

# Saturated-Liquid Viscosity of Ten Binary and Ternary Alternative Refrigerant Mixtures. Part I: Measurements<sup>†</sup>

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The saturated-liquid viscosities of 10 binary and ternary refrigerant mixtures composed of difluoromethane (R32), 1,1,1,2-tetrafluoroethane (R134a), pentafluoroethane (R125), and propane (R290) were measured in a sealed gravitational capillary viscometer with a straight vertical capillary from 245 to 345 K or to a maximum vapor pressure of 3 MPa. The maximum uncertainty of the measurements is estimated to be  $\pm 2.8\%$  for the mixtures of R32 + propane. The largest contribution to the total uncertainty is that of the saturated liquid and vapor densities of the mixtures, which had to be estimated. Comparisons with viscosity data from the literature show agreement within the mutual uncertainty for all but one data set. Similar agreement is found with predicted viscosities using the extended-corresponding-states model in NIST Standard Reference Database 23 (REFPROP), except for the mixtures of the polar fluids R32 and R134a with the nonpolar propane. Their viscosity–composition dependences are strongly nonlinear.

## Introduction

Mixtures of methane- and ethane-based hydrofluorocarbons (HFCs) are being investigated as substitutes for R12 (dichlorodifluoromethane,  $\text{CCl}_2\text{F}_2$ ) and R22 (difluorochloromethane,  $\text{CHClF}_2$ ). Whereas production of R12 was terminated by the end of 1995, the end of production of R22 as a working fluid in new refrigeration equipment is set for the year 2010. Other than HFCs, the hydrocarbon propane ( $\text{C}_3\text{H}_8$ ) is considered as a component in such mixtures because it facilitates their miscibility with mineral and alkylbenzene oils and improves the performance of refrigeration units. In fact, the miscibility of mineral lubricants is strongly increased by even low mass fractions of propane.<sup>1,2</sup> This is important for the maintenance-free durability of refrigeration equipment.

Viscosity measurements were carried out on binary systems of R32 ( $\text{CH}_2\text{F}_2$ ) with propane and R134a ( $\text{CF}_3\text{—CH}_2\text{F}$ ). Other binary systems studied were those of R134a with R125 ( $\text{CF}_3\text{—CHF}_2$ ) and with propane. The ternary mixtures were prepared from R32, R125, and R134a with R32 mole fractions of 0.3 and 0.33 and R125 mole fractions of 0.1 and 0.33. These measurements were part of a comprehensive program that also included measurements of the density, isochoric heat capacity, and thermal conductivity of the same samples.<sup>3</sup>

Mixtures of the systems R32 + R134a and R125 + R134a are considered as replacements for R22 in heat-pumping and air-conditioning equipment. In addition, they are constituent binaries of the ternary mixtures that were measured. They were also chosen for the theoretical information they would yield: R32 + R134a represents the case of a system of two highly polar compounds of different sizes, whereas R125 + R134a is a system of two compounds of similar size and polarity. Similar considerations guided the selection of the binaries of propane with R32 and R134a, with the difference being that propane is a nonpolar

molecule. In the system propane + R134a, the propane is paired with a polar compound of approximately equal size. In the system R32 + propane, it is paired with a considerably smaller and more polar compound. The ternary system R32 + R125 + R134a is currently the subject of intense industrial research as a possible replacement for R22. The measured molar composition 0.3 + 0.1 + 0.6, respectively, is close to that of the refrigerant blend R407C (0.38 + 0.18 + 0.44). The equimolar composition was measured for theoretical reasons.

This report is organized as follows. The preparation of the mixture samples is described next. Subsequently, the experimental procedure and analysis of the viscosity measurements are explained. These measurements differ markedly from conventional measurements of nonvolatile liquids in open gravitational capillary viscometers. The experimental viscosities are tabulated, and the influence of mixture composition is illustrated graphically. The measured data are compared with viscosities that were estimated by means of the extended-corresponding-states method in NIST Standard Reference Database 23 (REFPROP), version 6.01. They are also compared with literature data.

## Experimental Section

**Mixture Preparation.** The mixtures were gravimetrically prepared from pure compounds of research-grade purity and kept as vapors in 15.8-L aluminum cylinders. The purities of the pure fluids were determined by gas chromatography as follows: 99.9 mol % for R32 and R125 and 99.43 mol % for R134a. The major impurity of R134a was air, which was reduced from 0.57 mol % to a mole fraction of 75 ppm (mol/mol) by twice freezing the sample in liquid nitrogen and pumping off volatile gases. The propane supply included initially 500 ppm air and 400 ppm ethane. The air fraction dropped to 5 ppm after a vapor purge.

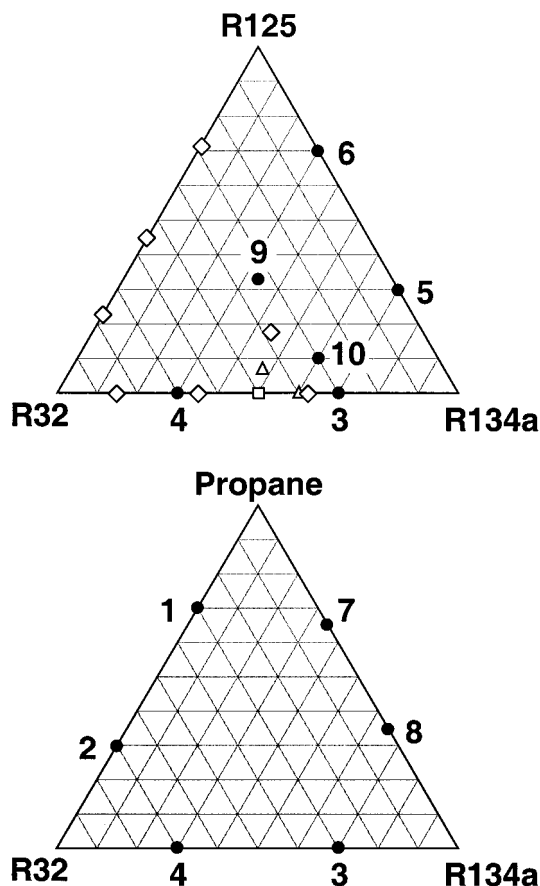
Two compositions were prepared for each of the five systems. Their mole and mass fractions are given in Table

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**Table 1. Constituents of the Mixtures and Their Compositions in Mole Fractions  $x$  and Mass Fractions  $w$** 

	component 1	component 2	component 3	$x_1$	$x_2$	$x_3$	$w_1$	$w_2$	$w_3$
DoE-1	R32	propane	–	0.299920	0.700080	–	0.33573	0.66427	–
DoE-2	R32	propane	–	0.700050	0.299950	–	0.73358	0.26642	–
DoE-3	R32	R134a	–	0.299557	0.700443	–	0.17902	0.82098	–
DoE-4	R32	R134a	–	0.700310	0.299690	–	0.54369	0.45631	–
DoE-5	R125	R134a	–	0.300150	0.699850	–	0.33533	0.66467	–
DoE-6	R125	R134a	–	0.699991	0.300009	–	0.73295	0.26705	–
DoE-7	propane	R134a	–	0.649822	0.350178	–	0.44437	0.55563	–
DoE-8	propane	R134a	–	0.351581	0.648412	–	0.18942	0.81058	–
DoE-9	R32	R125	R134a	0.332985	0.333424	0.333591	0.18958	0.43794	0.37248
DoE-10	R32	R125	R134a	0.300277	0.099952	0.599771	0.17589	0.13507	0.68904



**Figure 1.** Compositions of binary and ternary mixtures studied in this work and of literature viscosity data. ●, This work; ◇, Heide and Schenk, 1996;<sup>11</sup> △, Bivens et al., 1993;<sup>16</sup> □, Ripple and Matar, 1993.<sup>17</sup>

1 to the number of significant digits that follows from the uncertainty of the balance that was used for the mixture preparation. Whereas the binary mixtures of R32 + propane, R32 + R134a, and R125 + R134a were measured at nominal mole fractions of  $x_1 = 0.3$  and  $0.7$  each, the mixtures of propane + R134a were measured at nominal mole fractions of  $x_1 = 0.35$  and  $0.65$ . The nominal compositions of the ternary mixtures were  $0.33$  R32 +  $0.33$  R125 +  $0.34$  R134a and  $0.3$  R32 +  $0.1$  R125 +  $0.6$  R134a. The compositions of the mixtures studied in this work and of the literature viscosity data are summarized in Figure 1.

**Apparatus and Procedure.** The normal boiling-point temperatures of the investigated mixtures are below ambient temperature, which means that their vapor pressures at ambient temperature are higher than atmospheric pressure. Viscometry of such fluids requires sealed instruments to prevent the evaporation of the volatile samples during the measurements. The sealed gravitational capillary viscometer of this work has been employed before for

measurements of fluorinated propane derivatives<sup>4,5</sup> and three fluorinated ethane derivatives including R125,<sup>6</sup> as well as two of the pure components of the present mixtures, R32 and R134a, and ammonia.<sup>7</sup> The latter work contains a detailed description of the instrument, the measurement protocol, and a comprehensive discussion of the working theory. It was not necessary to remeasure the viscosity of the fourth pure component, R290 (propane), because a new reference correlation has been established recently based on a critical evaluation of the available literature data.<sup>8</sup> Thus, benchmark viscosities are available for all four pure constituents, with which the viscosities of their mixtures can be compared.

In preparation of the filling of the viscometer, a sample cylinder was placed upright on a hot plate and heated overnight to about 343 K at the plate. The temperature gradient across the height of the cylinder gave rise to natural convection inside the container, so that the mixture was stirred. A liquid volume of approximately 3 mL of each mixture was transferred for the measurements from a sample cylinder into the evacuated viscometer by cooling the lower reservoir of the instrument to approximately 240 K over evaporating liquid nitrogen. A homogeneous liquid phase was observed in the lower viscometer reservoir for all mixtures except DoE-2 ( $0.7$  R32 +  $0.3$  propane), which formed two liquid phases. The filled viscometer was subsequently weighed on a large precision balance before it was mounted on the viscometer stand, which was then immersed into the liquid bath of the thermostat. The viscometer contents were stirred well during these translational and rotational transfers of the instrument. The homogeneity of the liquid mixtures was controlled during the viscosity measurements, which consist in the visual observation of the downward movement of the vapor–liquid interface. Single liquid phases were observed during all of the measurements reported here. The sample liquid was stirred after each run when the viscometer was inverted to return the liquid from the lower to the upper reservoir.

