to test the ability of the general equation herein proposed to predict density of the liquid phase. Two tests have been made:

1. To compare this scheme with previous individual correlations developed by McLinden (Morrison and McLinden, 1986, McLinden, 1990) for the saturation liquid line of the several refrigerants.
2. To compare with recently available data on R32, R125, R134, R134a and R141b.

Figs. 8 and 9 show the results of these tests. In the first case our scheme is capable of producing density values as well as individual correlation, within mutual uncertainties (Morrison and McLinden, 1986, McLinden, 1990), especially for  $T^* < 0.8$ . Deviations between the two methods are, below this temperature, around  $\pm 1\%$ .

Fig. 9 shows the capability of our scheme to estimate the densities of refrigerants not included in the development scheme, over the ranges of pressure and temperature normally used in the refrigeration industries, from saturation line to compressed liquid, between vapour pressure and 20 MPa. It can be



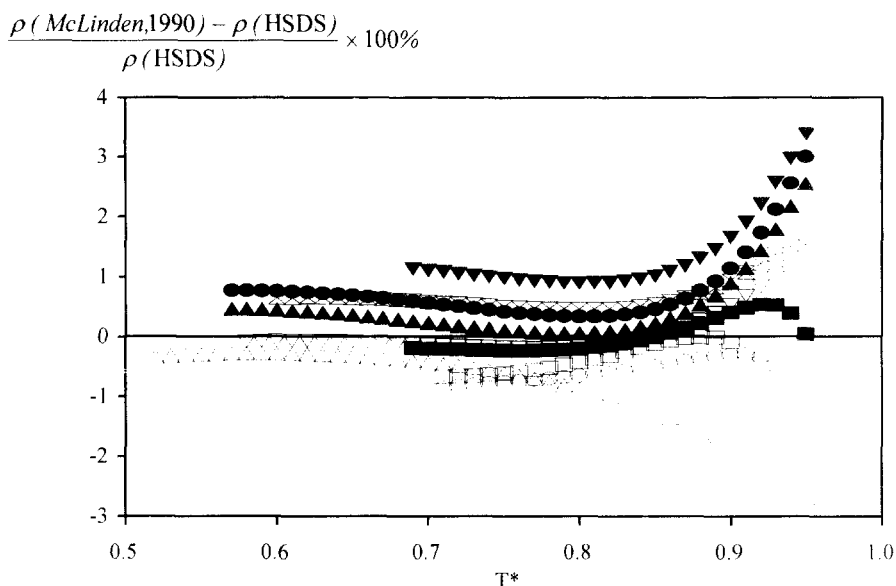


Fig. 8. Deviation plot from the liquid saturation line, given by McLinden (McLinden, 1990) for several refrigerants, and the reduced model here in presented.  $\square$ , R32;  $\bullet$ , R134a;  $\blacktriangledown$ , R125;  $\blacktriangle$ , R124;  $\nabla$ , R22;  $\Delta$ , R142b;  $\circ$ , R152a;  $\blacksquare$ , R23;  $-123$ .

seen that the model can predict the data within  $\pm 1.5\%$  for  $T^* \leq 0.9$ . Above this temperature, and as already stated, the modified HSDS equation is not capable of accommodating well the curvature of the  $\rho(p)$  isotherms as the critical point is approached.

