

Viscosity Measurements of Compressed Liquid Refrigerant Blend R-507A, Using a Vibrating-Wire Technique

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The refrigerant blend R-507A (50 wt % HFC-143a, 50 wt % HFC-125) is an azeotropic mixture of hydrofluorocarbon refrigerants, 1,1,1-trifluoroethane (HFC-143a) and pentafluoroethane (HFC-125). The paper reports viscosity measurements, performed with a vibrating-wire viscometer, of the refrigerant blend R-507A, at five temperatures in the range (253 to 293) K. The measurements were carried out at pressures from slightly above saturation up to 10 MPa, except for the isotherms at 253.26 K where the maximum pressure was 7.52 MPa and at 263.23 K where the maximum pressure was 7.09 MPa. The overall uncertainty of these measurements is estimated to be $\pm 1.0\%$. The data obtained were correlated by means of a modified hard-sphere based correlation technique. The root-mean-square deviation, rmsd, of the experimental results from the correlation equations is 0.23 %, and their bias is not significant. This correlation method has also been used to interpolate and extrapolate the present results to enable comparisons with measurements performed by other authors of the viscosity of liquid R-507A at different temperatures and pressures.

Introduction

The refrigerant blend R-507A is a mixture of hydrofluorocarbon refrigerants, 1,1,1-trifluoroethane (HFC-143a) and pentafluoroethane (HFC-125) (mass fraction 50 % HFC-143a and 50 % HFC-125). This azeotropic mixture is reported to be nonflammable and to have no ozone depletion potential¹ and seems to be a candidate fluid for several applications, namely as a replacement for R-502.¹

Although some of the new refrigerants in practical use are mixtures, experimental viscosity results on compressed liquid refrigerant mixtures are scarce in the literature.² Naturally, this lack of data is itself hindering the assessment of estimation methods. Furthermore, the existence of experimental viscosity data on the pure components is certainly an important piece of information regarding the development of estimation techniques for the viscosity of mixtures. Concerning the blend R-507A, it is therefore noteworthy that, to the knowledge of the authors, only recently has viscosity data on compressed liquid HFC-143a been made available in the literature.³

The present measurements of viscosity of the compressed liquid blend R-507A were performed with a vibrating-wire viscometer, at five temperatures in the range (253 to 293) K and pressures up to 10 MPa. The results were correlated with the molar volume, as given by the equation of state proposed by Lemmon.⁴ A previously described correlation scheme based on a hard-sphere theory^{5,6} was used for that purpose. The present results were compared with data obtained at thermodynamic saturation conditions by other authors, using the above correlation to perform the small extrapolations of the present data to the saturation pressure. It should be pointed out that viscosity

measurements on compressed liquid R-507A have been reported by another author.⁷ These results were obtained for five isotherms, over a temperature range of (300 to 421) K, and at pressures up to about 68 MPa.⁷ As no overlapping exists of the temperatures at which those data were obtained with the present measurements, a comparison could only be made by resorting to an extrapolation by means of the mentioned correlation scheme.

An oral description of part of the present work has been presented at the 16th Symposium on Thermophysical Properties.⁸

Experimental Section

Measurement Method and Equipment. The viscosity measurements were performed with a vibrating-wire viscometer, which has been described previously.^{3,9,10} The measurement method and the ancillary equipment were identical to those used for measuring the viscosity of compressed liquid 1,1,1-trifluoroethane (HFC-143a) and pentafluoroethane (HFC-125).³ Temperature stability during a measurement run was typically within ± 5 mK, and the pressure was measured with an estimated uncertainty of ± 0.05 MPa. The overall uncertainty of the present viscosity measurements is estimated to be $\pm 1.0\%$.

The calculation of viscosity requires, in addition to the density of the fluid, knowledge of the density of the wire material ρ_s , the wire radius R , and the internal damping, Δ_0 , of the wire. The latter parameters were the same as indicated in our previous publication.³

Fluid Samples. The blend R-507A was supplied by Solvay Fluor und Derivate GmbH, under the trade name Solkane. According to the supplier, the mixture, with a fixed composition, has a stated mass purity of 99.5 %. The fluid sample was introduced, from the liquid phase, in the measuring cell, without any further treatment, through a 65 μm filter (High Pressure Equipment Company, USA).

