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Dielectric Properties of Liquid Pentafluoroethane (HFC-125)

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This paper reports measurements of the static relative permittivity of HFC-125 in the liquid phase, performed by using the direct capacitance method at 10 kHz, for temperatures from 214 to 304 K and pressures up to 16 MPa. The repeatability of the measurements was found to be of the order of $\pm 0.7 \times 10^{-3}$ and the uncertainty is estimated to be better than $\pm 0.72 \times 10^{-2}$. We provide a complete set of tables of experimental data as a function of temperature, pressure, and density, which cover the dielectric property needs for most engineering applications. The data obtained were used to establish dielectric equations of state as a function of density and temperature and as a function of pressure and temperature. To study the dependence of the relative permittivity on temperature, pressure and density, we have applied various molecular theories of polar liquids. The apparent dipole moment obtained was $\mu^* = 2.482$ D.

KEY WORDS: dipole moment; HFC-125; Kirkwood equation; pentafluoroethane; relative permittivity; Vedam equation.

1. INTRODUCTION

The relative permittivity of alternative refrigerants is important for study of the electric properties of these polar fluids. It also gives insight into the development of the molecular theory of liquids, and at the same time, it is

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an important parameter for design of machinery used in the air-conditioning and refrigeration industry. One of the most urgent issues to be solved in these industries is to identify the optimum alternative refrigerant to replace R-502 (an azeotrope of HCFC-22 and CFC-115), which is used primarily in low-temperature refrigeration systems. A binary mixture of pentafluoroethane (HFC-125) and 1,1,1-trifluoroethane (HFC-143a, R-507) and a ternary mixture of HFC-125/134a/143a (R-404A) are considered as the most promising candidates to replace R-22 and R-502. Pentafluoroethane (HFC-125) was tested by the Program for Alternative Fluorocarbon Toxicity Testing (PAFT) and considered to be a nontoxic, nongenotoxic, and nonmutagenic compound.

The relative permittivity of HFC-125 was measured here as a continuation of our research program for the determination of the dielectric properties of some environmentally acceptable refrigerants and their mixtures. We have previously reported relative permittivity data and dipole moments in the liquid state for several pure fluids, such as 1,1-dichloro-1-fluoroethane (HCFC-141b), 1,1,1-trifluoro-2,2-dichloroethane (HFC-123), 1,1-difluoro-1-chloroethane (HCFC-142b), 1,1,1,2-tetrafluoroethane (HFC-134a) [1,2], difluoromethane (HFC-32) [3], and 1,1-difluoroethane (HFC-152a) [4], and for some refrigerant mixtures, namely, R-410A[4] [5], R-404A,[5] R-407C,[6] and R-507[7] [6]. In this work we present accurate experimental results of the relative permittivity of HFC-125 as a function of temperature and pressure in the liquid phase. Using a direct capacitance method, measurements were carried out at 10 kHz, from 214 to 304 K and at pressures from 2 to 16 MPa.

2. EXPERIMENTAL

The experimental technique can be used to perform absolute measurements of relative permittivity, based on a direct capacitance method. The sample handling and the apparatus performance have been described in detail by Gurova et al. [7], while a description of the cell has been presented by Mardolcar et al. [8]. The vacuum capacitance was measured prior to filling the cell with the sample. An impedance analyzer (Schlumberger Model 1260) was used, with an accuracy of 5×10^{-4} pF. This equipment was calibrated by Laboratório de Metrologia Eléctrica da Companhia Portuguesa Radio Marconi, Lisbon, using the standards of capacitance of

[4] HFC-based nearly azeotropic mixture composed of HFC-32/125.

[5] Ternary zeotropic blend composed of HFC-125/143a/134a.

[6] Zero ozone depletion blend composed of HFC-32/125/134a.

[7] Azeotrope of HFC-125 and HFC-143a.