Whereas compositional changes are negligible during measurements of nonvolatile liquid mixtures in open gravitational capillary viscometers, measurements in sealed instruments do involve such compositional changes with temperature. These changes have to be considered in the analysis of the experimental data. Ideally, the composition of the coexisting liquid and vapor should be determined by drawing samples from the viscometer at each temperature. Such an approach, however, was impractical with the present instrument. Therefore, the dependence on composition was introduced in the data analysis by measuring the bulk density of the mixture in the sealed viscometer after it had been filled. This value, together with the known sample composition, was then used to estimate the compositions and densities of the coexisting vapor and liquid phases at each temperature with the mixture models

in NIST Standard Reference Database 23 (REFPROP).<sup>9,10</sup>

The bulk density of a mixture in the viscometer was determined from the difference in mass of the filled and evacuated instrument and from its internal volume, which was calculated from its geometric dimensions. The temperature range of the measurements was 245–350 K or to a temperature corresponding to a maximum vapor pressure of about 3 MPa. Measurements were carried out in temperature intervals of 5 K with four repetitive runs at each temperature. Versions 5.0, 5.10, 5.18, and 6.01 (with several mixture model revisions) of NIST Standard Reference Database 23 (REFPROP) were used to analyze the viscosity measurements, as this database was undergoing a major revision at the same time. The property models for the mixtures improved steadily, so that the calculated mixture densities and compositions became more reliable. REFPROP, version 6.01, was used in the analysis of the results reported here. Based on our experiences, the earlier versions of REFPROP should no longer be used for refrigerant mixtures.

Comparative calculations showed that the estimated compositions and densities of the saturated liquid and vapor were rather insensitive even to substantial variations in the bulk density, on the order of  $\pm 10\%$  or more. Flash calculations based on the measured bulk densities and temperatures were successful for the mixtures that contain only hydrofluorocarbons. They did not converge for the nonpolar + polar mixtures containing propane. Therefore, the compositions and densities of the coexisting vapor and liquid phases of these mixtures were estimated by assuming that the composition of the liquid was equal to the known bulk composition and independent of temperature. To account for this assumption, the uncertainty in the predicted saturated liquid and vapor densities is estimated to be  $\pm 1\%$  for the mixtures containing propane (DoE-1, 2, 7, and 8) and  $\pm 0.2\%$  for those containing only hydrofluorocarbons. These margins were entered into the previous uncertainty analyses for R32 and R134a.<sup>7</sup> The previous uncertainty contribution of  $\pm 0.5\%$  due to calibration with toluene was supported in three recalibrations of the instrument, which were carried out during the present mixture measurements. The previous estimates for uncertainties due to surface-tension effects were also retained in view of experimental data for the surface tension of the hydrofluorocarbon mixtures.<sup>11–13</sup> Surface-tension data for the mixtures containing propane could not be located. However, these values should be lower than the surface tensions of the hydrofluorocarbon mixtures, so that the uncertainty margins due to surface-tension effects for the HFC mixtures are conservative for those containing propane.

The particular contribution to the total uncertainty of the viscosity measurements that arises from the uncertainty in the initial mixture composition was assessed for the mixture propane + R134a with a mole fraction of  $x_1 = 0.35$  (DoE-8). This system exhibits the strongest viscosity–composition dependence of all those measured in this work, because propane and R134a are respectively the pure components with the lowest and the highest viscosities. Using NIST Standard Reference Database 23 (REFPROP), version 6.01,<sup>10</sup> the variation in the viscosity of that mixture at 245 K due to an assumed variation in the mixture composition of  $\Delta x_1 = \pm 0.0001$  ranges between  $-0.02$  and  $0.008\%$ . As can be seen from Table 1, the uncertainty in the mixture mole fractions is 2 orders of magnitude smaller than this variation. Therefore, the viscosity uncertainty contribution due to the uncertainty of the initial mixture composition is negligible for this mixture, as well as for

**Table 2. Estimated Expanded Uncertainties<sup>a</sup> for the Present Mixture Viscosity Measurements in the Sealed Gravitational Capillary Viscometer**

	com- ponent 1	com- ponent 2	com- ponent 3	highest measured temperatures (K)	estimated expanded uncertainty (%)
DoE-1, 2	R32	propane	—	335, 305	2.8
DoE-3, 4	R32	R134a	—	335, 325	2.4
DoE-5, 6	R125	R134a	—	345, 335	2.2
DoE-7, 8	propane	R134a	—	335, 345	2.6
DoE-9, 10	R32	R125	R134a	330, 340	2.4

<sup>a</sup> Coverage factor  $k = 2$ .

all others that were measured in this work, because their viscosity–composition dependence is weaker than that of the system propane + R134a.

The estimated expanded uncertainties for the present measurements are compiled in Table 2. They pertain to the highest measured temperature for each mixture and should be about 0.2% smaller at the lowest temperatures. The estimated uncertainties of the present measurements range from  $\pm 2.2\%$  for the R125 + R134a mixtures (DoE-5 and DoE-6) up to  $\pm 2.8\%$  for the R32 + propane mixtures (DoE-1 and DoE-2) because of the higher uncertainty in the density prediction for this nonpolar + polar system. The precision of the measurements is  $\pm 0.5\%$ .

## Results and Discussion

The experimental results for the saturated liquid viscosity of the 10 mixtures are reported in Tables 3–12, together with the calculated densities and compositions of the liquid and vapor at each temperature. To establish traceability, information about the estimation of the saturated liquid and vapor densities,  $\rho_l$  and  $\rho_v$ , respectively, based on the measured bulk density  $\rho$  of each mixture is included in each table heading. The results are discussed in three parts. First, the temperature dependences of the mixture viscosities are compared with those of the pure fluids to deduce the dependence on composition. Second, the experimental viscosities are compared with values predicted with the mixture models and the extended-corresponding-states viscosity model of Klein et al.,<sup>14</sup> as implemented in REFPROP, version 6.01. Finally, the present results are compared with literature data for the systems that were measured in this work.

**Temperature and Composition Dependence of the Mixture Viscosities.** Because the measurements of the volatile mixtures were carried out at constant bulk density rather than at constant composition, the composition dependence of the mixture viscosity cannot be deduced from the results as simply as in the numerous measurements of nonvolatile mixtures. In that case, the composition dependence of mixture viscosity can be studied at constant pressure and temperature, and the results can then be interpreted in terms of various “mixing rules”. In the present viscosity measurements of volatile mixtures, the state parameters temperature, density, and composition vary simultaneously.

The experimental results are shown in viscosity–temperature diagrams (Figures 2–6). The data for each of the two compositions of a system are put in the context of the viscosities of the pure fluids to exhibit the viscosity–composition dependence in a system. Figure 2 shows the combined results for the R32 + propane mixtures (DoE-1 and DoE-2). They are typical of the results for the other mixtures that were measured. The saturated-liquid viscosities of both R32 + propane mixtures are lower than the



**Table 3. Saturated-Liquid Viscosities for Mixture DoE-1 (0.3 R32 + 0.7 Propane) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{l,R32}$	$w_{v,R32}$	$\eta$ (mPa s)
247.31	654.71	12.30	0.3357	0.6641	0.1510
247.32	654.69	12.31	0.3357	0.6641	0.1522
247.33	654.68	12.31	0.3357	0.6641	0.1527
247.34	654.66	12.31	0.3357	0.6641	0.1522
250.07	649.97	13.42	0.3357	0.6591	0.1479
250.07	649.98	13.42	0.3357	0.6591	0.1491
250.07	649.97	13.42	0.3357	0.6591	0.1479
250.07	649.97	13.42	0.3357	0.6591	0.1496
255.06	641.25	15.65	0.3357	0.6505	0.1386
255.06	641.25	15.65	0.3357	0.6505	0.1391
255.06	641.25	15.65	0.3357	0.6505	0.1386
255.06	641.25	15.65	0.3357	0.6505	0.1386
260.07	632.28	18.16	0.3357	0.6421	0.1309
260.06	632.28	18.15	0.3357	0.6421	0.1309
260.07	632.28	18.16	0.3357	0.6421	0.1318
260.07	632.27	18.16	0.3357	0.6421	0.1314
265.07	623.02	20.97	0.3357	0.6340	0.1232
265.07	623.02	20.97	0.3357	0.6340	0.1232
265.07	623.02	20.97	0.3357	0.6340	0.1232
265.07	623.02	20.97	0.3357	0.6340	0.1236
270.07	613.58	24.10	0.3357	0.6261	0.1168
270.07	613.57	24.11	0.3357	0.6261	0.1172
270.07	613.57	24.11	0.3357	0.6261	0.1175
275.08	603.80	27.63	0.3357	0.6181	0.1109
275.08	603.80	27.63	0.3357	0.6181	0.1109
275.07	603.81	27.62	0.3357	0.6181	0.1106
280.10	593.70	31.55	0.3357	0.6100	0.1053
280.10	593.70	31.55	0.3357	0.6100	0.1049
280.10	593.70	31.55	0.3357	0.6100	0.1049
280.10	593.70	31.55	0.3357	0.6100	0.1053
285.08	583.28	35.90	0.3357	0.6017	0.1004
285.08	583.28	35.90	0.3357	0.6017	0.0995
285.08	583.27	35.91	0.3357	0.6017	0.1001
285.08	583.27	35.91	0.3357	0.6017	0.1001
290.06	572.47	40.72	0.3357	0.5930	0.0947
290.06	572.46	40.72	0.3357	0.5930	0.0949
290.06	572.47	40.72	0.3357	0.5930	0.0944
290.06	572.47	40.72	0.3357	0.5930	0.0944
295.07	561.13	46.15	0.3357	0.5838	0.0891
295.07	561.14	46.15	0.3357	0.5838	0.0891
295.08	561.13	46.16	0.3357	0.5838	0.0888
295.07	561.13	46.15	0.3357	0.5838	0.0891
300.06	549.36	52.15	0.3357	0.5741	0.0836
300.06	549.36	52.16	0.3357	0.5741	0.0841
300.06	549.36	52.16	0.3357	0.5741	0.0839
300.06	549.36	52.15	0.3357	0.5741	0.0841
305.08	536.96	58.92	0.3357	0.5636	0.0791
305.07	536.97	58.91	0.3357	0.5636	0.0798
305.08	536.96	58.92	0.3357	0.5636	0.0791
305.08	536.95	58.92	0.3357	0.5636	0.0793
305.07	536.96	58.92	0.3357	0.5636	0.0793
310.10	523.83	66.48	0.3357	0.5522	0.0751
310.10	523.83	66.48	0.3357	0.5522	0.0751
310.10	523.82	66.48	0.3357	0.5522	0.0747
310.10	523.83	66.48	0.3357	0.5522	0.0749
315.07	510.01	74.92	0.3357	0.5401	0.0703
315.08	509.98	74.93	0.3357	0.5400	0.0701
315.07	509.99	74.92	0.3357	0.5400	0.0703
315.07	510.01	74.92	0.3357	0.5401	0.0703
320.12	495.02	84.59	0.3357	0.5266	0.0649
320.12	495.03	84.58	0.3357	0.5266	0.0652
320.12	495.03	84.58	0.3357	0.5266	0.0654
320.12	495.03	84.58	0.3357	0.5266	0.0656
325.11	478.99	95.48	0.3357	0.5121	0.0605
325.11	478.98	95.48	0.3357	0.5121	0.0605
325.11	478.97	95.49	0.3357	0.5121	0.0605
325.11	478.99	95.48	0.3357	0.5121	0.0606
330.08	461.62	107.91	0.3357	0.4964	0.0558
330.08	461.60	107.92	0.3357	0.4964	0.0557
330.08	461.60	107.92	0.3357	0.4963	0.0557
330.08	461.60	107.92	0.3357	0.4964	0.0556
335.09	441.99	122.52	0.3357	0.4791	0.0509
335.09	441.99	122.53	0.3357	0.4791	0.0507
335.09	441.98	122.53	0.3357	0.4791	0.0507
335.10	441.95	122.55	0.3357	0.4790	0.0506