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Table 1. Experimental Measurements of Viscosity, η , of R-507A along Five Isotherms at Nominal Temperatures, T_n , as a Function of Pressure, p , Together with Values of Density, ρ , Determined from the Equation of State Reported by Lemmon⁴

T_n	p	ρ	η	p	ρ	η
K	MPa	kg·m ⁻³	mPa·s	MPa	kg·m ⁻³	mPa·s
253.26	2.00	1236.3	0.2370	4.93	1248.2	0.2479
	2.01	1236.4	0.2374	4.96	1248.3	0.2479
	2.09	1236.7	0.2377	4.97	1248.3	0.2480
	2.17	1237.0	0.2378	4.99	1248.4	0.2482
	2.18	1237.1	0.2371	5.02	1248.5	0.2483
	2.22	1237.2	0.2379	5.07	1248.7	0.2487
	3.37	1242.0	0.2426	7.44	1257.5	0.2574
	3.38	1242.0	0.2427	7.50	1257.7	0.2576
	3.39	1242.1	0.2427	7.62	1258.1	0.2580
	3.41	1242.2	0.2425	7.72	1258.5	0.2587
	4.90	1248.0	0.2482			
263.23	1.99	1201.6	0.2076	3.72	1210.0	0.2140
	2.02	1201.7	0.2074	3.78	1210.3	0.2140
	2.07	1202.0	0.2081	4.69	1214.4	0.2177
	2.12	1202.2	0.2077	4.99	1215.8	0.2192
	3.66	1209.7	0.2139	5.01	1215.9	0.2195
	3.67	1209.7	0.2137	7.00	1224.5	0.2258
	3.67	1209.7	0.2139	7.05	1224.7	0.2257
	3.68	1209.8	0.2138	7.19	1225.3	0.2262
	3.68	1209.8	0.2137	7.22	1225.4	0.2270
	2.15	1165.5	0.1827	3.50	1173.4	0.1878
	2.16	1165.6	0.1825	5.03	1181.7	0.1923
273.24	2.19	1165.7	0.1826	5.04	1181.7	0.1925
	2.19	1165.7	0.1823	5.06	1181.9	0.1925
	2.23	1166.0	0.1829	7.36	1193.4	0.2011
	2.26	1166.2	0.1829	7.45	1193.8	0.2008
	2.27	1166.2	0.1829	7.45	1193.8	0.2008
	3.45	1173.1	0.1878	7.52	1194.1	0.2015
	3.45	1173.1	0.1876	9.71	1204.1	0.2103
	3.47	1173.2	0.1877	9.88	1204.9	0.2106
	3.48	1173.3	0.1873	9.88	1204.9	0.2104
	2.00	1124.1	0.1592	3.86	1137.4	0.1655
	2.06	1124.6	0.1592	3.88	1137.5	0.1657
283.34	2.12	1125.0	0.1591	4.97	1144.6	0.1693
	2.16	1125.3	0.1591	5.00	1144.8	0.1693
	2.22	1125.8	0.1593	5.03	1145.0	0.1696
	2.23	1125.9	0.1592	5.04	1145.1	0.1695
	2.23	1125.9	0.1596	7.60	1160.2	0.1780
	2.25	1126.0	0.1596	7.64	1160.4	0.1783
	2.26	1126.1	0.1595	7.68	1160.6	0.1784
	2.27	1126.2	0.1597	9.75	1171.6	0.1851
	3.72	1136.5	0.1650	9.86	1172.2	0.1854
	3.77	1136.8	0.1651	9.90	1172.4	0.1857
	3.77	1136.8	0.1654	9.92	1172.5	0.1857
293.33	3.82	1137.1	0.1654	9.93	1172.5	0.1856
	1.92	1079.1	0.1394	5.00	1105.9	0.1506
	1.94	1079.3	0.1392	5.03	1106.1	0.1506
	1.97	1079.6	0.1396	7.51	1123.8	0.1585
	2.02	1080.1	0.1394	7.56	1124.2	0.1584
	3.57	1094.3	0.1450	7.54	1124.0	0.1584
	3.69	1095.3	0.1454	9.77	1137.9	0.1667
	3.72	1095.3	0.1455	9.81	1138.2	0.1669
	3.75	1095.8	0.1457	9.85	1138.4	0.1666
	4.91	1105.2	0.1500	9.88	1138.6	0.1669
	4.91	1105.2	0.1503	9.93	1138.9	0.1672