1 pF, 10 pF, 100 pF, 1000 pF, 0.01 μF, 0.1 μF, and 1 μF with an uncertainty of 0.01 %. The technique employed a four-terminal connection to the cell in order to compensate for parasitic impedances. The mean value of a 10-dimensional sample taken at a 10-kHz frequency provides the experimental value of relative permittivity, which proved to be properly suited to the working accuracy. Since our last report the measuring process is now completely automatic and operated from a computer graphics user interface, making the data analysis faster and statistically more significant. The temperature of the cell was measured with a calibrated platinum resistance thermometer (100 Ω at 0°C) located near the sample. The resistance of the Pt-100 thermometer was determined with a four-wire measurement, with a 5.5 digital multimeter (Keithley Model 199 DMM), calibrated with three standard resistors. The sensor was calibrated at four points. The uncertainty of temperature measurement is 0.01 K. The pressure vessel is immersed in a cylindrical copper vessel cooled by a commercial cryostat (Julabo Model FPW90-SC), filled with ethanol, and operative in the range from 183 to 373 K with an uncertainty of 0.1 K.

For the measurements of capacitance under pressure, a high-pressure system was developed, composed of a HIP manual liquid-pressure generator and an air-operated, diaphragm-type compressor (Newport Scientific). The pressure was measured with a pressure transducer from Setra Systems with an uncertainty of 0.01 MPa. The sample of pentafluoroethane was obtained from Elf-Atochem, France, with an estimated purity of greater than 99.5 mass % and an estimated water content of less than 10 ppm. The refrigerant was measured without further purification, and no further purity checks were performed after handling. A schematic diagram of the apparatus for measurements of the relative permittivity in the liquid phase is presented in Fig. 1.

The measurements were performed for a series of 10 isotherms separated by $\approx$10 K, in steps of 1 MPa, from 2 to 16 MPa. The values of the density of HFC-125 were calculated using a 32-term modified Benedict–Webb–Rubin (MBWR) equation of state, proposed by Outcalt and McLinden [9], which represents available experimental data from 174 to 448 K and pressures to 68 MPa, with a maximum deviation of approximately 1.7 % from the experimental values that were used in the determination of the coefficients for the correlation.

The uncertainty of the experimental measurements of the relative permittivity with the present apparatus was found in previous papers to be better than 0.16 % [1, 2, 4, 10], for a confidence interval of 95 %.[8] This calculation

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[8] Here we use the ISO definition of uncertainty, with $k = 2$ (95 % confidence). Using the common calculation for uncertainty (accuracy), the values reported must be divided by two.

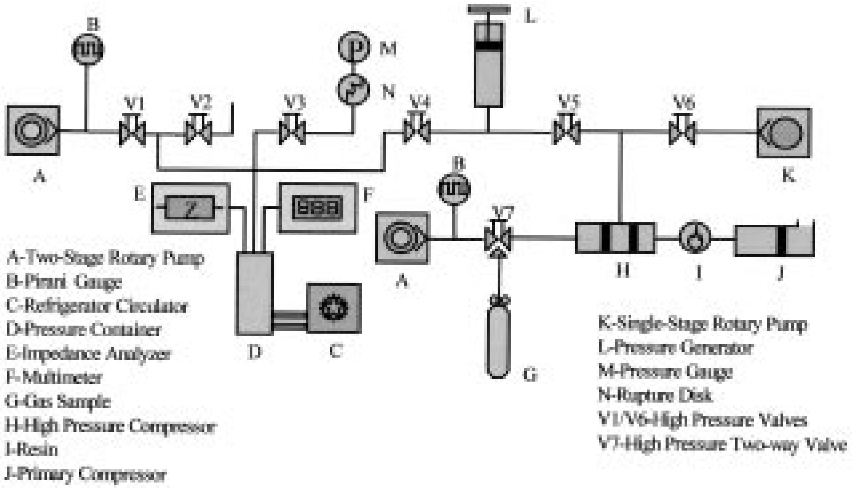

Fig. 1. Schematic diagram of the experimental apparatus.

