Viscosity of Compressed Liquid 1,1,1-Trifluoroethane (HFC-143a) and Pentafluoroethane (HFC-125)

Helena M. N. T. Avelino,[†] João M. N. A. Fareleira,* and Carla M. B. P. Oliveira[‡]

Centro de Química Estrutural, Instituto Superior Técnico, Universidade Técnica de Lisboa, Avenida Rovisco Pais, 1, 1049-001 Lisboa, Portugal

---

The viscosity of compressed liquid 1,1,1-trifluoroethane (HFC-143a) and pentafluoroethane (HFC-125) has been measured with a vibrating-wire viscometer at five temperatures between (254 and 293) K. The measurements were performed at pressures from above saturation up to 10 MPa, although for the isotherms at about 254 K the maximum pressure was approximately 5 MPa for HFC-143a and 7.4 MPa for HFC-125. For the isotherm at about 263 K, the highest pressure for HFC-143a was of the order of 7.5 MPa. The overall uncertainty of these results has been estimated to be less than ± 1.0 %. The measurements have been correlated using a scheme based on a hard-spheres model. The root mean square deviation of the experimental results from the correlations for HFC-143a and HFC-125 is ± 0.24 % and ± 0.25 %, respectively. The correlation scheme has been used to perform the small extrapolations of the present data to the saturation line to enable comparison with literature results at saturation pressure.

Introduction

In recent years, particular attention has been paid by many researchers to the replacement, in several industrial applications, of chlorinated hydrocarbons (CFCs) with chlorine-free molecules such as hydrofluorocarbons (HFCs). Although thermophysical property data for these substances should, in principle, be required to accomplish such replacement, a literature survey shows that most of the published thermophysical property data on HFCs concern a limited selection of those fluids, like 1,1,1,2-tetrafluoroethane (HFC-134a) and 1,1-difluoroethane (HFC-152a), while only a few measurements for other refrigerants, such as 1,1,1-trifluoroethane (HFC-143a), can be found, often covering modest ranges of thermodynamic conditions. HFC-143a is mainly used as a component in refrigeration fluid blends R404A [which is a mixture of (HFC-125 + HFC-134a + HFC-143a)] and R507 [which is a mixture of (HFC-125 + HFC-143a)].[1,2] The development of correlation schemes for the viscosity of these mixtures either requires or at least is facilitated by the existence of data for the viscosity of their pure components. However, for HFC-143a, no viscosity measurements could be found in the literature for the compressed liquid region. Moreover, only a limited number of data sets of the viscosity of the saturated liquid could be found in the literature. To contribute to the database of refrigerant fluids, we have measured the viscosities of HFC-143a and HFC-125 at temperatures between (254 and 293) K and at pressures up to 10 MPa.

Experimental Section

Measurement Method and Equipment. The viscosity measurements were performed with a vibrating-wire viscometer, which has been described previously.[3,4] Only the main features of the instrument are mentioned here. The central component of the measuring cell is a tungsten wire, one end of which is clamped in a stainless steel chuck that is supported from the pressure vessel lid. The lower end of the vertically suspended wire is attached to two weights through a system of pulleys that tension the wire. The two weights are mounted coaxially. In the present cell, these two weights have the same volume but are formed from materials having significantly different densities, such that the resulting force is used to apply a vertical tension on the wire. Variations in the buoyancy force acting on the weights are mutually compensated by the pulley system. This mechanical arrangement essentially eliminates variations in the frequency of the wire that arise from changes in the fluid density. This is the main modification introduced in the measuring cell in relation to the previous arrangement.[3,4] A similar device has been used before, namely, by Assael et al.[5] A sinusoidal current of variable frequency is passed through the wire, which is forced to perform transverse oscillations as it is immersed in a magnetic field, created by Nd−Fe−B magnets. The electromotive force induced at the wire by this motion is proportional to the velocity of the oscillations. The viscosity of the fluid surrounding the wire is obtained by analysis of the experimental resonance curve for the oscillations of the wire.[3,4,6]

The thermostatic system was essentially identical to the one described previously.[7,8] Temperature stability during a measurement run was typically within ± 5 mK. The temperature was measured on ITS-90 with a 100-Ω platinum resistance thermometer, calibrated by comparison with a Tinsley 5187L 25-Ω platinum resistance thermometer, which has been calibrated at the National Physical Laboratory, Teddington, England. The thermometer was mounted in the thermostatic bath close to the outside surface of the pressure vessel, and its readings have an estimated uncertainty of ± 0.01 K. Pressures were measured with a pressure transducer, model 206 (0 to 5000 psig), supplied by Setra Systems Inc., with an uncertainty of ± 0.05 MPa. The

---

* Corresponding author. E-mail: j.fareleira@ist.utl.pt.

[†] Also at Departamento de Engenharia Química, Instituto Superior de Engenharia de Lisboa, R. Conselheiro Emídio Navarro, 1-1950-062 Lisboa, Portugal.

[‡] Also at Universidade Aberta, R. da Escola Politécnica, 147, 1269-001, Lisboa, Portugal.

10.1021/je0601356 CCC: $30.25

Table 1. Average Wire Radius, R, Internal Damping Factor, Δ_0, and Wire Density, ρ_s, as Reported by Oliveira,[12] at a Temperature of 298.15 K and Pressure of 0.1 MPa

R	49.13 μm
Δ_0	7.85×10^{-5}
ρ_s	19.23×10^3 kg·m^{-3}

overall uncertainty of the present viscosity measurements is estimated to be less than ± 1.0 %.

Working Equations. The performance of the vibrating-wire instrument is based on a rigorous analysis of the dynamics of the wire immersed in a fluid.[9,10] The viscosity is obtained from the analysis of the resonance curve for the transverse oscillations of the wire obtained experimentally. The relation between the experimental parameters and the hydrodynamic analysis of the wire motion is enabled by an electromechanical model, whose main features have been described elsewhere.[3,4,6] The calculation of viscosity requires, in addition to the density of the fluid, the density of the wire material ρ_s, the wire radius R, and the internal damping, Δ_0, of the wire. The parameter, Δ_0, was obtained from the response of the wire in vacuo,[11] and the density of the wire material was taken to be equal to that reported by Oliveira.[12] The average radius of the wire, R, was obtained from a measurement of the resonance curve when the wire was immersed in methylbenzene at a temperature of 298.15 K and pressure of 0.1 MPa; at this condition, the viscosity and density were obtained from the correlations of Gonçalves et al.[13] and Assael et al.,[14] respectively. The values of R, ρ_s, and Δ_0 at $T = 298.15$ K and $p = 0.1$ MPa are listed in Table 1.