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities were calculated with REFPROP, version 6.01, at each temperature for the constant saturated-liquid composition of mass fraction  $w_{l,R32} = 0.3357$ . Flash calculations for the measured mixture bulk density  $\rho = 257.5$  kg m<sup>-3</sup> did not converge.

**Table 4. Saturated-Liquid Viscosities for Mixture DoE-2 (0.7 R32 + 0.3 Propane) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{l,R32}$	$w_{v,R32}$	$\eta$ (mPa s)
245.91	862.83	12.49	0.7336	0.6505	0.1666
245.92	862.79	12.50	0.7336	0.6506	0.1666
245.93	862.77	12.50	0.7336	0.6506	0.1661
250.05	852.58	14.38	0.7336	0.6577	0.1582
250.04	852.59	14.38	0.7336	0.6577	0.1563
250.03	852.62	14.38	0.7336	0.6577	0.1568
250.03	852.63	14.37	0.7336	0.6577	0.1592
255.07	839.78	16.98	0.7336	0.6659	0.1456
255.07	839.77	16.98	0.7336	0.6659	0.1477
255.07	839.77	16.98	0.7336	0.6659	0.1472
255.07	839.78	16.98	0.7336	0.6659	0.1464
260.06	826.73	19.91	0.7336	0.6735	0.1368
260.06	826.73	19.91	0.7336	0.6735	0.1380
260.06	826.74	19.91	0.7336	0.6734	0.1376
260.06	826.74	19.91	0.7336	0.6734	0.1376
265.04	813.38	23.26	0.7336	0.6805	0.1297
265.04	813.40	23.25	0.7336	0.6805	0.1287
265.04	813.38	23.26	0.7336	0.6805	0.1315
265.04	813.39	23.25	0.7336	0.6805	0.1315
270.09	799.34	27.11	0.7336	0.6872	0.1228
270.09	799.36	27.10	0.7336	0.6872	0.1229
270.09	799.34	27.11	0.7336	0.6872	0.1228
270.09	799.34	27.11	0.7336	0.6872	0.1238
275.05	785.11	31.42	0.7336	0.6932	0.1146
275.05	785.09	31.43	0.7336	0.6932	0.1145
275.05	785.12	31.42	0.7336	0.6932	0.1148
275.05	785.10	31.43	0.7336	0.6932	0.1157
280.08	770.16	36.39	0.7336	0.6990	0.1096
280.08	770.14	36.39	0.7336	0.6990	0.1096
280.09	770.14	36.39	0.7336	0.6990	0.1096
280.08	770.15	36.39	0.7336	0.6990	0.1102
285.09	754.56	42.04	0.7336	0.7042	0.1032
285.09	754.58	42.03	0.7336	0.7042	0.1029
285.12	754.49	42.07	0.7336	0.7042	0.1034
285.12	754.49	42.07	0.7336	0.7042	0.1036
290.07	738.36	48.42	0.7336	0.7090	0.0969
290.08	738.33	48.43	0.7336	0.7090	0.0973
290.07	738.36	48.42	0.7336	0.7090	0.0967
290.08	738.34	48.43	0.7336	0.7090	0.0964
295.03	721.39	55.71	0.7336	0.7135	0.0908
295.04	721.38	55.72	0.7336	0.7135	0.0906
295.03	721.39	55.71	0.7336	0.7135	0.0910
295.03	721.40	55.71	0.7336	0.7135	0.0908
300.10	703.04	64.22	0.7336	0.7176	0.0852
300.10	703.03	64.23	0.7336	0.7176	0.0852
300.10	703.04	64.22	0.7336	0.7176	0.0857
300.10	703.02	64.23	0.7336	0.7176	0.0854
305.08	683.83	73.95	0.7336	0.7214	0.0782
305.09	683.81	73.96	0.7336	0.7214	0.0794
305.08	683.82	73.95	0.7336	0.7214	0.0794
305.09	683.79	73.97	0.7336	0.7214	0.0790

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities were calculated with REFPROP, version 6.01, at each temperature for the constant saturated-liquid composition of mass fraction  $w_{l,R32} = 0.7336$ . Flash calculations for the measured mixture bulk density  $\rho = 267.4$  kg m<sup>-3</sup> did not converge.

viscosity of the nonpolar fluid propane, which, in turn, is lower than the viscosity of R32. Despite their quite different compositions, the viscosities of these mixtures are almost equal, whereas the addition of 30 mol % of propane to R32 lowers the viscosity dramatically. Obviously, the viscosity–composition dependence of the system R32 + propane is nonlinear, as it exhibits a minimum at a mole fraction of approximately 0.30 R32 at 250 K. The viscosity minimum broadens at higher temperatures, and its locus moves from lower to higher R32 concentrations.

On the other hand, the saturated liquid viscosities of the R32 + R134a mixtures are spaced almost equidistantly between those of the pure fluids, as seen in Figure 3. This indicates a rather linear dependence on composition,

**Table 5. Saturated-Liquid Viscosities for Mixture DoE-3 (0.3 R32 + 0.7 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