reflects the uncertainty in the temperature, pressure, and capacitance measurements (in the fluid, including the effect of the hydrostatic pressure in the capacitor components, and in vacuum). Vacuum points were stable at the level of 10^{-4} pF over the duration of this study. The presence of impurities causes an extra source of uncertainty. These impurities, as the water content is less than 10 ppm and the purity of the sample is greater than 99.5 mass %, can amount to a maximum of 0.5 mass %. In the absence of information about the type of compounds that make up the impurities, and their respective relative permittivities for the same temperature and pressure, this problem cannot be solved quantitatively. However, if we assume that the impurities have a relative permittivity that is 40 % different from that of pentafluoropentane, the maximum error possible is of the order of 1 part in 10^3, a value comparable to the repeatability of the measurements (7 parts in 10^4). Using this result in the uncertainty estimate, the uncertainty for the relative permittivity measurements with HFC-125 was estimated to be $\pm 0.72 \times 10^{-2}$.

3. RESULTS AND DISCUSSION

The following relation gives the relative permittivity of the fluid,

$$\varepsilon = \frac{C(p, T)}{C_0(T)} \tag{1}$$

where $C(p, T)$ is the geometric capacitance at pressure p and temperature T, and $C_0(T)$ is the capacitance under vacuum at a temperature T. Table I presents the data obtained as a function of pressure and density for each isotherm for HFC-125. The experimental data of the relative permittivity were fitted by a maximum-likelihood iterative χ^2 method (each iteration implemented by a Levenberg–Marquardt procedure) to a function in density and temperature of the following form (T in K and ρ in $kg \cdot m^{-3}$),

$$\varepsilon(\rho, T) = a_0 + \frac{a_1}{T} + a_2\rho + \frac{a_3\rho}{T} \tag{2}$$

with a standard deviation of 0.08 %.

Table I. Experimental Values of the Relative Permittivity of Pentafluoroethane (HFC-125)[a]

p (MPa)	ρ ($kg \cdot m^{-3}$)	ε	p (MPa)	ρ ($kg \cdot m^{-3}$)	ε	p (MPa)	ρ ($kg \cdot m^{-3}$)	ε
$T_n = 303.74$ K			$T_n = 273.19$ K			$T_n = 243.31$ K		
16.00	1298.24	5.14852	16.00	1405.28	6.26914	16.00	1502.49	7.63996
14.99	1291.85	5.11837	15.00	1401.02	6.24447	15.00	1499.48	7.61840
14.00	1285.30	5.08702	14.00	1396.63	6.21937	14.00	1496.39	7.59681
13.00	1278.39	5.05436	13.00	1392.13	6.19277	13.00	1493.24	7.57477
11.99	1271.08	5.02044	12.00	1387.47	6.16666	12.00	1490.05	7.55295
11.00	1263.55	4.98402	11.00	1382.66	6.13874	11.00	1486.79	7.53001
10.00	1255.47	4.94645	10.00	1377.71	6.11013	10.00	1483.46	7.50717
9.00	1246.91	4.90617	9.00	1372.58	6.08062	9.00	1480.08	7.48382
7.99	1237.75	4.86352	8.00	1367.24	6.04995	8.00	1476.62	7.46020
7.00	1228.03	4.81868	7.00	1361.72	6.01879	7.00	1473.06	7.43593
6.00	1217.47	4.77033	6.00	1355.98	5.98646	6.00	1469.45	7.41098
5.00	1205.91	4.71625	5.00	1349.98	5.95193	5.00	1465.74	7.38610
4.01	1193.26	4.66147	4.00	1343.69	5.91568	4.00	1461.93	7.35937
3.00	1178.91	4.59737	3.00	1337.08	5.87978	3.00	1458.01	7.33261
2.01	1162.67	4.52497	2.00	1330.19	5.84240	2.00	1454.02	7.30559
$T_n = 294.08$ K			$T_n = 263.32$ K			$T_n = 233.19$ K		
16.00	1333.08	5.48165	16.00	1438.08	6.68550	16.00	1534.25	8.19469
15.00	1327.53	5.45345	15.00	1434.29	6.66185	15.00	1531.54	8.17440
14.00	1321.77	5.42441	14.00	1430.39	6.63776	14.00	1528.79	8.15388
13.00	1315.77	5.39408	13.00	1426.41	6.61299	13.00	1525.99	8.13338
12.00	1309.51	5.36303	12.00	1422.32	6.58781	12.00	1523.14	8.11288
11.00	1302.99	5.33044	11.00	1418.12	6.56213	11.00	1520.24	8.09128
10.00	1296.17	5.29617	10.00	1413.78	6.53548	10.00	1517.29	8.06963