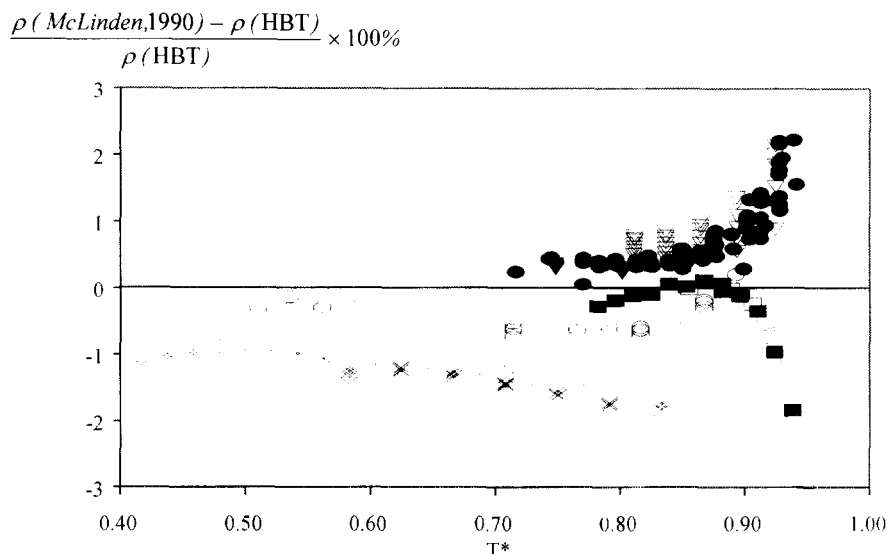


Fig. 9. Deviation plot from the experimental data obtained for several refrigerants and general equation of state here in developed.  $\square$ , R32 (Holcomb et al., 1992);  $\blacksquare$ , R32 (Widiatmo et al., 1994);  $\bullet$ , R134a (Morrison and Ward, 1991);  $\blacktriangledown$ , R134a (Maezawa et al., 1990);  $\nabla$ , R125 (Defibaugh and Morrison, 1993);  $\times$ , R141b (Maezawa et al., 1991a);  $\circ$ , R134 (Maezawa et al., 1991a).

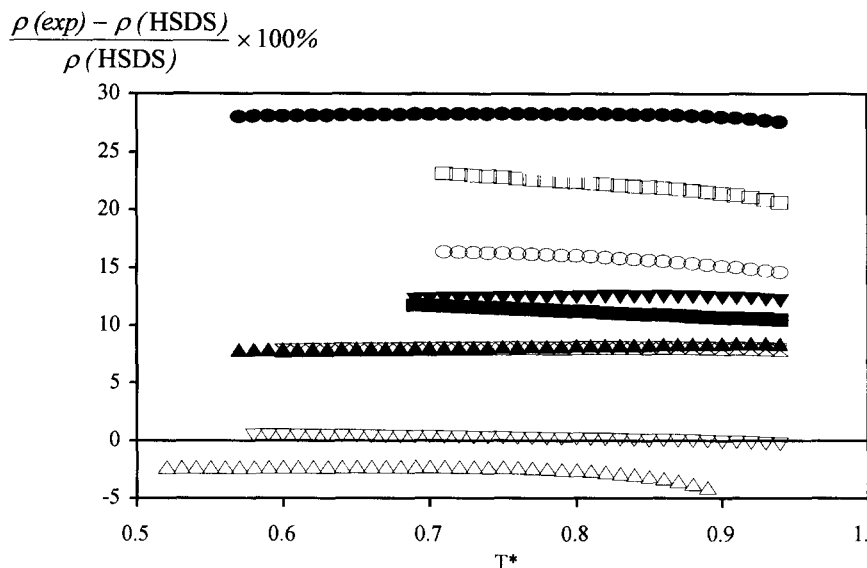


Fig. 10. Deviation plot from the liquid saturation line, given by McLinden (McLinden, 1990) for several refrigerants, and the HBT technique (Reid et al., 1986).  $\square$ , R32;  $\bullet$ , R134a;  $\blacktriangledown$ , R125;  $\blacktriangle$ , R124;  $\nabla$ , R22;  $\Delta$ , R142b;  $\circ$ , R152a;  $\blacksquare$ , R23;  $\otimes$ , R123.

In addition we have compared this scheme with an estimation method proposed by Hankinson, Brobst and Thomson (Reid et al., 1986). If we use an estimation method not based in the correlation of parameters over a wide range of states, as in this estimation technique, we can obtain systematic differences that can amount to 30%, as in the case of R134a, one of the most important replacement fluids, as it can be seen in Fig. 10.

## 5. Conclusions

A new form of the Carnahan and Starling De Santis equation of state, the Hard Sphere De Santis (HSDS) equation of state has been used to develop an general scheme for the estimation of the liquid densities of environmentally acceptable refrigerants, within  $\pm 1.5\%$  for  $T^* \leq 0.9$  and  $3\%$  for  $0.9 \leq T^* \leq 0.95$ , and pressures between vapour pressure and 20 MPa, based only on the knowledge of the critical constants,  $p_c$  and  $T_c$ . This scheme is at present limited to halocarbons of the methane and ethane series and we hope to extend it in the future to propane derivatives and haloethers. If the results obtained are similar it could be transformed into a universal prediction scheme for refrigerants. As it is now, it represents the most accurate and powerful general method of estimation of halocarbon liquid densities in the saturation line and in the compressed liquid region, based on a theoretical repulsive part and on an experimentally adjustable attractive part. It is very easy to use PC environment as it only needs 11 adjusted constants in addition to the fixed hard sphere ten virial constants. It is also easily attached to phase-equilibria and transport properties estimation packages.

In a companion paper the scheme is applied to the estimation of the density of the binary and ternary mixtures of halocarbon refrigerants.

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