T (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$W_{l,R32}$	$W_{v,R32}$	$\eta$ (mPa s)
255.00	1301.99	8.79	0.1764	0.3776	0.2851
254.96	1302.14	8.77	0.1764	0.3777	0.2892
254.97	1302.10	8.77	0.1764	0.3777	0.2863
254.96	1302.11	8.77	0.1764	0.3777	0.2855
260.08	1285.75	10.62	0.1759	0.3679	0.2692
260.08	1285.75	10.62	0.1759	0.3679	0.2726
260.09	1285.74	10.62	0.1759	0.3679	0.2685
260.08	1285.75	10.62	0.1759	0.3679	0.2689
260.08	1285.75	10.62	0.1759	0.3679	0.2685
265.13	1269.61	12.74	0.1755	0.3585	0.2512
265.14	1269.58	12.75	0.1755	0.3585	0.2516
265.14	1269.59	12.75	0.1755	0.3585	0.2514
265.13	1269.60	12.74	0.1755	0.3585	0.2506
270.11	1253.62	15.15	0.1750	0.3497	0.2342
270.11	1253.63	15.15	0.1750	0.3497	0.2381
270.11	1253.61	15.15	0.1750	0.3497	0.2344
270.11	1253.62	15.15	0.1750	0.3497	0.2358
275.06	1236.83	17.90	0.1745	0.3411	0.2210
275.06	1236.83	17.90	0.1745	0.3411	0.2226
275.06	1236.83	17.90	0.1745	0.3411	0.2218
275.06	1236.83	17.90	0.1745	0.3411	0.2242
280.09	1219.69	21.09	0.1740	0.3326	0.2083
280.09	1219.69	21.09	0.1740	0.3326	0.2087
280.09	1219.69	21.09	0.1740	0.3326	0.2083
280.09	1219.69	21.09	0.1740	0.3326	0.2083
247.70	1324.91	6.59	0.1770	0.3922	0.3147
247.70	1324.91	6.59	0.1770	0.3922	0.3157
247.70	1324.91	6.59	0.1770	0.3922	0.3153
247.70	1324.91	6.59	0.1770	0.3922	0.3133
250.15	1317.47	7.27	0.1768	0.3872	0.3047
250.15	1317.49	7.27	0.1768	0.3872	0.3046
250.15	1317.48	7.27	0.1768	0.3872	0.3048
250.15	1317.48	7.27	0.1768	0.3872	0.3046
285.08	1202.22	24.72	0.1734	0.3244	0.1955
285.08	1202.23	24.71	0.1734	0.3244	0.1962
285.08	1202.22	24.72	0.1734	0.3244	0.1950
285.08	1202.23	24.72	0.1734	0.3244	0.1956
290.07	1183.76	28.82	0.1729	0.3164	0.1834
290.07	1183.76	28.82	0.1729	0.3164	0.1834
290.07	1183.77	28.82	0.1729	0.3164	0.1847
295.07	1164.70	33.53	0.1723	0.3085	0.1739
295.08	1164.70	33.54	0.1723	0.3085	0.1733
295.08	1164.70	33.54	0.1723	0.3085	0.1727
295.07	1164.71	33.53	0.1723	0.3085	0.1751
300.07	1145.73	38.85	0.1717	0.3008	0.1633
300.07	1145.74	38.85	0.1717	0.3008	0.1627
300.07	1145.73	38.85	0.1717	0.3008	0.1641
300.07	1145.73	38.85	0.1717	0.3008	0.1635
305.05	1125.80	44.89	0.1711	0.2932	0.1541
305.06	1125.78	44.90	0.1711	0.2931	0.1530
305.05	1125.79	44.89	0.1711	0.2931	0.1541
305.05	1125.78	44.89	0.1711	0.2931	0.1538
310.05	1104.76	51.75	0.1705	0.2856	0.1433
310.06	1104.75	51.75	0.1705	0.2856	0.1437
310.06	1104.75	51.75	0.1705	0.2856	0.1441
310.06	1104.74	51.76	0.1705	0.2856	0.1442
315.08	1082.15	59.64	0.1698	0.2780	0.1355
315.08	1082.15	59.64	0.1698	0.2780	0.1363
315.08	1082.14	59.64	0.1698	0.2780	0.1357
315.08	1082.14	59.64	0.1698	0.2780	0.1356
320.06	1059.69	68.53	0.1692	0.2705	0.1277
320.07	1059.67	68.54	0.1692	0.2705	0.1273
320.07	1059.67	68.54	0.1692	0.2705	0.1274
320.07	1059.67	68.54	0.1692	0.2705	0.1281
325.09	1034.54	78.86	0.1685	0.2629	0.1194
325.10	1034.53	78.86	0.1685	0.2629	0.1192
325.10	1034.52	78.86	0.1685	0.2629	0.1203
325.10	1034.52	78.87	0.1685	0.2629	0.1195
330.15	1009.17	90.77	0.1678	0.2552	0.1116
330.15	1009.13	90.79	0.1678	0.2552	0.1113
330.15	1009.12	90.79	0.1678	0.2552	0.1110
330.16	1009.12	90.79	0.1678	0.2552	0.1110
335.10	981.04	104.34	0.1671	0.2476	0.1039
335.10	981.04	104.33	0.1671	0.2476	0.1039
335.10	981.03	104.34	0.1671	0.2476	0.1036
335.10	981.03	104.34	0.1671	0.2476	0.1035

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities and compositions were obtained from REFPROP, version 6.01, by flash calculations for the measured mixture bulk density  $\rho = 438.76$  kg m<sup>-3</sup>.

**Table 6. Saturated-Liquid Viscosities for Mixture DoE-4 (0.7 R32 + 0.3 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

T (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$W_{l,R32}$	$W_{v,R32}$	$\eta$ (mPa s)
246.07	1237.78	8.64	0.5423	0.7694	0.2529
246.08	1237.75	8.65	0.5423	0.7694	0.2532
246.64	1236.07	8.84	0.5422	0.7686	0.2510
246.65	1236.06	8.84	0.5422	0.7686	0.2523
246.65	1236.06	8.84	0.5422	0.7686	0.2498
246.65	1236.04	8.84	0.5422	0.7686	0.2516
250.01	1225.98	8.79	0.5420	0.7640	0.2409
250.01	1225.97	8.80	0.5420	0.7640	0.2423
250.01	1225.97	8.80	0.5420	0.7640	0.2420
250.01	1225.96	8.80	0.5420	0.7640	0.2426
254.99	1210.54	10.60	0.5416	0.7572	0.2266
254.99	1210.52	10.60	0.5416	0.7571	0.2264
255.01	1210.48	10.60	0.5416	0.7571	0.2271
260.03	1194.89	12.70	0.5411	0.7502	0.2122
260.04	1194.88	12.71	0.5411	0.7502	0.2127
260.04	1194.88	12.71	0.5411	0.7502	0.2121
260.04	1194.87	12.71	0.5411	0.7502	0.2128
264.99	1178.55	15.10	0.5405	0.7434	0.2022
264.99	1178.54	15.10	0.5405	0.7434	0.2012
265.00	1178.52	15.11	0.5405	0.7434	0.2037
265.00	1178.51	15.11	0.5405	0.7434	0.2019
269.91	1161.32	17.82	0.5400	0.7366	0.1913
269.91	1161.31	17.82	0.5400	0.7366	0.1911
269.91	1161.31	17.82	0.5400	0.7366	0.1912
269.92	1161.29	17.82	0.5400	0.7366	0.1914
275.02	1144.45	21.07	0.5393	0.7295	0.1801
275.04	1144.37	21.08	0.5393	0.7295	0.1801
275.04	1144.35	21.08	0.5393	0.7294	0.1798
275.05	1144.31	21.09	0.5393	0.7294	0.1817
280.13	1126.46	24.79	0.5386	0.7223	0.1701
280.14	1126.46	24.79	0.5386	0.7223	0.1708
280.14	1126.46	24.79	0.5386	0.7223	0.1716
280.14	1126.45	24.79	0.5386	0.7223	0.1716
285.15	1107.96	28.96	0.5379	0.7151	0.1631
285.16	1107.96	28.96	0.5379	0.7151	0.1612
285.16	1107.95	28.96	0.5379	0.7151	0.1609
285.16	1107.94	28.97	0.5379	0.7151	0.1611
290.15	1089.40	33.69	0.5372	0.7078	0.1525
290.15	1089.40	33.69	0.5372	0.7078	0.1533
290.16	1089.38	33.69	0.5372	0.7078	0.1544
290.16	1089.37	33.69	0.5372	0.7078	0.1519
295.14	1069.44	39.09	0.5365	0.7004	0.1433
295.14	1069.44	39.09	0.5365	0.7004	0.1436
295.14	1069.44	39.09	0.5365	0.7004	0.1435
295.14	1069.44	39.09	0.5365	0.7004	0.1436
300.06	1049.78	45.12	0.5358	0.6929	0.1338
300.06	1049.77	45.13	0.5358	0.6929	0.1357
305.01	1028.44	52.09	0.5351	0.6852	0.1293
305.01	1028.44	52.09	0.5351	0.6852	0.1293
305.02	1028.43	52.09	0.5351	0.6852	0.1245
305.02	1028.43	52.09	0.5351	0.6852	0.1260
305.02	1028.43	52.09	0.5351	0.6852	0.1280
305.02	1028.42	52.09	0.5351	0.6851	0.1300
305.02	1028.42	52.09	0.5351	0.6851	0.1285
310.16	1006.25	60.34	0.5344	0.6769	0.1209
310.16	1006.23	60.34	0.5344	0.6769	0.1209
310.16	1006.23	60.34	0.5344	0.6769	0.1207
310.16	1006.22	60.35	0.5344	0.6769	0.1207
315.01	983.04	69.33	0.5337	0.6689	0.1136
315.02	983.02	69.33	0.5337	0.6689	0.1141
315.02	983.01	69.33	0.5337	0.6689	0.1146
315.02	983.01	69.33	0.5337	0.6689	0.1152
319.99	958.26	79.88	0.5330	0.6605	0.1065
319.99	958.26	79.89	0.5330	0.6605	0.1071
319.99	958.25	79.89	0.5330	0.6605	0.1068
319.99	958.25	79.89	0.5330	0.6605	0.1073
325.03	931.21	92.40	0.5324	0.6516	0.0997
325.03	931.18	92.41	0.5324	0.6516	0.0999
325.03	931.16	92.42	0.5324	0.6516	0.0999
325.04	931.16	92.42	0.5324	0.6516	0.1000

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities and compositions were obtained from REFPROP, version 6.01, by flash calculations for the measured mixture bulk density  $\rho = 501.25$  kg m<sup>-3</sup>.