[a] T_n is a nominal temperature. All experimental points measured at a given temperature T, close to T_n, were adjusted to this temperature, by using $\varepsilon(T_n, p) = \varepsilon(T, p) + (\partial\varepsilon/\partial T)_p (T_n - T)$.

Table I. *(Continued)*

p (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	ε	p (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	ε	p (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	ε
$T_n = 294.08$ K			$T_n = 263.32$ K			$T_n = 233.19$ K		
9.00	1288.98	5.26028	9.00	1409.34	6.50866	9.00	1514.30	8.04760
8.00	1281.36	5.22241	7.99	1404.73	6.48060	8.00	1511.21	8.02494
7.00	1273.35	5.18247	7.00	1400.01	6.45191	7.00	1508.12	8.00224
6.00	1264.86	5.14094	6.00	1395.15	6.42282	6.00	1504.97	7.97943
5.00	1255.71	5.09609	5.00	1390.11	6.39290	5.00	1501.75	7.95558
4.00	1245.88	5.04838	4.00	1384.87	6.36180	4.00	1498.47	7.93196
3.00	1235.13	4.99649	3.00	1379.42	6.32898	3.00	1495.12	7.90791
2.00	1223.34	4.94095	2.00	1373.77	6.29534	2.00	1491.68	7.88279
$T_n = 283.21$ K			$T_n = 253.29$ K			$T_n = 223.20$ K		
16.00	1371.13	5.87740	16.00	1470.69	7.14283	16.00	1565.11	8.80421
15.00	1366.32	5.85120	15.00	1467.33	7.12118	15.00	1562.67	8.78479
14.00	1361.32	5.82372	14.00	1463.87	7.09862	14.00	1560.19	8.76537
13.00	1356.16	5.79544	13.00	1460.34	7.07545	13.00	1557.65	8.74478
12.00	1350.82	5.76684	12.00	1456.73	7.05170	12.00	1555.08	8.72433
11.00	1345.28	5.73689	11.00	1453.04	7.02735	11.00	1552.49	8.70343
10.00	1339.53	5.70600	10.00	1449.25	7.00258	10.00	1549.87	8.68295
9.00	1333.53	5.67351	9.00	1445.37	6.97736	9.00	1547.24	8.66253
8.00	1327.27	5.64054	8.00	1441.40	6.95149	8.00	1544.55	8.64173
7.00	1320.73	5.60552	7.00	1437.32	6.92499	7.00	1541.81	8.62041
6.00	1313.84	5.56917	6.00	1433.13	6.89847	6.00	1539.04	8.59877
5.00	1305.86	5.53081	5.00	1428.82	6.87060	5.00	1536.20	8.57692
4.00	1298.92	5.49199	4.00	1424.39	6.84253	4.01	1533.36	8.55498
3.00	1290.73	5.44786	3.00	1419.79	6.81299	3.01	1530.44	8.53212
2.00	1281.98	5.40315	2.00	1415.05	6.78240	2.00	1527.43	8.50964
$T_n = 214.32$ K								
16.00	1592.05	9.40676						
15.00	1589.84	9.38960						
14.00	1587.61	9.37041						
13.00	1585.33	9.35174						
12.00	1583.02	9.33209						
11.00	1580.68	9.31215						
10.00	1578.31	9.29232						
9.00	1575.92	9.27243						
8.00	1573.53	9.25279						
7.00	1571.10	9.23317						
6.00	1568.64	9.21291						
5.00	1566.12	9.19204						
4.01	1563.58	9.17107						
3.00	1560.97	9.15001						
2.00	1558.52	9.12981						