Fluid Samples. 1,1,1-Trifluoroethane (HFC-143a) was supplied by Solvay Fluor und Derivate GmbH, Hannover, Germany, under the trade name Solkane R143a (nominal mass purity specification of 99.5 %), while pentafluoroethane (HFC-125) was supplied by Avantec, Saint-Priest, France (minimum mass purity of 99.5 %). The fluid samples were introduced in the measuring cell without any further treatment through a 65 μm filter (High-Pressure Equipment Company, USA).

Results

The viscosity of compressed liquid HFC-143a and HFC-125 was measured along five isotherms at temperatures between (254 and 293) K. The measurements were performed at pressures from above saturation up to 10 MPa, although for the isotherms at about 254 K the maximum pressure was approximately 5 MPa for HFC-143a and 7.4 MPa for HFC-125. For the isotherm at about 263 K, the highest pressure for HFC-143a was of the order of 7.5 MPa. The results are shown in Table 2 for HFC-143a and in Table 3 for HFC-125. The viscosity measurements were performed at temperatures close to the nominal temperatures, T_n, shown in Tables 2 and 3. The raw viscosity data were subsequently corrected to the corresponding value of T_n through linear interpolation, which did not exceed ± 0.08 K for both fluids.

The density values, required to obtain the viscosity from the working equations, were calculated from the modified Benedict−Webb−Rubin (MBWR) equations of state, published by Outcalt and McLinden.[15,16] According to those authors, the experimental liquid phase pVT data sets used in the correlation have average absolute deviations smaller than 0.05 % for the density of HFC-143a.[15] Regarding HFC-125, Outcalt and McLinden state that all the pVT data used for the correlation have an average absolute deviation of 0.092 % for the density of HFC-125.[16]

Table 2. Experimental Measurements of Viscosity, η, of HFC-143a along Five Isotherms at Nominal Temperatures, T_n, as a Function of Pressure Together with Values of Density, ρ, Determined from the Equation of State Reported by Outcalt and McLinden[15]

T_n / K	p / MPa	ρ / kg·m^{-3}	η / mPa·s	p / MPa	ρ / kg·m^{-3}	η / mPa·s	p / MPa	ρ / kg·m^{-3}	η / mPa·s	p / MPa	ρ / kg·m^{-3}	η / mPa·s	p / MPa	ρ / kg·m^{-3}	η / mPa·s
254.35	1.34	1090.3	0.2054	1.45	1090.8	0.2063	2.07	1092.7	0.2088	4.17	1100.2	0.2145	5.02	1103.1	0.2172
	1.35	1090.5	0.2063	1.96	1092.3	0.2082	3.11	1096.3	0.2114	4.19	1100.3	0.2147	5.05	1103.4	0.2171
	1.35	1090.5	0.2059	1.96	1092.4	0.2080	3.16	1096.5	0.2116	4.21	1100.3	0.2147	5.10	1103.5	0.2176
	1.35	1090.6	0.2062	1.97	1092.4	0.2078	3.17	1096.5	0.2115	4.21	1100.3	0.2147			
	1.36	1090.6	0.2065	2.01	1092.5	0.2080	3.21	1096.6	0.2122	4.94	1102.7	0.2173			
	1.36	1090.7	0.2057	2.01	1092.5	0.2081	3.28	1096.9	0.2120	4.97	1102.9	0.2167			
	1.41	1090.2	0.2060	2.04	1092.7	0.2083	3.32	1097.1	0.2119	5.00	1103.2	0.2176			
263.02	1.30	1063.0	0.1828	1.99	1065.8	0.1855	3.56	1072.6	0.1901	4.91	1078.0	0.1942	7.31	1086.9	0.2019
	1.32	1062.9	0.1828	2.01	1065.9	0.1851	3.59	1072.7	0.1898	4.95	1078.1	0.1939	7.38	1087.2	0.2024
	1.32	1063.0	0.1829	2.02	1065.9	0.1857	3.64	1072.9	0.1906	6.99	1085.9	0.2018	7.39	1087.3	0.2015
	1.33	1063.0	0.1827	2.02	1065.9	0.1859	3.67	1073.0	0.1905	7.09	1086.2	0.2010	7.42	1087.4	0.2011
	1.33	1063.0	0.1826	3.42	1072.4	0.1899	3.67	1073.0	0.1908	7.16	1086.4	0.2015	7.46	1087.5	0.2024
	1.33	1063.0	0.1826	3.43	1072.0	0.1898	4.80	1077.6	0.1943	7.20	1086.6	0.2011	7.46	1087.5	0.2022
	1.97	1065.7	0.1855	3.45	1072.2	0.1898	4.84	1077.8	0.1940	7.23	1086.6	0.2016	7.52	1087.7	0.2017
	1.98	1065.8	0.1857	3.52	1072.4	0.1903	4.87	1077.8	0.1940	7.31	1087.0	0.2013			
273.21	1.42	1029.7	0.1617	1.86	1032.1	0.1629	3.49	1040.3	0.1682	4.96	1047.4	0.1724	7.96	1060.4	0.1805
	1.43	1029.8	0.1619	1.89	1032.1	0.1626	3.50	1040.3	0.1680	5.01	1047.6	0.1720	9.53	1066.8	0.1854
	1.44	1029.8	0.1619	3.45	1040.2	0.1682	4.85	1047.0	0.1724	5.03	1047.6	0.1723	9.54	1066.9	0.1852
	1.44	1029.8	0.1619	3.45	1040.2	0.1681	4.86	1047.0	0.1722	7.78	1059.8	0.1804	9.58	1067.0	0.1856
	1.45	1029.8	0.1619	3.45	1040.2	0.1679	4.88	1047.1	0.1719	7.80	1059.9	0.1805	9.61	1067.1	0.1851
	1.81	1031.8	0.1629	3.46	1040.2	0.1677	4.88	1047.5	0.1724	7.81	1059.9	0.1806	9.77	1067.7	0.1850
	1.82	1031.9	0.1627	3.49	1040.3	0.1678	4.91	1047.2	0.1722	7.90	1060.2	0.1803	9.88	1068.1	0.1853
283.22	1.24	992.9	0.1416	1.57	995.0	0.1429	2.05	998.3	0.1442	5.22	1016.7	0.1543	9.72	1039.0	0.1679
	1.24	992.9	0.1416	1.90	997.4	0.1433	3.70	1008.3	0.1495	5.26	1017.0	0.1536	9.78	1039.2	0.1678
	1.26	993.0	0.1418	1.93	997.6	0.1433	3.70	1008.3	0.1495	5.26	1017.0	0.1540	9.94	1040.0	0.1680
	1.47	994.5	0.1416	1.95	997.7	0.1432	3.72	1008.4	0.1496	7.40	1028.0	0.1615	10.05	1040.4	0.1678
	1.51	994.7	0.1424	1.97	997.8	0.1440	3.85	1009.1	0.1497	7.50	1028.4	0.1610			
	1.53	994.8	0.1424	2.00	998.0	0.1435	5.13	1016.8	0.1537	7.54	1028.7	0.1607			
293.17	1.37	954.3	0.1253	1.85	958.5	0.1268	3.86	973.9	0.1335	4.97	981.6	0.1380	7.62	997.8	0.1453
	1.39	954.4	0.1255	1.86	958.5	0.1266	3.87	973.9	0.1333	5.01	981.8	0.1381	9.74	1009.5	0.1507
	1.39	954.4	0.1255	1.89	958.8	0.1271	3.89	974.1	0.1335	7.45	996.9	0.1448	9.77	1009.7	0.1506
	1.40	954.5	0.1252	1.92	959.0	0.1267	3.89	974.1	0.1335	7.49	997.1	0.1446	9.82	1009.9	0.1507
	1.42	954.7	0.1256	1.94	959.1	0.1270	3.89	974.0	0.1336	7.52	997.1	0.1450	9.88	1010.2	0.1510
	1.43	954.8	0.1258	3.82	973.6	0.1333	4.90	981.0	0.1378	7.55	997.4	0.1449	9.97	1010.7	0.1511
	1.43	954.8	0.1256	3.83	973.7	0.1336	4.92	981.2	0.1377	7.56	997.5	0.1449	10.07	1011.2	0.1517
	1.44	954.8	0.1258	3.85	973.8	0.1337	4.96	981.5	0.1380	7.59	997.4	0.1451			