**Table 7. Saturated-Liquid Viscosities for Mixture DoE-5 (0.3 R125 + 0.7 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$W_{l,R125}$	$W_{v,R125}$	$\eta$ (mPa s)
249.74	1389.77	9.70	0.3314	0.5535	0.3372
249.76	1389.73	9.71	0.3313	0.5535	0.3386
249.77	1389.70	9.71	0.3313	0.5534	0.3440
249.78	1389.67	9.72	0.3313	0.5534	0.3399
255.15	1371.99	11.90	0.3306	0.5426	0.3126
255.15	1371.97	11.90	0.3306	0.5426	0.3149
255.16	1371.95	11.90	0.3306	0.5426	0.3149
255.16	1371.93	11.91	0.3306	0.5425	0.3161
260.09	1354.74	14.21	0.3299	0.5328	0.2929
260.09	1354.74	14.21	0.3299	0.5328	0.2939
260.09	1354.74	14.21	0.3299	0.5328	0.2939
260.09	1354.74	14.22	0.3299	0.5328	0.2971
265.10	1338.15	16.93	0.3292	0.5232	0.2750
265.10	1338.14	16.93	0.3292	0.5232	0.2741
265.11	1338.13	16.94	0.3292	0.5232	0.2759
265.11	1338.13	16.94	0.3292	0.5232	0.2750
270.12	1320.50	20.04	0.3284	0.5137	0.2576
270.13	1320.50	20.04	0.3284	0.5137	0.2585
270.13	1320.50	20.05	0.3284	0.5137	0.2585
270.13	1320.50	20.05	0.3284	0.5137	0.2576
275.09	1302.19	23.56	0.3275	0.5044	0.2417
275.09	1302.19	23.56	0.3275	0.5044	0.2417
275.09	1302.19	23.56	0.3275	0.5044	0.2409
275.09	1302.18	23.56	0.3275	0.5044	0.2432
280.07	1283.71	27.55	0.3267	0.4953	0.2297
280.07	1283.71	27.55	0.3267	0.4953	0.2297
280.07	1283.70	27.55	0.3267	0.4953	0.2290
280.19	1283.23	27.66	0.3266	0.4951	0.2295
285.05	1264.79	32.10	0.3258	0.4863	0.2145
285.07	1264.73	32.12	0.3258	0.4863	0.2145
285.07	1264.72	32.12	0.3258	0.4863	0.2151
285.07	1264.72	32.12	0.3258	0.4863	0.2151
290.09	1244.64	37.29	0.3248	0.4773	0.1978
290.09	1244.63	37.29	0.3248	0.4773	0.1978
290.09	1244.62	37.30	0.3248	0.4773	0.1983
290.10	1244.62	37.30	0.3248	0.4773	0.1978
295.09	1224.64	43.15	0.3238	0.4685	0.1860
295.09	1224.63	43.15	0.3238	0.4685	0.1856
295.09	1224.63	43.16	0.3238	0.4685	0.1851
295.10	1224.62	43.16	0.3238	0.4685	0.1851
300.08	1203.64	49.74	0.3228	0.4598	0.1741
300.08	1203.64	49.74	0.3228	0.4598	0.1749
300.08	1203.64	49.74	0.3228	0.4598	0.1745
300.08	1203.63	49.74	0.3228	0.4598	0.1745
305.08	1181.16	57.23	0.3218	0.4510	0.1646
305.08	1181.16	57.23	0.3218	0.4510	0.1639
305.08	1181.15	57.23	0.3218	0.4510	0.1631
305.08	1181.15	57.23	0.3218	0.4510	0.1631
310.07	1158.64	65.68	0.3208	0.4424	0.1542
310.07	1158.64	65.68	0.3208	0.4424	0.1527
310.07	1158.64	65.68	0.3208	0.4424	0.1524
310.07	1158.64	65.69	0.3208	0.4424	0.1527
315.09	1134.54	75.39	0.3197	0.4336	0.1431
315.09	1134.54	75.39	0.3197	0.4336	0.1431
315.10	1134.53	75.39	0.3197	0.4336	0.1431
315.10	1134.52	75.40	0.3197	0.4336	0.1424
320.09	1108.53	86.35	0.3187	0.4250	0.1336
320.10	1108.46	86.38	0.3186	0.4249	0.1338
320.10	1108.45	86.38	0.3186	0.4249	0.1338
320.11	1108.39	86.41	0.3186	0.4249	0.1338
325.06	1081.64	98.89	0.3176	0.4163	0.1249
325.06	1081.63	98.89	0.3176	0.4163	0.1249
325.07	1081.61	98.90	0.3176	0.4163	0.1246
325.07	1081.60	98.90	0.3176	0.4163	0.1251
330.05	1052.71	113.36	0.3165	0.4075	0.1154
330.05	1052.69	113.37	0.3165	0.4075	0.1154
330.05	1052.69	113.37	0.3165	0.4075	0.1159
330.05	1052.68	113.37	0.3165	0.4075	0.1154
335.08	1021.01	130.58	0.3154	0.3986	0.1067
335.10	1020.84	130.67	0.3154	0.3986	0.1069
335.11	1020.79	130.71	0.3154	0.3985	0.1071
340.08	986.29	150.67	0.3143	0.3897	0.0979
340.08	986.27	150.69	0.3143	0.3897	0.0979
340.09	986.26	150.69	0.3143	0.3897	0.0979
340.09	986.26	150.70	0.3143	0.3897	0.0983
345.11	946.91	175.43	0.3132	0.3806	0.0888
345.11	946.89	175.45	0.3132	0.3806	0.0891
345.11	946.87	175.46	0.3132	0.3806	0.0895
345.11	946.86	175.47	0.3132	0.3805	0.0886

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities and compositions were obtained from REFPROP, version 6.01, by flash calculations for the measured mixture bulk density  $\rho = 387.13$  kg m<sup>-3</sup>.

**Table 8. Saturated-Liquid Viscosities for Mixture DoE-6 (0.7 R125 + 0.3 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$W_{l,R125}$	$W_{v,R125}$	$\eta$ (mPa s)
248.93	1413.73	14.37	0.7294	0.8638	0.3125
248.89	1413.89	14.35	0.7286	0.8639	0.3124
248.85	1414.01	14.33	0.7286	0.8639	0.3117
249.32	1412.39	14.58	0.7285	0.8634	0.3113
250.14	1409.44	15.03	0.7283	0.8626	0.3073
250.14	1409.43	15.03	0.7283	0.8626	0.3085
250.14	1409.42	15.03	0.7283	0.8626	0.3078
250.14	1409.43	15.03	0.7283	0.8626	0.3066
255.12	1390.51	18.03	0.7274	0.8577	0.2870
255.13	1390.50	18.04	0.7274	0.8577	0.2878
255.13	1390.50	18.04	0.7274	0.8577	0.2903
255.13	1390.49	18.04	0.7274	0.8577	0.2915
260.08	1371.68	21.45	0.7264	0.8528	0.2677
260.08	1371.70	21.45	0.7264	0.8528	0.2689
260.08	1371.70	21.45	0.7264	0.8528	0.2691
260.08	1371.69	21.45	0.7264	0.8528	0.2686
265.10	1352.59	25.44	0.7255	0.8478	0.2504
265.10	1352.59	25.44	0.7255	0.8478	0.2508
265.10	1352.58	25.44	0.7255	0.8478	0.2511
265.10	1352.60	25.43	0.7255	0.8478	0.2513
270.09	1332.60	29.94	0.7245	0.8429	0.2320
270.09	1332.60	29.94	0.7245	0.8429	0.2315
270.09	1332.60	29.94	0.7245	0.8429	0.2323
270.09	1332.60	29.94	0.7245	0.8429	0.2327
275.10	1311.60	35.12	0.7235	0.8379	0.2172
275.11	1311.58	35.13	0.7235	0.8378	0.2183
275.10	1311.60	35.12	0.7235	0.8379	0.2202
275.11	1311.58	35.13	0.7235	0.8378	0.2164
280.09	1290.58	40.98	0.7226	0.8328	0.2047
280.09	1290.59	40.98	0.7226	0.8328	0.2056
280.09	1290.59	40.98	0.7226	0.8328	0.2052
280.10	1290.57	40.98	0.7226	0.8328	0.2047
285.13	1267.94	47.72	0.7216	0.8276	0.1932
285.13	1267.94	47.72	0.7216	0.8276	0.1912
285.13	1267.94	47.72	0.7216	0.8276	0.1912
285.12	1267.94	47.72	0.7216	0.8276	0.1917
285.12	1267.94	47.72	0.7216	0.8276	0.1917
290.06	1245.73	55.17	0.7206	0.8223	0.1802
290.06	1245.72	55.17	0.7206	0.8223	0.1817
290.04	1245.79	55.15	0.7206	0.8223	0.1787
290.04	1245.80	55.15	0.7206	0.8223	0.1825
295.10	1221.04	63.90	0.7197	0.8168	0.1663
295.10	1221.04	63.90	0.7197	0.8168	0.1664
295.11	1221.02	63.91	0.7197	0.8168	0.1678
295.10	1221.03	63.91	0.7197	0.8168	0.1686
300.08	1195.59	73.66	0.7187	0.8112	0.1562
300.08	1195.61	73.66	0.7187	0.8112	0.1562
300.08	1195.60	73.66	0.7187	0.8112	0.1561
300.08	1195.60	73.66	0.7187	0.8112	0.1551
305.09	1168.99	84.94	0.7177	0.8053	0.1458
305.10	1168.96	84.95	0.7177	0.8053	0.1463
305.10	1168.98	84.94	0.7177	0.8053	0.1452
305.09	1168.98	84.94	0.7177	0.8053	0.1455
310.11	1141.35	97.85	0.7168	0.7992	0.1344
310.11	1141.37	97.84	0.7168	0.7992	0.1350
310.11	1141.35	97.85	0.7168	0.7992	0.1352
310.11	1141.36	97.84	0.7168	0.7992	0.1360
315.11	1110.76	112.77	0.7158	0.7929	0.1254
315.12	1110.75	112.77	0.7158	0.7929	0.1254
315.11	1110.76	112.77	0.7158	0.7929	0.1257
315.12	1110.73	112.78	0.7158	0.7929	0.1254
320.07	1078.53	129.96	0.7148	0.7863	0.1154
320.07	1078.52	129.97	0.7148	0.7863	0.1150
320.07	1078.54	129.96	0.7148	0.7863	0.1154
320.07	1078.52	129.97	0.7148	0.7863	0.1155
325.16	1042.32	151.01	0.7139	0.7792	0.1060
325.16	1042.33	151.00	0.7139	0.7792	0.1065
325.16	1042.30	151.02	0.7139	0.7792	0.1060
325.16	1042.34	151.00	0.7139	0.7792	0.1061
330.12	1001.94	175.61	0.7129	0.7720	0.0964
330.10	1002.14	175.48	0.7129	0.7720	0.0965
330.10	1002.14	175.48	0.7129	0.7720	0.0967
330.11	1002.08	175.52	0.7129	0.7720	0.0965
335.14	955.82	206.81	0.7119	0.7643	0.0868
335.14	955.80	206.82	0.7119	0.7643	0.0864
335.14	955.80	206.82	0.7119	0.7643	0.0856
335.14	955.76	206.85	0.7119	0.7643	0.0880

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities and compositions were obtained from REFPROP, version 6.01, by flash calculations for the measured mixture bulk density  $\rho = 390.14$  kg m<sup>-3</sup>.