Table II. Coefficients of the Dielectric Equations of State [Eqs. (2) and (3)]

a_0	a_1 (K)	$10^{-3}a_2$ ($\mathrm{kg^{-1}\cdot m^3}$)	a_3 ($\mathrm{K\,m^3\cdot kg^{-1}}$)
5.141 ± 0.086	-1866 ± 29	-3.3867 ± 0.0521	2.471 ± 0.017
b_0	b_1 (K)	$10^{-2}b_2$ ($\mathrm{MPa^{-1}}$)	b_3 ($\mathrm{K\cdot MPa^{-1}}$)
-6.364 ± 0.031	3314 ± 7.8	8.908 ± 0.309	-15.426 ± 0.784

For industrial applications the data were also fitted to a function in pressure and temperature according to the equation (p in MPa and T in K)

$$\varepsilon(p, T) = b_0 + \frac{b_1}{T} + b_2 p + \frac{b_3 p}{T} \tag{3}$$

with a standard deviation of 0.21%. The coefficients of Eqs. (2) and (3) with their uncertainties are given in Table II.

An analysis of the experimental data of relative permittivity as a function of density is also presented in this paper. The Vedam formalism was applied based on the work of Vedam et al. [11, 12] and Diguet [13]. According to this theory, the variation of the relative permittivity with pressure is a function of the deformation of the volume, showing a nonlinear behavior in the case of liquids. This nonlinearity can be reduced when the variation of ε, Δ, is analyzed as a function of the Eulerian deformation, Σ, also named the Eulerian strain. It is possible to verify that Σ provides a linear relation for Δ independently of the type of molecules that compose the fluid. We have used the relation between $\varepsilon^{1/2}$ and the Eulerian strain Σ, which is defined, according to the Vedam relation, as

$$\Delta = \varepsilon^{1/2}(\rho) - \varepsilon^{1/2}(\rho_0) = A\Sigma + B \tag{4}$$

$$\Sigma = \frac{1}{2}\left[\left(1 - \frac{\rho}{\rho_0}\right)^{2/3}\right] \tag{5}$$

Here ρ_0 is the reference density, taken in this case as the saturation value for each isotherm. The saturation density data of HFC-125 were calculated using the equation of state provided by Outcalt and McLinden [9].

The calculations show that the function Δ indeed represents a linear variation with the Eulerian strain Σ, as shown in Fig. 2. Table III presents the values of the coefficients A and B of the Vedam equation for each isotherm. The intercept values are close to zero for all isotherms, $B \cong 0$.