Table 3. Experimental Measurements of Viscosity, η, of HFC-125 along Five Isotherms at Nominal Temperatures, T_n, as a Function of Pressure Together with Values of Density, ρ, Determined from the Equation of State Reported by Outcalt and McLinden[16]

T_n/K	p/MPa	ρ/kg·m^{-3}	η/mPa·s	p/MPa	ρ/kg·m^{-3}	η/mPa·s	p/MPa	ρ/kg·m^{-3}	η/mPa·s	p/MPa	ρ/kg·m^{-3}	η/mPa·s	p/MPa	ρ/kg·m^{-3}	η/mPa·s
253.99	1.44	1409.5	0.2707	1.46	1409.6	0.2710	3.28	1418.4	0.2789	5.06	1426.4	0.2874	7.36	1436.3	0.2984
	1.45	1409.5	0.2710	1.47	1409.6	0.2711	3.28	1418.2	0.2794	5.10	1426.4	0.2875	7.38	1436.3	0.2982
	1.45	1409.5	0.2710	1.48	1409.7	0.2707	3.28	1418.2	0.2790	5.14	1426.8	0.2875			
	1.46	1409.6	0.2710	3.25	1418.2	0.2794	5.05	1426.4	0.2876	7.35	1436.2	0.2984			
263.23	1.43	1370.8	0.2381	3.05	1380.1	0.2452	5.06	1390.8	0.2535	7.56	1403.1	0.2645	9.82	1413.3	0.2745
	1.44	1370.8	0.2373	3.06	1380.1	0.2453	5.07	1390.8	0.2538	7.61	1403.3	0.2645			
	1.44	1370.8	0.2375	3.07	1380.2	0.2456	5.08	1390.9	0.2538	7.64	1403.4	0.2646			
	1.46	1370.9	0.2382	5.03	1390.6	0.2531	5.11	1391.0	0.2534	9.77	1413.1	0.2743			
	1.48	1371.1	0.2377	5.05	1390.7	0.2529	7.53	1402.9	0.2643	9.80	1413.3	0.2740			
273.08	1.46	1326.8	0.2076	2.95	1337.3	0.2145	5.04	1350.7	0.2224	7.49	1364.9	0.2327	9.69	1376.5	0.2421
	1.48	1326.9	0.2075	2.99	1337.5	0.2145	5.04	1350.7	0.2226	7.51	1365.0	0.2324	9.71	1376.6	0.2416
	1.49	1327.0	0.2074	5.02	1350.5	0.2234	7.48	1364.8	0.2324	7.52	1365.0	0.2326			
	1.50	1327.1	0.2074	5.03	1350.6	0.2229	7.49	1364.9	0.2327	7.53	1365.1	0.2324			
	1.50	1327.1	0.2073	5.03	1350.6	0.2233	7.49	1364.9	0.2324	9.68	1376.5	0.2416			
	2.94	1337.2	0.2147	5.04	1350.7	0.2224	7.49	1364.9	0.2324	9.69	1376.5	0.2419			
283.29	1.55	1277.4	0.1781	3.01	1290.4	0.1850	5.22	1307.9	0.1954	7.59	1324.3	0.2059	10.04	1339.5	0.2155
	1.58	1277.7	0.1789	3.01	1290.4	0.1850	5.22	1307.9	0.1960	7.60	1324.4	0.2064	10.04	1339.5	0.2159
	1.59	1277.8	0.1788	3.03	1290.6	0.1852	5.22	1307.9	0.1953	7.60	1324.4	0.2057	10.05	1339.5	0.2155
	1.60	1277.9	0.1785	3.03	1290.6	0.1852	5.22	1307.9	0.1955	7.60	1324.4	0.2055	10.07	1339.6	0.2155
	1.62	1278.1	0.1783	3.06	1290.9	0.1850	5.23	1307.9	0.1953	7.60	1324.4	0.2059	10.08	1339.7	0.2157
	3.00	1290.3	0.1852	5.19	1307.6	0.1957	5.27	1308.2	0.1956	7.60	1324.4	0.2056			
293.13	2.02	1229.1	0.1577	2.14	1230.5	0.1583	7.53	1281.9	0.1818	10.04	1300.3	0.1918	10.04	1300.3	0.1914
	2.03	1229.2	0.1581	5.08	1261.1	0.1720	7.53	1281.9	0.1813	10.04	1300.3	0.1917	10.05	1300.4	0.1914
	2.07	1229.7	0.1583	5.10	1261.3	0.1716	7.53	1281.9	0.1813	10.03	1300.2	0.1915			
	2.10	1230.0	0.1584	5.13	1261.5	0.1719	7.53	1281.9	0.1811	10.03	1300.2	0.1915			
	2.11	1230.1	0.1583	5.13	1261.5	0.1715	9.99	1300.0	0.1918	10.04	1300.3	0.1916			