Results

The viscosity of compressed liquid R-507A was measured along five isotherms at temperatures between (253 and 293) K. The measurements were performed at pressures from slightly above saturation up to 10 MPa, except for the isotherms at 253.26 K where the maximum pressure was 7.52 MPa and at 263.23 K where the maximum pressure was 7.09 MPa. The viscosity measurements were performed at temperatures close to the nominal temperatures, T_n , shown in Table 1. The raw viscosity data were subsequently corrected to the corresponding value of T_n through linear interpolation, which did not exceed 0.02 K. The density values, required to obtain the viscosity from

the working equations, were calculated using the equation of state published by Lemmon.⁴ According to the author, the deviations of the pseudopure fluid equation of state from the full mixture model¹¹ do not exceed ± 0.1 %, except in the critical region.⁴

Discussion

Data Correlation. All the measurements of the viscosity of R-507A were correlated with the fluid density using the same method that has been applied to its pure components,³ HFC-143a and HFC-125. The correlation 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 liquid⁶ and has been proposed by Li et al.⁵ This correlation uses a reduced dimensionless viscosity, η^* , which, using base SI units, is defined by

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

where η is the viscosity; M is the molar mass; T is the temperature; V is the molar volume; and R is the gas constant. Using the procedure described elsewhere,³ the data are correlated by a single equation of the form

$$\ln \eta^* = \sum_{j=0}^n a_j \left(\frac{V_0}{V - V_0} \right)^{-j} \quad (2)$$

where V_0 is a slightly temperature-dependent characteristic volume and $n = 4$ for the present results. The choice of the value for n has been made from the observation that the utilization of a higher number of parameters in eq 2 did not produce significantly smaller values for the statistical parameters AAD, rmsd, or bias. This method enables the optimization of an expression of the type of eq 2, dedicated specifically to one fluid or to one set of measurements, and it has been applied before to several liquids (2,2,4-trimethylpentane,¹² HFC-134a,¹³ and toluene^{14,15}).

In the present work, the blend R-507A was treated as a pseudopure fluid. Hence a reference value for V_0 , at an arbitrary reference temperature, T_{ref} , was calculated, following a procedure previously applied to refrigerants by Teja et al.¹⁶ Assuming that V_0 is, at this temperature, equivalent to the volume of close-packing of hard spheres,⁶ it was calculated from the value of the hard-sphere diameter, σ_{HS}

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

In eq 3, σ_{HS} is the hard-sphere diameter and N_A is Avogadro's constant. The diameter, σ_{HS} , for R-507A was obtained from the Lennard-Jones parameters, σ_{LJ} and ϵ_{LJ} , reported by Nabizadeh and Mayinger,¹⁷ making use of a correlation proposed by Hammonds and Heyes.¹⁸

The reference temperature for R-507A was chosen to be 273.24 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 as described before.³ The molar volumes of R-507A were calculated from the equation of state developed by Lemmon.⁴ In Table 2, the values of V_0 at all the nominal temperatures of the present measurements are shown, including the reference temperature.

The values of V_0 in Table 2 were calculated as described before³ and are represented by the empirical relation

$$V_0 \cdot 10^6 / (\text{m}^3 \cdot \text{mol}^{-1}) = a + b(T/\text{K}) + c(T/\text{K})^2 \quad (4)$$

The fitting parameters a , b , and c are given in Table 3.

Table 2. Characteristic Volumes, V_0 , for R-507A at the Nominal Temperatures of the Present Viscosity Measurements

R-507A	
T_i/K	$10^6 V_0 \text{ m}^3 \cdot \text{mol}^{-1}$
254.26	51.80
263.23	51.58
273.24	51.35
283.34	51.07
293.33	50.77

Table 3. Fitting Parameters of Equation 4

R-507A	
a	47.02
b	$5.747 \cdot 10^{-2}$
c	$-1.524 \cdot 10^{-4}$

Table 4. Fitting Parameters of Equation 2 and the Corresponding Average Absolute Deviation, AAD, Bias, and Root-Mean-Square Deviation, rmsd, of the Present Data from Equation 2, Calculated According to Equations 5 to 7

R-507A	
a_0	1.73484
a_1	0.20853
a_2	0.19403
a_3	0.12037
a_4	-0.05353
AAD/%	0.19
bias/%	0.00
rmsd/%	0.23

The coefficients, a_i , of eq 2 that best represent the present results of the viscosity measurements of R-507A, using the values of V_0 shown in Table 2, are listed in Table 4.