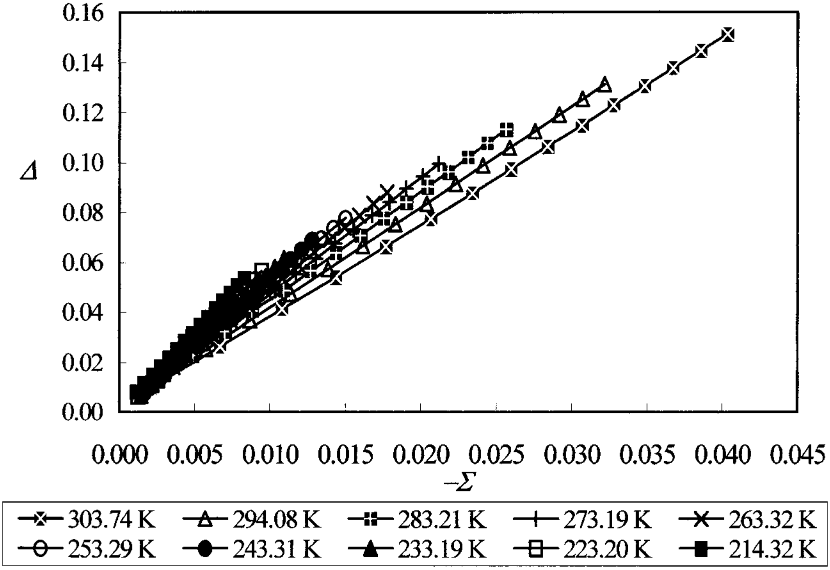

Fig. 2. Variation of Δ with the Eulerian strain, Σ [Eq. (4)].

The slope of the linear variation of Δ with Σ is negative for all temperatures, decreasing linearly with an increase in temperature, as expected from the theory. Assuming that $B = 0$ [Eq. (4)], it is possible to use the Vedam relation to estimate the relative permittivity values. In the present work, as reported before [5, 6, 10, 14], we have estimated the new value of A', by fitting the experimental results as a function of Σ and forcing the constant

Table III. Values of the Constants A and B of the Vedam Equation [Eq. (4)] and of the Constant A' in Eq. (6)

T (K)	ρ_{sat} (kg · m^{-3})	$\varepsilon(\rho_{\text{sat}})$	A	B	A'
214.32	1553.2	9.080	−6.3685	0.00088	−6.5186
223.20	1521.7	8.473	−6.1038	−0.00142	−5.8903
233.19	1485.2	7.846	−5.7912	−0.00184	−5.5538
243.31	1446.7	7.263	−5.5071	−0.00151	−5.3404
253.29	1406.9	6.733	−5.2290	−0.00071	−5.1622
263.32	1364.7	6.238	−4.9207	0.00037	−4.9502
273.19	1320.5	5.781	−4.6442	0.00091	−4.7040
283.21	1272.0	5.342	−4.3302	0.00176	−4.4249
294.08	1214.0	4.884	−4.0091	0.00200	−4.0941
303.74	1155.7	4.484	−3.7114	0.00125	−3.7535

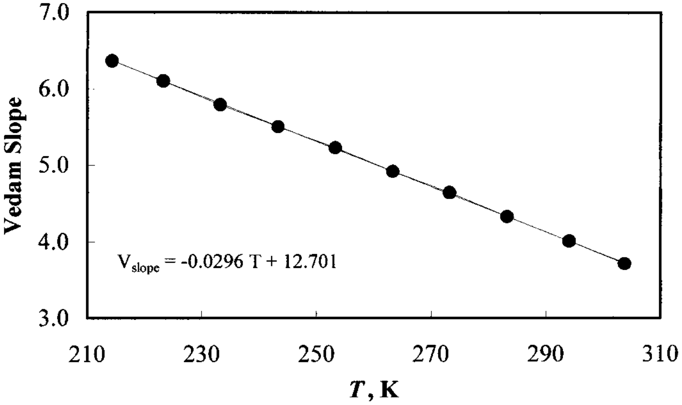

Fig. 3. Graphical representation of the Vedam slope.

B to be equal to zero. In this case the Vedam equation takes the following form:

$$\Delta = A'\Sigma \tag{6}$$

The new values of the slope A' according to Eq. (6) are also presented in Table III and its variation with temperature in Fig. 3. We can estimate the variation of the relative permittivity with density for each isotherm. Fig. 4 presents the deviations between the estimated values and the experimental data of the relative permittivity, calculated according to this method. As can be seen, they are smaller than 0.25%. However, a systematic deviation does exist because of the constrained regression. This systematic difference is never present if we use the full equation [Eq. (4)] for correlating the data. Either equation is very useful for interpolation.