Discussion

Correlation. All the measurements of the viscosity of HFC-143a and HFC-125 were correlated with the fluid density using a semiempirical method proposed by Li et al.[17] This technique is a heuristic development of the application of the kinetic theory of a dense hard-sphere fluid to the van der Waals model of a liquid.[18] This correlation technique uses a dimensionless viscosity, η^*, which, using SI units, is defined by[18]

$$\eta^* = 6.035 \times 10^8 \eta V^{2/3}/(MRT)^{1/2} \tag{1}$$

where M is the molar mass, R is the gas constant, T is the temperature, and V is the molar volume. The quantity η^* is assumed to depend on V/V_0 only, where V_0 is a characteristic molar volume that is slightly dependent on temperature, accounting for the finite gradient of the repulsive part of a potential for real molecules. Accordingly, the curves of $\eta^*(T)$ versus $\ln(V/V_0)$ obtained at different temperatures can form one single curve of the type

$$\eta^* = f_\eta\left(\frac{V}{V_0}\right) \tag{2}$$

by horizontal shifts along the $\ln(V/V_0)$ axis, corresponding to the temperature dependence of V_0. This kind of method enables the optimization of an expression of the type of eq 2 dedicated specifically to one fluid or to one set of measurements. The technique has been used to describe, for example, viscosity measurements of compressed liquid 2,2,4-trimethylpentane,[19] HFC-134a,[20] and toluene.[21] The optimization of a specific form of curve of the type of eq 2 was also the adopted method for the development of a reference correlation for the viscosity of toluene.[14] For the present work, as the purpose was to develop accurate correlations of the viscosity of single fluids and following the point of view stated in ref 14, the optimization of the form of the curve was also preferred, rather than utilizing a universal equation, as the one developed by Assael et al.[22]

Table 4. Characteristic Volumes, V_0, for HFC-143a and HFC-125, at the Nominal Temperatures of the Present Viscosity Measurements

HFC-143a		HFC-125	
T_n/K	$10^6\ V_0$/m^3·mol^{-1}	T_n/K	$10^6\ V_0$/m^3·mol^{-1}
254.35	53.20	253.99	58.35
263.01	52.91	263.23	58.09
273.21	52.63	273.08	57.82
283.22	52.38	283.29	57.55
293.17	52.19	293.13	57.30

In the present work, a reference value for V_0 at an arbitrary reference temperature, T_{ref}, was calculated for each of the fluids following a procedure previously applied to refrigerants by Teja et al.[23] Assuming that V_0 is at this temperature equivalent to the volume of close-packing of hard-spheres,[18] it was calculated from the value of the hard-sphere diameter, σ_{HS}:

$$V_0 = ({\sigma_{HS}}^3/2^{0.5})N_A \tag{3}$$

The hard-sphere diameters, σ_{HS}, were calculated from the Lennard−Jones potential parameters, σ_{LJ} and ϵ_{LJ}, published by Takahashi et al.[24] for HFC-143a and by Kiselev et al.[25] for HFC-125 using a correlation proposed by Hammonds and Heyes.[26]

The reference temperature, T_{ref}, for HFC-143a was chosen to be 273.21 K, and for HFC-125, it was 273.08 K. The values of V_0 at each of the other nominal temperatures, T_n, at which the present measurements were performed have been obtained by "horizontal" shifts of the curves of $\eta^*(T_n)$ versus $\ln(V/V_0)$ in order to superimpose each isotherm on the curve of $\eta^*(T_{ref})$ at the reference temperature, T_{ref}. The molar volumes of HFC-143a and of HFC-125 were calculated from the corresponding MBWR equations of state reported by Outcalt and McLinden.[15,16] In Table 4, the values of V_0 at all the nominal temperatures of the present measurements are shown, including the reference temperatures indicated above.

Table 5. Fitting Parameters of Equation 4

	HFC-143a	HFC-125
a/m^3·mol^{-1}	74.58	73.29
b/K^{-1}	-1.344×10^{-2}	-8.566×10^{-2}
c/K^{-2}	1.980×10^{-4}	1.063×10^{-4}

Table 6. Fitting Parameters of Equation 5

	HFC-143a	HFC-125
a_0	2.01358	1.42048
a_1	−0.94793	0.56661
a_2	1.475397	0.24535
a_3	−0.57104	−0.10438
a_4	0.007774	0.01106

The values of V_0 in Table 4 are represented by the empirical relationship

$$V_0 \times 10^6/(\mathrm{m}^3\cdot\mathrm{mol}^{-1}) = a + bT + T^2 \tag{4}$$

The regression coefficients a, b, and c are listed in Table 5 for both fluids studied.