The relative deviations of our measurements from eq 2 are shown in Figure 1. The maximum deviation does not exceed ± 0.56 %; the corresponding average absolute deviation, AAD, of the experimental points from the fitting is 0.19 %; and their bias is not significant (being several orders of magnitude smaller than the AAD). The root-mean-square deviation, rmsd, of the experimental results from the correlations is 0.23 %. The quantities AAD, bias, and rmsd used were defined as follows:

$$\text{AAD}/\% = \frac{100}{N} \sum_i^N \left| \frac{\eta_{\text{expt},i}}{\eta_{\text{calcd},i}} - 1 \right| \quad (5)$$

$$\text{bias}/\% = \frac{100}{N} \sum_i^N \left(\frac{\eta_{\text{expt},i}}{\eta_{\text{calcd},i}} - 1 \right) \quad (6)$$

$$\text{rmsd}/\% = 100 \left[\frac{1}{N} \sum_i^N \left(\frac{\eta_{\text{expt},i}}{\eta_{\text{corr},i}} - 1 \right)^2 \right]^{1/2} \quad (7)$$

where $\eta_{\text{expt},i}$ is the i th viscosity experimental datum; $\eta_{\text{corr},i}$ is the corresponding value of the viscosity calculated by means of the correlation eq 2; and N is the number of experimental data in the set of measurements used to define the correlation equation.

Comparison with Literature Data. For comparison purposes, the only viscosity measurements on compressed liquid R-507A that could be found in the literature have been reported by Laesecke.⁷ Those data were obtained using a torsional crystal viscometer, along five isotherms, over a temperature range of approximately (300 to 421) K and at pressures up to about 68 MPa. The uncertainty of such measurements is, in general, estimated⁷ not to exceed ± 2 %. As the temperature ranges of those measurements and of the present results do not overlap, a comparison can only be made with resort to data extrapolation. Moreover, most of the data published by Laesecke⁷ were

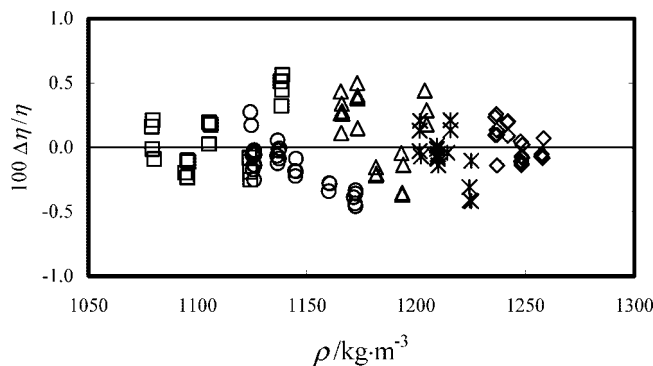


Figure 1. Relative deviation, $\Delta\eta/\eta = \eta_{\text{expt},i}/\eta_{\text{corr},i} - 1$, of the present viscosity measurements of R-507A (Table 1), $\eta_{\text{expt},i}$, from the correlation of the present experimental results given by equation 2, $\eta_{\text{corr},i}$, as a function of density along the isotherms: \diamond , 253.26 K; $*$, 263.23 K; \triangle , 273.24 K; \square , 283.22 K.