The only true molecular theory that can be applied to the present data, in the absence of data on the refractive index of the liquid, is the theory of molecular polarizability developed by Kirkwood [15] after the definition of Onsager's local field [16]. In this theory, the apparent dipole moment of the liquid μ^* is calculated from the following relation:

$$\frac{(\varepsilon-1)(2\varepsilon+1)}{9\varepsilon}\left(\frac{M}{\rho}\right)=\frac{N_A}{3}\left[\alpha+\frac{(\mu^*)^2}{3\varepsilon_0 k_B T}\right] \tag{7}$$

where M is the relative molar mass of the fluid, N_A is the Avogadro constant, α is the molecular polarizability of the molecule, ε_0 is the electric permittivity in vacuum, T is the absolute temperature, k_B is the Boltzmann constant, and ρ is the density determined using the equation of state of

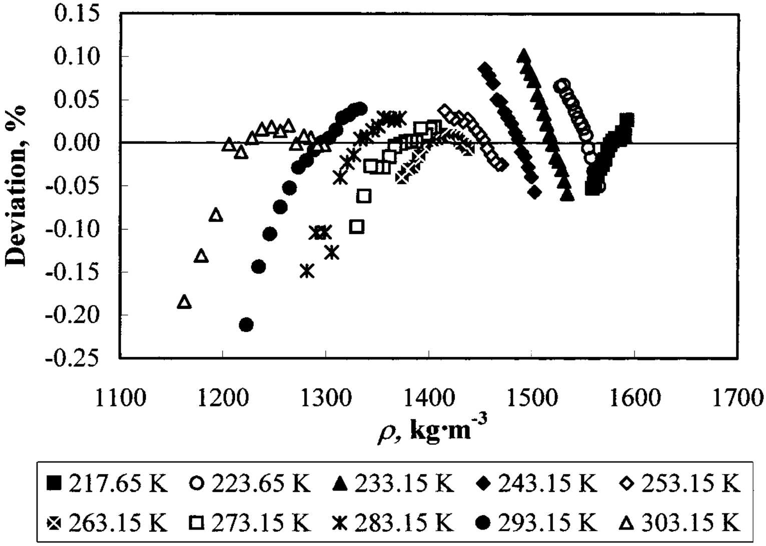

Fig. 4. Deviations of the calculated relative permittivity values from the experimental data $[100(\varepsilon_{\mathrm{VR}}-\varepsilon_{\exp})/\varepsilon_{\exp}]$, according to the Vedam relation [Eq. (6)].

Outcalt and McLinden [9], evaluated at the experimental (T, p) points. The apparent dipole moment is $\mu^* = g^{1/2}\mu$, where μ is the dipole moment in the ideal-gas state and g is the Kirkwood correlation parameter, which represents the restriction to rotation imposed by a cage of molecules surrounding a given molecule. Kirkwood, on the basis of a quasi-crystalline model, defined this parameter g as

$$g=\frac{\mu^{*2}}{\mu^2}=1+\sum_{i=1}^{\infty} z_i\langle\cos\gamma_i\rangle \tag{8}$$

where z_i is the number of neighbors to the central molecule under consideration in the ith coordination shell, and $\langle\cos\gamma_i\rangle$ is the average cosine angle γ formed by the dipole moments of molecules in the ith shell with the dipole of the central molecule. For nonpolar or nonassociated liquids $g\approx 1$, but for polar liquids it may differ considerably from unity. The greater the value of g, the bigger the orientational order imposed by the neighbors.

The value of μ^* can be calculated by a linear regression of the left-hand side of Eq. (7) as a function of $1/T$. The experimental measurements were used to calculate the Kirkwood function, and Fig. 5 shows its variation with $1/T$ for HFC-125, as well as the value of the apparent dipole moment obtained ($\mu^* = 2.482$ D). Using the value of the dipole moment of