The viscosity of HFC-143a and HFC-125 was correlated with the molar volume by

$$\ln \eta^* = \sum_{i=0}^{4} a_i \left(\frac{V_0}{V - V_0}\right)^{-i} \tag{5}$$

The coefficients a_i, listed in Table 6, were adjusted to best represent the results listed in Tables 2 and 3. The relative deviations of our measurements from the viscosity values calculated from eq 5, with the parameters of Table 6, are shown in Figures 1 and 2 for HFC-143a and HFC-125, respectively. The maximum deviations do not exceed ± 0.52 % for HFC-143a and ± 0.60 % for HFC-125. The corresponding values of the root mean square deviation (rmsd) are ± 0.24 % and ± 0.25 %, respectively, with

$$\mathrm{rmsd} = 100 \times \left[\frac{1}{N}\sum\left(\frac{\eta_{\mathrm{exp},i} - \eta_{\mathrm{calc},i}}{\eta_{\mathrm{calc},i}}\right)^2\right]^{1/2} \tag{6}$$

where $\eta_{\mathrm{exp},i}$ is the result of the ith viscosity measurement, $\eta_{\mathrm{calc},i}$ is the calculated value of the viscosity, and N is the number of experimental data in the set of measurements used to define the correlation equation.

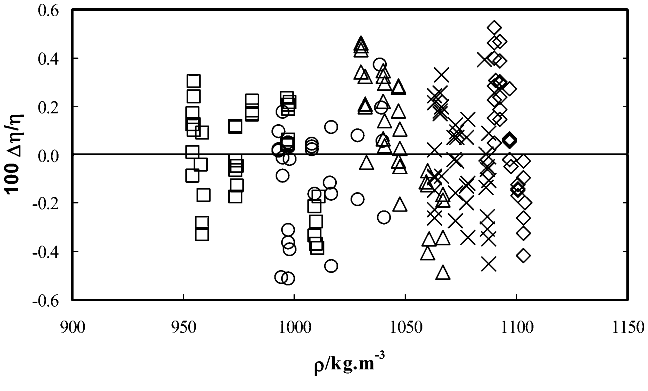

Figure 1. Deviation, $\Delta\eta/\eta = \{\eta(\mathrm{exp}) - \eta(\mathrm{calc})\}/\eta(\mathrm{calc})$, of the viscosity measurements of HFC-143a (Table 2), η(exp), from the correlation eq 5, η(calc), as a function of density, for several isotherms: ◇, 254.35 K; ×, 263.01 K; △, 273.21 K; ○, 283.22 K; □, 293.17 K.

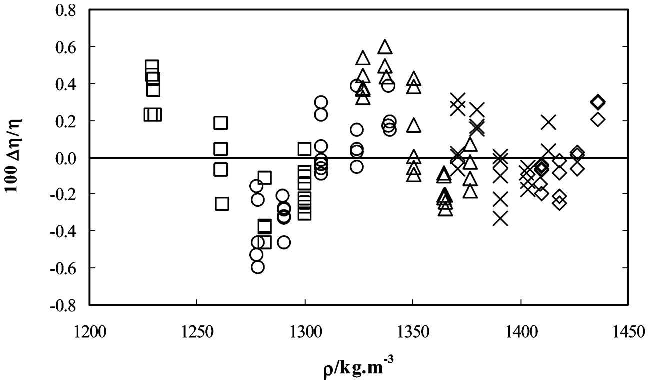

Figure 2. Deviation, $\Delta\eta/\eta = \{\eta(\mathrm{exp}) - \eta(\mathrm{calc})\}/\eta(\mathrm{calc})$, of the viscosity measurements of HFC-125 (Table 3), η(exp), from the correlation eq 5, η(calc), as a function of density, for several isotherms: ◇, 253.99 K; ×, 263.23 K; △, 273.08 K; ○, 283.29 K; □, 293.13 K.

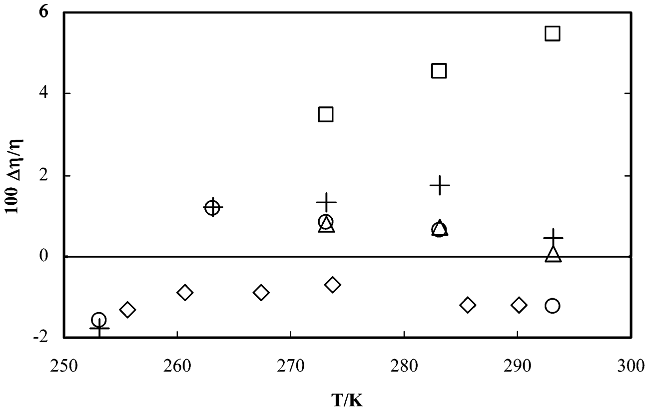

Figure 3. Deviation, $\Delta\eta/\eta = \{\eta(\mathrm{lit}) - \eta(\mathrm{calc})\}/\eta(\mathrm{calc})$, of published viscosity data of HFC-143a, η(lit), along the saturation line, from eq 5, η(calc), as a function of temperature, for the temperature range (250 to 293) K: ◇, ref 28; +, ref 29; △, ref 30; □, ref 27; ○, refs 31 and 32.