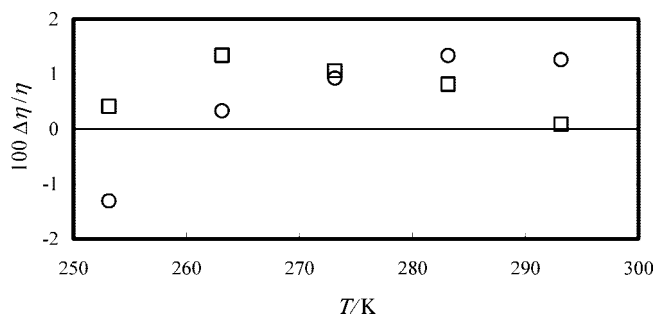


Figure 2. Deviation, $\Delta\eta/\eta = \eta_{\text{lit}}/\eta_{\text{corr}} - 1$, of literature viscosity data on R-507A, η_{lit} , from the correlation of the present experimental results given by eq 2, η_{corr} , as a function of temperature: \circ , ref 19; \square , refs 20 and 21.

obtained at supercritical or near critical temperatures. As a consequence, only the compressed liquid R-507A viscosity data published by that author at approximately 300 K were compared with the present results. For this purpose, the latter were extrapolated by means of the correlation eqs 2 and 4, with the parameters given in Tables 3 and 4. Only two data points of the isotherm obtained by Laesecke⁷ are within the pressure range of the present work. Those points deviate from the extrapolation of the present data by -0.22 % at 3.6 MPa and $+0.63$ % at 6.6 MPa, which is an excellent agreement. Moreover, an extrapolation of eq 2 along the isotherm at about 300 K up to a pressure of approximately 67 MPa shows that the absolute deviations of the data published by Laesecke⁷ do not exceed 1.9 %, which is still within the nominal uncertainty of the experimental method.

Measurements of the kinematic viscosity of liquid R-507A, at temperatures overlapping those of the present work, were performed along the saturation line, by Fröba et al.¹⁹ Those results, obtained with a surface light scattering technique, have an estimated uncertainty of ± 2 % and, for comparison with the present data, have been converted to dynamic viscosities, using the density values reported by the same authors.¹⁹ Recently, these results have been recalculated by Fröba using a more rigorous description of the liquid–vapor interface.^{20,21} The deviations of both sets of data from the correlation represented by eq 2, with the parameters given in Table 4, along the temperature range of the present results are shown in Figure 2. This comparison required the extrapolation of the present results to the saturation pressure by means of eq 2. The comparison shows that the results first reported by Fröba et al.¹⁹ have deviations within ± 1.3 % from the present values, while the corrected data^{20,21} deviate from (0.01 to 1.32) %. Therefore,

it may be concluded that, over the temperature range of our measurements, the deviations are well within the combined uncertainty of the experimental methods.

It is interesting to note that the viscosity of R-507A is predicted in excellent agreement with the present results, as a mole fraction average of the viscosity of its pure components at the same temperature and pressure, as calculated by the corresponding correlation equations of the experimental data published previously³

$$\eta_{\text{mixt}}(T, p) = x_i \eta_i(T, p) + x_j \eta_j(T, p) \quad (8)$$

The deviations of the values calculated by eq 8 from the experimental data shown in Table 1 are within (−0.56 to +0.84) % and are characterized by an AAD of 0.33 % and a bias of +0.19 %, where the following definitions have been used:

$$\text{AAD}/\% = \frac{100}{N} \sum_i^N \left| \frac{\eta_{\text{calcd},i}}{\eta_{\text{expt},i}} - 1 \right| \quad (9)$$

$$\text{bias}/\% = \frac{100}{N} \sum_i^N \left(\frac{\eta_{\text{calcd},i}}{\eta_{\text{expt},i}} - 1 \right) \quad (10)$$

where $\eta_{\text{expt},i}$ is the i th viscosity experimental datum of the present work (see Table 1); $\eta_{\text{calcd},i}$ is the corresponding value of the viscosity calculated by means of eq 8; and N is the number of experimental points in Table 1. It is noteworthy that the correlation eq 8 has been used²² as a reference for calculating the viscosity of mixing.

Conclusions

New viscosity measurements of compressed liquid R-507A in the temperature range (253 to 293) K and pressures up to 10 MPa have been presented. To the knowledge of the authors, the present results are the first set of viscosity data of compressed liquid R-507A, over that temperature range. The overall uncertainty of the results, obtained using a vibrating-wire instrument, is estimated not to exceed ± 1.0 %. The experimental results were fitted successfully with a correlation scheme based on a hard-sphere model. This was used for extrapolating the present data to the saturation line and to other conditions of temperature and pressure, to enable comparisons with literature data. Good agreement has been found with two sets of recent viscosity measurements of liquid R-507A.

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