**Table 9. Saturated-Liquid Viscosities for Mixture DoE-7 (0.65 propane + 0.35 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

T (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{1,R290}$	$w_{v,R290}$	$\eta$ (mPa s)
246.16	781.50	8.27	0.4444	0.4366	0.1838
246.18	781.47	8.27	0.4444	0.4366	0.1842
246.19	781.44	8.27	0.4444	0.4366	0.1838
246.20	781.42	8.28	0.4444	0.4366	0.1837
250.09	774.33	9.45	0.4444	0.4347	0.1752
250.09	774.33	9.45	0.4444	0.4347	0.1742
250.09	774.33	9.45	0.4444	0.4347	0.1746
250.09	774.33	9.45	0.4444	0.4347	0.1751
254.96	765.28	11.10	0.4444	0.4325	0.1652
254.96	765.28	11.11	0.4444	0.4325	0.1640
254.96	765.27	11.11	0.4444	0.4325	0.1650
254.96	765.27	11.11	0.4444	0.4325	0.1641
260.10	755.62	13.06	0.4444	0.4304	0.1549
260.10	755.61	13.06	0.4444	0.4304	0.1560
260.10	755.61	13.06	0.4444	0.4304	0.1551
260.10	755.61	13.06	0.4444	0.4304	0.1565
265.15	745.96	15.25	0.4444	0.4286	0.1462
265.15	745.96	15.25	0.4444	0.4286	0.1460
265.16	745.95	15.26	0.4444	0.4286	0.1457
265.16	745.95	15.26	0.4444	0.4286	0.1462
270.07	736.36	17.64	0.4444	0.4270	0.1371
270.07	736.36	17.64	0.4444	0.4270	0.1370
270.07	736.36	17.64	0.4444	0.4270	0.1365
270.07	736.36	17.64	0.4444	0.4270	0.1368
275.07	726.37	20.35	0.4444	0.4256	0.1295
275.07	726.36	20.35	0.4444	0.4256	0.1295
275.07	726.36	20.35	0.4444	0.4256	0.1293
275.07	726.36	20.35	0.4444	0.4256	0.1300
280.06	716.18	23.36	0.4444	0.4244	0.1218
280.06	716.18	23.36	0.4444	0.4244	0.1227
280.06	716.18	23.36	0.4444	0.4244	0.1224
280.06	716.18	23.36	0.4444	0.4244	0.1224
285.01	705.88	26.70	0.4444	0.4234	0.1158
285.01	705.88	26.70	0.4444	0.4234	0.1156
285.01	705.88	26.70	0.4444	0.4234	0.1158
285.01	705.88	26.70	0.4444	0.4234	0.1157
290.03	695.13	30.46	0.4444	0.4226	0.1097
290.03	695.13	30.46	0.4444	0.4226	0.1095
290.03	695.13	30.46	0.4444	0.4226	0.1094
290.03	695.13	30.46	0.4444	0.4226	0.1095
295.03	684.09	34.61	0.4444	0.4220	0.1030
295.03	684.09	34.61	0.4444	0.4220	0.1030
295.03	684.09	34.61	0.4444	0.4220	0.1031
295.03	684.08	34.61	0.4444	0.4220	0.1031
300.10	672.56	39.29	0.4444	0.4216	0.0974
300.11	672.55	39.29	0.4444	0.4216	0.0970
300.11	672.55	39.29	0.4444	0.4216	0.0974
300.11	672.55	39.30	0.4444	0.4216	0.0974
305.09	660.79	44.41	0.4444	0.4213	0.0914
305.09	660.79	44.41	0.4444	0.4213	0.0921
305.09	660.79	44.41	0.4444	0.4213	0.0918
305.09	660.79	44.41	0.4444	0.4213	0.0921
310.07	648.72	50.07	0.4444	0.4212	0.0868
310.07	648.72	50.07	0.4444	0.4212	0.0868
310.07	648.72	50.07	0.4444	0.4212	0.0868
310.08	648.71	50.07	0.4444	0.4212	0.0870
315.07	636.07	56.38	0.4444	0.4213	0.0821
315.07	636.07	56.38	0.4444	0.4213	0.0822
315.07	636.06	56.38	0.4444	0.4213	0.0822
315.08	636.06	56.38	0.4444	0.4213	0.0823
320.00	623.09	63.27	0.4444	0.4216	0.0778
320.00	623.09	63.27	0.4444	0.4216	0.0778
320.01	623.08	63.28	0.4444	0.4216	0.0778
320.01	623.08	63.28	0.4444	0.4216	0.0778
325.12	608.96	71.29	0.4444	0.4220	0.0734
325.12	608.96	71.29	0.4444	0.4220	0.0735
325.12	608.96	71.29	0.4444	0.4220	0.0735
325.13	608.95	71.30	0.4444	0.4220	0.0735
330.09	594.54	79.94	0.4444	0.4226	0.0698
330.09	594.54	79.94	0.4444	0.4226	0.0696
330.09	594.54	79.95	0.4444	0.4226	0.0695
330.09	594.53	79.95	0.4444	0.4226	0.0698
335.09	579.17	89.76	0.4444	0.4234	0.0656
335.09	579.17	89.76	0.4444	0.4234	0.0657
335.09	579.16	89.76	0.4444	0.4234	0.0657
335.09	579.16	89.76	0.4444	0.4234	0.0657

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities were calculated with REFPROP, version 6.01, at each temperature for the constant saturated-liquid composition of mass fraction  $w_{1,R290} = 0.4444$ . Flash calculations for the measured bulk density  $\rho = 243.1$  kg m<sup>-3</sup> did not converge.

**Table 10. Saturated-Liquid Viscosities for Mixture DoE-8 (0.35 propane + 0.65 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

T (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{1,R290}$	$w_{v,R290}$	$\eta$ (mPa s)
245.91	1006.18	10.46	0.1894	0.4396	0.2371
245.91	1006.18	10.46	0.1894	0.4396	0.2366
245.92	1006.17	10.46	0.1894	0.4396	0.2363
245.92	1006.16	10.46	0.1894	0.4396	0.2362
250.08	995.80	11.97	0.1894	0.4261	0.2220
250.09	995.79	11.97	0.1894	0.4261	0.2242
250.09	995.79	11.97	0.1894	0.4261	0.2230
250.09	995.78	11.98	0.1894	0.4261	0.2224
255.08	983.81	14.01	0.1894	0.4108	0.2084
255.08	983.80	14.02	0.1894	0.4108	0.2100
255.08	983.80	14.02	0.1894	0.4108	0.2091
255.09	983.79	14.02	0.1894	0.4108	0.2097
255.09	983.79	14.02	0.1894	0.4108	0.2097
260.08	971.60	16.31	0.1894	0.3963	0.1966
260.08	971.60	16.31	0.1894	0.3963	0.1965
260.08	971.60	16.31	0.1894	0.3963	0.1965
260.08	971.60	16.31	0.1894	0.3963	0.1954
265.06	959.25	18.89	0.1894	0.3826	0.1845
265.06	959.25	18.89	0.1894	0.3826	0.1842
265.06	959.24	18.89	0.1894	0.3826	0.1837
265.06	959.24	18.89	0.1894	0.3826	0.1855
270.07	946.61	21.79	0.1894	0.3696	0.1730
270.07	946.61	21.79	0.1894	0.3696	0.1724
270.08	946.61	21.79	0.1894	0.3696	0.1729
270.08	946.61	21.79	0.1894	0.3696	0.1728
275.07	933.82	25.03	0.1894	0.3574	0.1621
275.07	933.82	25.03	0.1894	0.3574	0.1625
275.07	933.82	25.03	0.1894	0.3574	0.1626
275.07	933.82	25.03	0.1894	0.3574	0.1620
280.09	920.57	28.66	0.1894	0.3457	0.1525
280.09	920.57	28.66	0.1894	0.3457	0.1522
280.09	920.57	28.66	0.1894	0.3457	0.1527
280.09	920.57	28.66	0.1894	0.3457	0.1523
285.06	907.24	32.68	0.1894	0.3349	0.1433
285.06	907.24	32.68	0.1894	0.3349	0.1438
285.06	907.24	32.68	0.1894	0.3348	0.1436
285.06	907.24	32.68	0.1894	0.3348	0.1433
290.08	893.37	37.17	0.1894	0.3244	0.1369
290.08	893.37	37.17	0.1894	0.3244	0.1364
290.08	893.37	37.17	0.1894	0.3244	0.1361
290.08	893.37	37.17	0.1894	0.3244	0.1367
295.03	879.37	42.11	0.1894	0.3147	0.1278
295.03	879.37	42.11	0.1894	0.3147	0.1277
295.03	879.37	42.11	0.1894	0.3147	0.1282
295.03	879.37	42.11	0.1894	0.3147	0.1278
300.05	864.76	47.66	0.1894	0.3054	0.1202
300.05	864.75	47.66	0.1894	0.3054	0.1204
300.05	864.75	47.66	0.1894	0.3054	0.1211
300.05	864.75	47.66	0.1894	0.3054	0.1207
305.06	849.66	53.83	0.1894	0.2965	0.1131
305.07	849.65	53.83	0.1894	0.2965	0.1134
305.07	849.65	53.83	0.1894	0.2965	0.1132
305.07	849.65	53.83	0.1894	0.2965	0.1134
310.03	834.20	60.60	0.1894	0.2881	0.1073
310.03	834.20	60.60	0.1894	0.2881	0.1072
310.03	834.20	60.60	0.1894	0.2881	0.1075
310.03	834.19	60.60	0.1894	0.2881	0.1072
315.05	817.93	68.22	0.1894	0.2801	0.1005
315.05	817.92	68.23	0.1894	0.2801	0.1005
315.06	817.92	68.23	0.1894	0.2801	0.1006
315.06	817.92	68.23	0.1894	0.2801	0.1003
320.07	801.07	76.67	0.1894	0.2724	0.0950
320.07	801.05	76.68	0.1894	0.2724	0.0949
320.07	801.05	76.68	0.1894	0.2724	0.0952
320.08	801.03	76.69	0.1894	0.2724	0.0953
325.10	783.44	86.18	0.1894	0.2650	0.0882
325.11	783.42	86.18	0.1894	0.2650	0.0884
325.11	783.42	86.19	0.1894	0.2650	0.0889
325.11	783.42	86.19	0.1894	0.2650	0.0884
330.09	764.96	96.66	0.1894	0.2579	0.0831
330.09	764.96	96.66	0.1894	0.2579	0.0830
330.09	764.96	96.66	0.1894	0.2579	0.0831
330.09	764.95	96.67	0.1894	0.2579	0.0832
335.17	745.10	108.73	0.1894	0.2509	0.0781
335.17	745.10	108.74	0.1894	0.2509	0.0777
335.17	745.10	108.74	0.1894	0.2509	0.0778
335.17	745.10	108.74	0.1894	0.2509	0.0781
340.08	724.86	121.83	0.1894	0.2443	0.0736
340.08	724.85	121.83	0.1894	0.2443	0.0734
340.08	724.85	121.83	0.1894	0.2443	0.0732
340.08	724.84	121.84	0.1894	0.2443	0.0733
345.08	702.57	136.96	0.1894	0.2378	0.0691
345.08	702.56	136.97	0.1894	0.2378	0.0689
345.08	702.56	136.97	0.1894	0.2378	0.0692
345.09	702.55	136.98	0.1894	0.2378	0.0691

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities were calculated with REFPROP, version 6.01, at each temperature for the constant saturated-liquid composition of mass fraction  $w_{1,R290} = 0.1894$ . Flash calculations for the measured bulk density  $\rho = 314.1$  kg m<sup>-3</sup> did not converge.