Comparison with Literature Data. No results of viscosity measurements have been found by the authors in the literature for HFC-143a in the compressed liquid region. Therefore, the present results for this fluid can only be compared with literature data obtained along the saturation line. This requires a modest extrapolation of the present results to the saturation pressure by means of eq 5, with the parameters given in Table 6. The density at saturation has been obtained from the ancillary equations published by Outcalt and McLinden for HFC-143a and HFC-125.[15,16] For HFC-143a, the maximum extrapolation in terms of the fluid density was smaller than 0.35 %, while for HFC-125, the same quantity did not exceed 0.78 %. The saturated liquid viscosity of HFC-143a reported by Kumagai and Takahashi,[27] Ripple and Defibaugh,[28] and Fröba et al.[29] are shown in Figure 3 as deviations from the correlation of the measurements performed in the present work along their temperature range. The first two sets of data were both obtained using capillary viscometers, while the latter set was measured using a surface light-scattering technique. It should be noted that the data published by Kumagai and Takahashi[27] have been corrected for buoyancy effects by Kumagai and Yokoyama.[30] In Figure 3, the deviations of these corrected values from the present results are also shown. The nominal uncertainty of the measurements by Kumagai and Takahashi[27] has been estimated as ± 0.5 %. It can be observed in Figure 3 that the deviations of the uncorrected set of results amount to a maximum 5.5 %, while after correction the maximum absolute deviation is 0.81 %. It may therefore be concluded that the corrected data agree with the correlation of the present results within the mutual uncertainty of the measurements. The same conclusion may be drawn from the comparison of the data set published by Ripple and Defibaugh,[28] whose measurements have an estimated

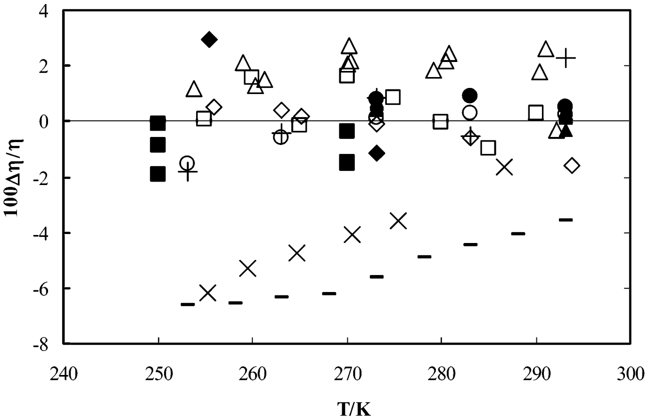

Figure 4. Deviation, $\Delta\eta/\eta = \{\eta(\text{lit}) - \eta(\text{calc})\}/\eta(\text{calc})$, of published viscosity data of HFC-125, η(lit), from eq 5, η(calc), as a function of temperature, for the temperature range (254 to 293) K: □, ref 33 (saturation line data); ◇, ref 36 (saturation line data); ●, ref 34 (saturation line data); ×, ref 37 (saturation line data); △, ref 28 (saturation line data); +, ref 29 (saturation line data); ■, ref 33 (compressed liquid data); ▲, ref 34 (compressed liquid data); ◆, ref 35 (compressed liquid data); −, ref 38; ○, refs 31 and 32.

uncertainty of ± 2.4 %. The results obtained by Fröba et al.[29] have been presented as kinematic viscosity. For comparison purposes, the corresponding dynamic viscosities have been computed using the density data utilized by those authors.[29] The viscosity data reported by Fröba et al.[29] deviate from eq 5, from (−1.8 to +0.4 %). It can therefore be concluded that they agree with the correlation of the present results within the uncertainty of their measurements, as they estimate an uncertainty of ± 2 % for the kinematic viscosity of HFC-143a. It is noteworthy that those measurements[29] have been recalculated by Fröba using a more rigorous description of the liquid−vapor interface.[31,32] The deviations of these new values from the present data are also shown in Figure 3. The differences encountered vary from (−1.6 to 1.2 %), within the uncertainty of the data. Therefore, over the temperature range of our measurements, all the viscosity data, used in the comparisons above, for liquid HFC-143a at saturation agree with the present extrapolated results, the absolute deviations being well within the mutual uncertainty of the experimental methods except for the uncorrected data set by Kumagai and Takahashi.[27]

Viscosity data for HFC-125 are reported in the literature both for the compressed liquid phase and for the liquid under saturation pressure. Both kinds of data are compared with the present results in the temperature range from (250 to 293) K and from saturation pressure to 10 MPa in Figure 4. In this figure the deviations of the literature data from the correlation eq 5 of the present results are shown. The comparison with the literature data under saturation pressure was made performing the necessary extrapolations of the present results as described above. Three viscosity measurement sets have been found of compressed liquid HFC-125, namely those published by Diller and Peterson,[33] by Assael and Polimatidou,[34] and by Wilson et al.[35] The highest pressure attained in those measurements was 50 MPa in the first of those data sets and 17 MPa in the second, but the comparisons of the literature viscosity data of HFC-125 compressed liquid phase with the present results extend up to 10 MPa only. Wilson et al.[35] measured the viscosity of liquid HFC-125 at pressures slightly above saturation. The viscosity of liquid HFC-125 reported by Diller and Peterson[33] differs from the present results, as correlated by eq 5, by less than ± 2 %, which is within the uncertainty of ± 3 %, claimed by those authors, who used a torsional crystal viscometer. It is noteworthy that the results reported by Diller and Peterson[33] at 250 K, lie below the lowest temperature of the present measurements; therefore, the comparison between the two data sets required a small extrapolation of the current data set using eq 5. Within the ranges of temperature and pressure used for the comparisons, the viscosity values of Assael and Polimatidou[34] obtained using a vibrating-wire viscometer with a stated uncertainty of ± 0.5 % differ by less than 0.74 % from eq 5, which is within the mutual uncertainties of the measurement methods. The measurements reported by Wilson et al.[35] of the viscosity of HFC-125, above saturation pressure, with a claimed uncertainty of ± 2 % only include two points within the range of the present measurements, namely, at (255.37 and 273.15) K and at a pressure of 2.8 MPa. The deviations are about 2.9 % and 1.2 %, respectively. The latter is clearly in agreement with our own measurements, and the former seems to be close to the mutual uncertainty of the methods. A good agreement has been found between the results reported by Ripple and Defibaugh[28] along the saturation line, obtained using a capillary viscometer, with a nominal uncertainty of ± 2.4 % and the present measurements, as their deviations from eq 5 do not exceed ±1.6 %. The same kind of agreement has been found for the results obtained with a capillary viscometer reported by Ripple and Matar,[36] as they differ from eq 5 less than ± 3 % within their nominal uncertainty limits, which go from (± 3 to ± 5 %). Fröba et al.[29] have reported kinematic viscosity data for HFC-125 along the saturation line, using a surface light-scattering technique, with a claimed uncertainty of ± 2 %. To compare with the present results, the density values indicated by those authors have been used to compute the corresponding dynamic viscosity. The deviations encountered from correlation eq 5 vary from (−1.8 to +2.3 %), which is commensurate with the mutual uncertainty of the experimental methods. The results of these measurements[26] have been recalculated by Fröba,[31,32] using a more rigorous description of the liquid−vapor interface. The deviations of these new values from the present data are also compared in Figure 4. The differences encountered vary from (−1.5 to +0.3 %), well within the uncertainty of the data. The values documented by Oliveira and Wakeham[37] obtained with a vibrating-wire viscometer with a nominal uncertainty of ± 0.6 % deviate from eq 5 by as much as −5.9 %, which largely exceeds the mutual absolute uncertainty of the measurement sets. Sun et al.[38] have measured the viscosity of liquid HFC-125 at saturation pressure using a capillary device with a claimed uncertainty of ± 3 %. All those data show negative deviations from the correlation of the present results, from (−6.6 to −3.6 %) most of them outside the mutual uncertainty of the measurements. Among the data sets for HFC-125 subjected to comparison with the present data, only the last two, from Oliveira and Wakeham[37] and Sun et al.,[38] are not commensurate with the mutual uncertainty of the measurements. However, it is noteworthy that Assael and Polimatidou[34] have already called the attention and suggested a possible explanation, for the large differences encountered between the viscosity of liquid HFC-125 as measured by Oliveira and Wakeham[37] and other literature data.