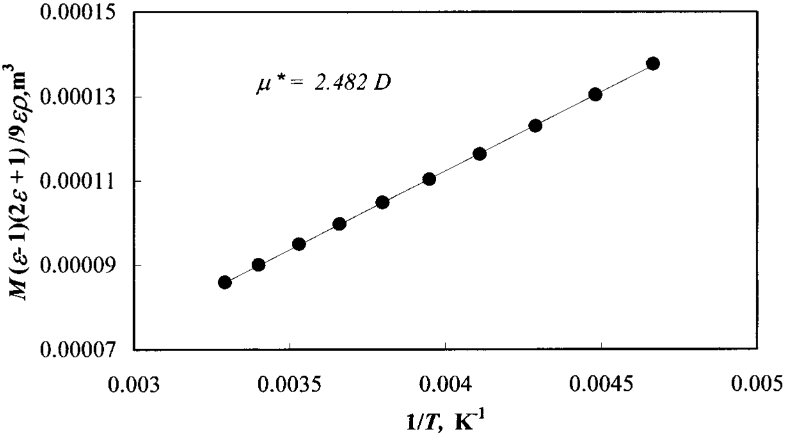

Fig. 5. Kirkwood function vs $1/T$ for HFC-125: (●) HFC-125; (——) linear fit.

HFC-125 ($\mu = 1.563$ D) in the gas phase [17], the value of the Kirkwood parameter g was found to be equal to 2.52.

4. CONCLUSIONS

This work contributes to our knowledge about the behavior of polar fluids in the liquid state. It presents accurate relative permittivity measurements of an environmentally safe refrigerant, pentafluoroethane (HFC-125), in the temperature range from 214 to 304 K at pressures from 2 to 16 MPa. The experimental values were correlated as a function of density and temperature and as a function of pressure and temperature, generating two dielectric equations of state for this fluid. The Eulerian formalism was used to analyze the data, and as in previous studies, it can be concluded that it represents a powerful correlation and interpolation method for the dependence of the relative permittivity on density.

The Kirkwood theory, which allows the evaluation of the value of the apparent dipole moment μ^*, was also applied to calculate the Kirkwood correlation factor, g, found to be 2.52. When compared with the previous study of 1,1,1-trifluoro-2,2-dichloroethane (HFC-123), 1,1-difluoro-1-chloroethane (HCFC-142b), 1,1-dichloro-1-fluoroethane (HCFC-141b), 1,1,1-trifluoroethane (HFC-143a), and 1,1-difluoroethane (HFC-152a) [4], it is possible to conclude that there is some restricted rotation of the molecule in the liquid state.

This conclusion is supported by recent calculations of Costa Cabral et al. [18], who performed density functional and density functional self-consistent reaction field calculations (SCIPCM; self-consistent isodensity polarizable continuum model) for a series of HFC molecules including CH_2FCF_3 (HFC-134a), CHF_2CH_3 (HFC-152a), and CH_3CF_3 (HFC-143a). Particular emphasis has been placed on the calculation of electronic polarizabilities and to the prediction of the dipole moment in a dielectric medium representing the liquid environment. The liquid was assumed to have no structure and a constant relative permittivity. The value obtained for the dipole moment was 1.91 D, a value 22% greater than the gas-phase value but still only 77% of the liquid-phase value determined experimentally. This result was expected, as the model incorporates only electrostatic interactions with a continuum dielectric medium.

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NOMENCLATURE

a_i	Coefficients of Eq. (2)
b_i	Coefficients of Eq. (3)
g	Kirkwood factor
$k_{\mathbf{B}}$	Boltzmann constant
p	Pressure
A, B	Coefficients of Eq. (4)
A'	Coefficient of Eq. (6)
C	Geometric capacitance of the cell filled with the liquid
C_0	Geometric capacitance of the cell in vacuum
$N_{\mathbf{A}}$	Avogadro's number
T	Absolute temperature

Greek Letters

α	Polarizability
χ^2	Iterative method
ε	Relative permittivity (dielectric constant) of the liquid
ε_0	Relative permittivity (dielectric constant) in vacuum
ρ	Density
ρ_0	Reference density
μ	Dipole moment in the gaseous phase
μ^*	Apparent dipole moment in the liquid phase
Δ	Defined by Eq. (4)
Σ	Eulerian strain

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