**Table 11. Saturated-Liquid Viscosities for Mixture DoE-9 (0.33 R32 + 0.33 R125 + 0.34 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

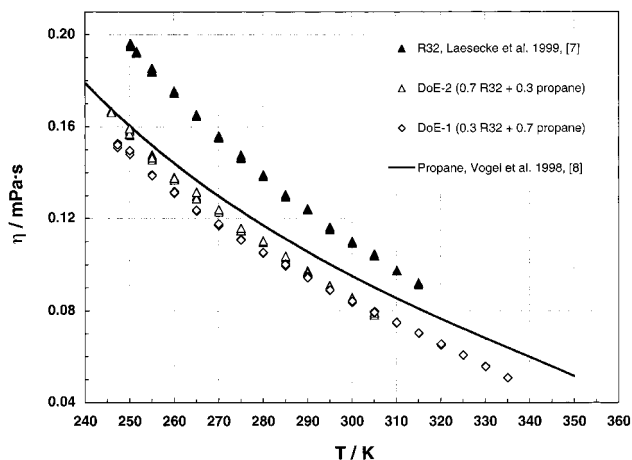
<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{l,R32}$	$w_{v,R32}$	$w_{l,R125}$	$w_{v,R125}$	$\eta$ (mPa s)
245.65	1353.04	10.16	0.1881	0.2768	0.4361	0.5400	0.2913
245.66	1353.03	10.16	0.1881	0.2768	0.4361	0.5400	0.2944
245.66	1353.01	10.16	0.1881	0.2768	0.4361	0.5400	0.2913
245.67	1353.00	10.16	0.1881	0.2768	0.4361	0.5400	0.2923
245.73	1352.82	10.18	0.1881	0.2768	0.4361	0.5400	0.2954
245.74	1352.78	10.19	0.1881	0.2768	0.4361	0.5400	0.2923
245.75	1352.77	10.19	0.1881	0.2768	0.4361	0.5400	0.2933
245.76	1352.72	10.20	0.1881	0.2768	0.4361	0.5399	0.2923
249.97	1338.09	11.98	0.1879	0.2745	0.4359	0.5368	0.2733
249.99	1338.03	11.99	0.1879	0.2745	0.4359	0.5368	0.2742
249.91	1338.32	11.95	0.1879	0.2745	0.4359	0.5369	0.2733
249.98	1338.08	11.98	0.1879	0.2745	0.4359	0.5368	0.2751
255.18	1319.88	14.51	0.1875	0.2717	0.4355	0.5330	0.2578
255.17	1319.91	14.51	0.1875	0.2717	0.4355	0.5330	0.2570
255.17	1319.89	14.51	0.1875	0.2717	0.4355	0.5330	0.2570
255.18	1319.88	14.51	0.1875	0.2717	0.4355	0.5330	0.2562
255.17	1319.92	14.51	0.1875	0.2717	0.4355	0.5330	0.2545
255.18	1319.89	14.51	0.1875	0.2717	0.4355	0.5330	0.2553
255.16	1319.96	14.50	0.1875	0.2717	0.4355	0.5331	0.2529
255.25	1319.64	14.55	0.1875	0.2717	0.4355	0.5330	0.2577
260.12	1302.58	17.28	0.1872	0.2691	0.4351	0.5295	0.2378
260.12	1302.59	17.28	0.1872	0.2691	0.4351	0.5295	0.2385
260.12	1302.60	17.28	0.1872	0.2691	0.4351	0.5295	0.2378
260.11	1302.61	17.28	0.1872	0.2691	0.4351	0.5295	0.2393
265.13	1284.50	20.53	0.1868	0.2664	0.4347	0.5258	0.2257
265.12	1284.52	20.52	0.1868	0.2664	0.4347	0.5258	0.2244
265.12	1284.53	20.52	0.1868	0.2664	0.4347	0.5258	0.2238
265.12	1284.52	20.52	0.1868	0.2664	0.4347	0.5258	0.2244
270.17	1265.40	24.26	0.1863	0.2636	0.4343	0.5222	0.2089
270.17	1265.39	24.26	0.1863	0.2636	0.4343	0.5222	0.2089
270.19	1265.35	24.27	0.1863	0.2636	0.4343	0.5222	0.2100
270.16	1265.44	24.25	0.1863	0.2636	0.4343	0.5222	0.2095
270.14	1265.50	24.24	0.1863	0.2636	0.4343	0.5222	0.2101
275.11	1247.11	28.44	0.1858	0.2608	0.4338	0.5186	0.1970
275.11	1247.12	28.44	0.1858	0.2608	0.4338	0.5186	0.1991
275.10	1247.14	28.43	0.1858	0.2608	0.4338	0.5186	0.1996
275.11	1247.13	28.44	0.1858	0.2608	0.4338	0.5186	0.1996
280.06	1227.75	33.20	0.1853	0.2580	0.4333	0.5149	0.1853
280.07	1227.72	33.20	0.1853	0.2580	0.4333	0.5149	0.1857
280.07	1227.71	33.21	0.1853	0.2580	0.4332	0.5149	0.1852
280.08	1227.68	33.21	0.1853	0.2580	0.4332	0.5149	0.1862
285.14	1206.87	38.77	0.1848	0.2550	0.4327	0.5111	0.1741
285.15	1206.84	38.78	0.1848	0.2550	0.4327	0.5111	0.1754
285.15	1206.85	38.77	0.1848	0.2550	0.4327	0.5111	0.1762
285.14	1206.87	38.77	0.1848	0.2550	0.4327	0.5111	0.1762
290.09	1186.58	44.94	0.1843	0.2519	0.4321	0.5074	0.1653
290.09	1186.58	44.94	0.1843	0.2519	0.4321	0.5074	0.1669
290.09	1186.58	44.94	0.1843	0.2519	0.4321	0.5074	0.1653
290.10	1186.57	44.94	0.1843	0.2519	0.4321	0.5074	0.1641
295.07	1164.18	52.01	0.1837	0.2488	0.4315	0.5035	0.1537
295.07	1164.18	52.01	0.1837	0.2488	0.4315	0.5035	0.1530
295.07	1164.19	52.01	0.1837	0.2488	0.4315	0.5035	0.1540
295.07	1164.18	52.01	0.1837	0.2488	0.4315	0.5035	0.1537
300.12	1141.45	60.16	0.1831	0.2454	0.4309	0.4995	0.1438
300.13	1141.44	60.16	0.1831	0.2454	0.4309	0.4995	0.1445
300.12	1141.48	60.15	0.1831	0.2454	0.4309	0.4995	0.1454
300.12	1141.45	60.16	0.1831	0.2454	0.4309	0.4995	0.1432
305.09	1117.58	69.33	0.1825	0.2419	0.4302	0.4954	0.1364
305.08	1117.59	69.33	0.1825	0.2419	0.4302	0.4954	0.1352
305.09	1117.56	69.34	0.1825	0.2419	0.4302	0.4954	0.1364
305.09	1117.57	69.33	0.1825	0.2419	0.4302	0.4954	0.1361
310.16	1092.14	80.03	0.1819	0.2382	0.4295	0.4911	0.1276
310.17	1092.06	80.06	0.1819	0.2382	0.4295	0.4911	0.1265
310.16	1092.11	80.05	0.1819	0.2382	0.4295	0.4911	0.1273
310.17	1092.08	80.06	0.1819	0.2382	0.4295	0.4911	0.1270
315.13	1065.23	92.15	0.1813	0.2343	0.4288	0.4867	0.1163
315.13	1065.24	92.15	0.1813	0.2343	0.4288	0.4867	0.1174
315.14	1065.19	92.17	0.1813	0.2343	0.4288	0.4867	0.1174
315.13	1065.24	92.15	0.1813	0.2343	0.4288	0.4867	0.1181
320.10	1036.43	106.10	0.1807	0.2302	0.4281	0.4822	0.1094
320.09	1036.45	106.09	0.1807	0.2302	0.4281	0.4822	0.1096
320.08	1036.50	106.07	0.1807	0.2302	0.4281	0.4822	0.1100
320.09	1036.47	106.08	0.1807	0.2302	0.4281	0.4822	0.1094
325.12	1005.70	122.74	0.1801	0.2258	0.4274	0.4774	0.1016
325.12	1005.74	122.72	0.1801	0.2258	0.4274	0.4774	0.1020
325.12	1005.72	122.73	0.1801	0.2258	0.4274	0.4774	0.1024
325.12	1005.71	122.74	0.1801	0.2258	0.4274	0.4774	0.1018
330.10	971.57	142.14	0.1795	0.2213	0.4267	0.4725	0.0935
330.10	971.59	142.13	0.1795	0.2213	0.4267	0.4725	0.0932
330.09	971.66	142.09	0.1795	0.2213	0.4267	0.4725	0.0936
330.10	971.57	142.14	0.1795	0.2213	0.4267	0.4725	0.0930

<sup>a</sup> The data are listed in the order of measurements. Liquid and vapor densities and compositions were obtained from REFPROP, version 6.01, by flash calculations for the measured mixture bulk density  $\rho = 401 \text{ kg m}^{-3}$ .