Conclusions

New viscosity measurements of compressed liquid HCF-143a and HFC-125 in the temperature range (254 to 293) K and pressures up to 10 MPa have been presented. To the knowledge of the authors, the present results for the former fluid are the first set of viscosity data of compressed liquid HFC-143a. The overall uncertainty of the results obtained using a vibrating-wire instrument is estimated not to exceed ± 1.0 %. The measurements performed on HFC-125 were primarily aimed at testing the equipment, which suffered some minor changes in relation to a previous version as described above. Good agreement with most of the literature values for HFC-125 was found, including the majority of the viscosity measurements of the compressed liquid phase. A good general agreement has also been verified with the HFC-143a saturated liquid viscosity. The experimental results for HFC-125 and HFC-143a were fitted successfully with a correlation scheme based on a hard-spheres model, which was used for extrapolating the present data to the saturation line to enable comparisons with literature data at the saturation pressure.

Acknowledgment

The authors are grateful to Prof. U. V. Mardolcar (Department of Physics, Instituto Superior Técnico, Lisbon, Portugal) and Solvay-Fluor und Derivate GmbH, Germany, for having kindly supplied the sample of HFC-143a. The authors also thank Dr.-Ing. A. P. Fröba (Lehrstuhl für Technische Thermodynamik, Friedrich-Alexander-Universität, Erlangen-Nürnberg, Germany) for calling their attention to the existence of corrected data sets on the viscosity of HFC-125 and HFC-143a, obtained by the surface light scattering method.

Literature Cited

(1) Srinivasan, K.; Oellrich, L. R. Saturation properties of the refrigerant R143a. *Int. J. Refrig.* **1997**, *20*, 332−338.
(2) Fröba, A. P.; Will, S.; Leipertz, A. Thermophysical properties of R143a (1,1,1-trifluoroethane). *Int. J. Refrig.* **2001**, *24*, 734−743.
(3) Correia da Mata, J. L. G.; Fareleira, J. M. N. A.; Oliveira, C. M. B. P.; Wakeham, W. A.; Caetano, F. J. P. A new instrument to perform simultaneous measurements of density and viscosity of fluids by a dual vibrating-wire technique. *High Temp.−High Pressures* **2001**, *33*, 669−676.
(4) Caetano, F. J. P.; Correia da Mata, J. L. G.; Fareleira, J. M. N. A.; Oliveira, C. M. B. P.; Wakeham, W. A. Viscosity measurements of liquid toluene at low temperatures using a dual vibrating-wire technique. *Int. J. Thermophys.* **2004**, *25*, 1−11.
(5) Assael, M. J.; Oliveira, C. P.; Papadaki, M.; Wakeham, W. A. Vibrating-wire viscometers for liquids at high-pressures *Int. J. Thermophys.* **1992**, *13*, 593−615.
(6) Correia da Mata, J. L. G. Desenvolvimento de técnicas e instrumentação para a medida simultânea de densidade e viscosidade de líquidos. Ph.D. Thesis, Universidade Técnica de Lisboa, IST, Lisboa, 2004.
(7) Pádua, A. A. H.; Fareleira, J. M. N. A.; Calado, J. C. G.; Wakeham, W. A. Validation of an accurate vibrating-wire densimeter: density and viscosity of liquids over wide ranges of temperature and pressure. *Int. J. Thermophys.* **1996**, *17*, 781−802.
(8) Pádua, A. A. H. Densidade de líquidos a altas pressões. Ph.D. Thesis, Universidade Técnica de Lisboa, IST, Lisboa, 1994.
(9) Retsina, T.; Richardson, S. M.; Wakeham, W. A. The theory of a vibrating-rod densimeter. *Appl. Sci. Res.* **1986**, *43*, 127−158.
(10) Richardson, S. M. *Fluid Mechanics*; Hemisphere: New York, 1989.
(11) Pádua, A. A. H.; Fareleira, J. M. N. A.; Calado, J. C. G.; Wakeham, W. A. Electromechanical model for vibrating-wire instruments. *Rev. Sci. Instrum.* **1998**, *69*, 2392−2399.
(12) Oliveira, C. M. B. P. Viscosity of liquid hydrocarbons at high pressure. Ph.D. Thesis, Imperial College of Science, Technology and Medicine, London, 1991.
(13) Gonçalves, F. A.; Hamano, K.; Sengers, J. V.; Kestin, J. Viscosity of toluene in the temperature range 25−75 °C. *Int. J. Thermophys.* **1987**, *8*, 641−647.
(14) Assael, M. J.; Avelino, H. M. N. T.; Dalaouti, N. K.; Fareleira, J. M. N. A.; Harris, K. R. Reference correlation for the viscosity of liquid toluene from 213 to 373 K at pressures to 250 MPa. *Int. J. Thermophys*, **2001**, *22*, 789−799.
(15) Outcalt, S. L.; McLinden, M. O. An equation of state for the thermodynamic properties of R143a (1,1,1-trifluoroethane). *Int. J. Thermophys*. **1997**, *18*, 1445−1463.
(16) Outcalt, S. L.; McLinden, M. O. Equations of state for the thermodynamic properties of R32 (difluoromethane) and R125 (pentafluoroethane). *Int. J. Thermophys*. **1995**, *16*, 79−89.
(17) Li, S. F. Y.; Trengove, R. D.; Wakeham, W. A.; Zalaf, M. The Transport Coeficients of Polyatomic Liquids. *Int. J. Thermophys* **1986**, *7*, 273−284.
(18) Dymond, J. H. Hard-Sphere Theories of Transport-Properties. *Chem. Soc. Rev.* **1985**, *14*, 317−356.
(19) Pádua, A. A. H.; Fareleira, J. M. N. A.; Calado, J. C. G.; Wakeham, W. A. Density and Viscosity Measurements of 2,2,4-Trimethylpentane (Isooctane) from 198 K to 348 K and up to 100 MPa. *J. Chem. Eng. Data* **1996**, *41*, 1488−1494.
(20) Pádua, A. A. H.; Fareleira, J. M. N. A.; Calado, J. C. G.; Wakeham, W. A. Density and viscosity measurements of 1,1,1,2-tetrafluoroethane (HFC-134a) from 199 K to 298 K and up to 100 MPa. *J. Chem. Eng. Data* **1996**, *41*, 731−735.
(21) Avelino, H. M. N. T.; Fareleira, J. M. N. A.; Wakeham, W. A. Simultaneous measurement of the density and viscosity of compressed liquid toluene. *Int. J. Thermophys*, **2003**, *24*, 323−336.
(22) Assael, M. J.; Dymond, J. H.; Polimatidou, S. K. Correlation and prediction of dense fluid transport coefficients. VII. Refrigerants. *Int. J. Thermophys.* **1995**, *16*, 761−772.
(23) Teja, A. S.; Smith, R. L.; King, R. K.; Sun, T. F. Correlation and prediction of the transport properties of refrigerants using two modified rough hard-sphere models. *Int. J. Thermophys.* **1999**, *20*, 149−161.
(24) Takahashi, M.; Shibasaki-Kitakawa, N.; Yokoyama, C. Viscosity of gaseous HFC-143a (1,1,1-trifluoroethane) under hugh pressures. *Int. J. Thermophys.* **1999**, *20*, 435−443.
(25) Kiselev, S. B.; Perkins; R. A.; Huber, M. L. Transport properties of refrigerants R32, R125, R134a, and R125 + R32 mixtures in and beyond the critical region. *Int. J. Refrig.* **1999**, *22*, 509−520.
(26) Hammonds, K. D.; Heyes, D. M. Transport-coeficients of model simple liquids. molecular-dynamics study and effective hard-sphere. *J. Chem. Soc., Faraday Trans*. **1988**, *84*, 705−725.
(27) Kumagai, A.; Takahashi, S. Viscosity of saturated liquid fluorocarbon refrigerants from 273 to 353 K. *Int. J. Thermophys*. **1991**, *12*, 105−117.
(28) Ripple, D.; Defibaugh, D. Viscosity of the saturated liquid phase of three fluorinated ethanes: R152a, R143a, and R125. *J. Chem. Eng. Data* **1997**, *42*, 360−364.
(29) Fröba, A. P.; Will, S.; Leipertz, A. Saturated liquid viscosity and surface tension of alternative refrigerants. *Int. J. Thermophys.* **2000**, *21*, 1225−1253.
(30) Kumagai, A.; Yokoyama, C. Revised viscosities of saturated liquid halocarbon refrigerants from 273 to 353K. *Int. J. Thermophys.* **2000**, *21*, 909−912.
(31) Fröba, A. P. Personal communication, 2006.
(32) Fröba, A. P. Simultane bestimmung von viskosität und oberflächenspannung transparenter fluide mittels oberflächenlichtstreuung. Ph.D. Thesis, Friedrich-Alexander-Universität, Erlangen-Nürnberg, 2002.
(33) Diller, D. E.; Peterson, S. M. Measurements of the viscosities of saturated and compressed fluid 1-chloro-1,2,2,2-tetrafluoroethane (R124) and pentafluoroethane (R125) at temperatures between 120 and 420 K. *Int. J. Thermophys.* **1993**, *14*, 55−66.
(34) Assael, M. J.; Polimatidou, S. K. Measurements of the viscosity of liquid R22, R124, and R125 in the temperature range 273−333 K at pressures up to 17 MPa. *Int. J. Thermophys.* **1994**, *15*, 779−790.
(35) Wilson, L. C.; Wilding, W. V.; Wilson, G. M.; Rowley, R. L.; Felix, R. L.; Chilsolm-Carter, T. Thermophysical properties of HFC-125. *Fluid Phase Equilib.* **1992**, *80*, 167−177.
(36) Ripple, D.; Matar, O. Viscosity of the saturated liquid phase of six halogenated compounds and three mixtures. *J. Chem. Eng. Data* **1993**, *38*, 560−564.
(37) Oliveira, C. M. B. P.; Wakeham, W. A. The viscosity of R32 and R125 at saturation. *Int. J. Thermophys.* **1993**, *14*, 1131−1143.
(38) Sun, L.-Q.; Zhu, M.-S.; Han, L.-Z.; Lin, Z.-Z. Viscosity of difluoromethane and pentafluoroethane along the saturation line. *J. Chem. Eng. Data* **1996**, *41*, 292−296.

Received for review March 23, 2006. Accepted May 30, 2006. The present work has been supported by Fundação para a Ciência e a Tecnologia (Portugal) under PRAXIS and POCTI Programmes, with European Union funds (FEDER) and National funds.

JE0601356