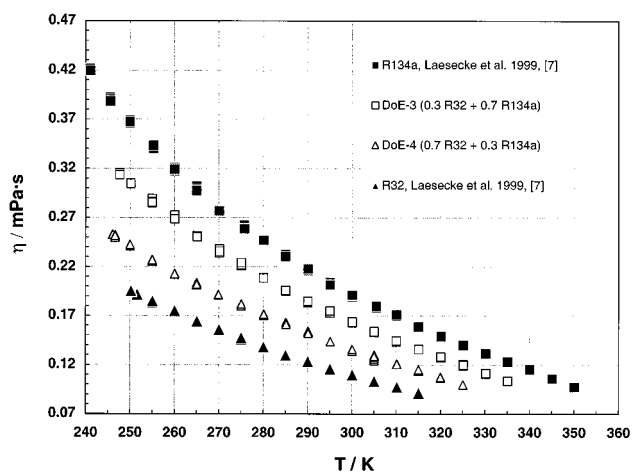
**Table 12. Saturated-Liquid Viscosities for Mixture DoE-10 (0.3 R32 + 0.1 R125 + 0.6 R134a) Measured in the Sealed Gravitational Capillary Viscometer<sup>a</sup>**

<i>T</i> (K)	$\rho_l$ (kg m <sup>-3</sup> )	$\rho_v$ (kg m <sup>-3</sup> )	$w_{l,R32}$	$w_{v,R32}$	$w_{l,R125}$	$w_{v,R125}$	$\eta$ (mPa s)
251.32	1322.03	9.07	0.1733	0.3284	0.1336	0.2212	0.2936
251.27	1322.19	9.05	0.1733	0.3284	0.1336	0.2212	0.2968
251.23	1322.32	9.03	0.1733	0.3285	0.1336	0.2212	0.2973
251.18	1322.47	9.02	0.1733	0.3285	0.1336	0.2213	0.2962
255.15	1309.55	10.47	0.1729	0.3234	0.1334	0.2182	0.2807
255.16	1309.53	10.48	0.1729	0.3234	0.1334	0.2182	0.2802
255.16	1309.53	10.48	0.1729	0.3234	0.1334	0.2182	0.2815
255.15	1309.54	10.47	0.1729	0.3234	0.1334	0.2182	0.2824
260.15	1293.49	12.57	0.1724	0.3170	0.1331	0.2143	0.2698
260.14	1293.50	12.57	0.1724	0.3170	0.1331	0.2143	0.2680
260.14	1293.51	12.56	0.1724	0.3171	0.1331	0.2144	0.2674
260.14	1293.50	12.57	0.1724	0.3170	0.1331	0.2143	0.2651
265.12	1277.07	14.97	0.1719	0.3108	0.1328	0.2106	0.2501
265.13	1277.06	14.97	0.1719	0.3108	0.1328	0.2106	0.2491
265.13	1277.06	14.97	0.1719	0.3108	0.1328	0.2106	0.2473
265.13	1277.05	14.97	0.1719	0.3108	0.1328	0.2106	0.2480
270.17	1259.48	17.77	0.1714	0.3046	0.1325	0.2069	0.2304
270.18	1259.47	17.77	0.1714	0.3046	0.1325	0.2069	0.2318
270.17	1259.48	17.77	0.1714	0.3046	0.1325	0.2069	0.2337
270.18	1259.46	17.77	0.1714	0.3046	0.1325	0.2069	0.2360
275.22	1242.74	20.98	0.1708	0.2985	0.1322	0.2032	0.2199
275.22	1242.73	20.98	0.1708	0.2985	0.1322	0.2032	0.2201
275.22	1242.73	20.98	0.1708	0.2985	0.1322	0.2032	0.2209
275.22	1242.73	20.98	0.1708	0.2985	0.1322	0.2032	0.2196
280.09	1224.69	24.50	0.1702	0.2926	0.1319	0.1997	0.2020
280.09	1224.70	24.50	0.1702	0.2926	0.1319	0.1997	0.2046
280.08	1224.71	24.49	0.1702	0.2926	0.1319	0.1997	0.2029
280.08	1224.71	24.50	0.1702	0.2926	0.1319	0.1997	0.2046
285.14	1206.46	28.67	0.1695	0.2866	0.1316	0.1961	0.1918
285.13	1206.46	28.67	0.1695	0.2866	0.1316	0.1961	0.1917
285.13	1206.47	28.67	0.1695	0.2866	0.1316	0.1961	0.1898
285.13	1206.47	28.67	0.1695	0.2866	0.1316	0.1961	0.1918
290.08	1187.70	33.29	0.1688	0.2807	0.1312	0.1927	0.1808
290.05	1187.80	33.26	0.1688	0.2807	0.1312	0.1927	0.1788
290.05	1187.81	33.26	0.1688	0.2807	0.1312	0.1927	0.1786
290.06	1187.77	33.27	0.1688	0.2807	0.1312	0.1927	0.1795
295.14	1168.00	38.68	0.1681	0.2747	0.1308	0.1891	0.1681
295.14	1168.00	38.68	0.1681	0.2747	0.1308	0.1891	0.1694
295.15	1167.99	38.69	0.1681	0.2747	0.1308	0.1891	0.1680
295.15	1167.99	38.68	0.1681	0.2747	0.1308	0.1891	0.1686
300.13	1147.48	44.69	0.1674	0.2687	0.1305	0.1857	0.1587
300.13	1147.47	44.69	0.1674	0.2687	0.1305	0.1857	0.1585
300.13	1147.47	44.69	0.1674	0.2687	0.1305	0.1857	0.1580
300.13	1147.48	44.69	0.1674	0.2687	0.1305	0.1857	0.1583
30							

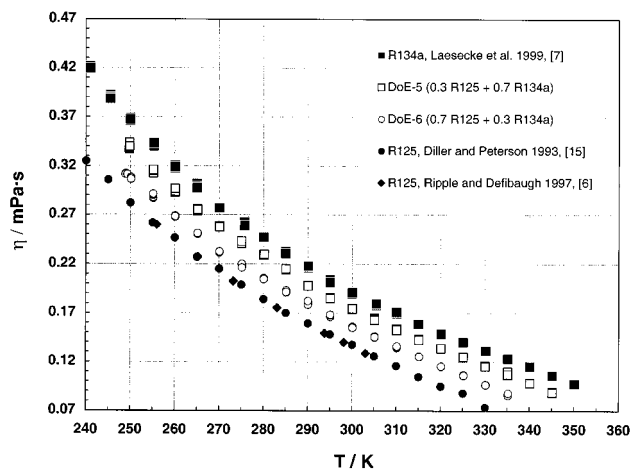




**Figure 2.** Temperature dependence of experimental saturated-liquid viscosities of the mixtures DoE-1 and DoE-2 (R32 + propane) in comparison with those of the pure fluids.

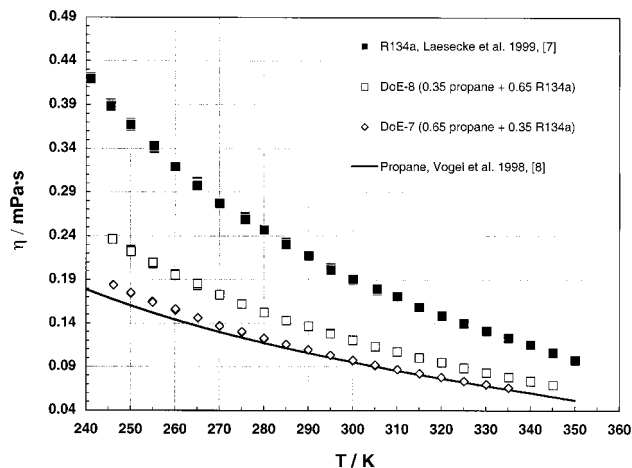


**Figure 3.** Temperature dependence of experimental saturated-liquid viscosities of the mixtures DoE-3 and DoE-4 (R32 + R134a) in comparison with those of the pure fluids.

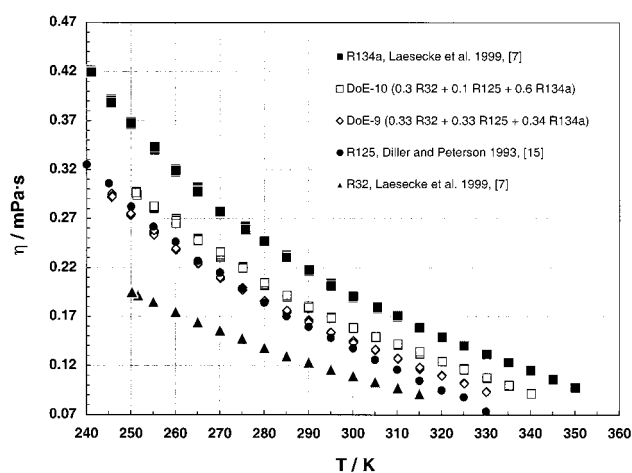


**Figure 4.** Temperature dependence of experimental saturated-liquid viscosities of the mixtures DoE-5 and DoE-6 (R125 + R134a) in comparison with those of the pure fluids.

and DoE-6). These compounds have the greatest similarity in their chemical structures because they differ by only a hydrogen/fluorine atom. Reference data for the viscosity of pure R125 are the measurements of Ripple and Defibaugh<sup>6</sup> with the present capillary viscometer and the saturated liquid data that Diller and Peterson<sup>15</sup> measured in the torsional-crystal viscometer. These data sets agree



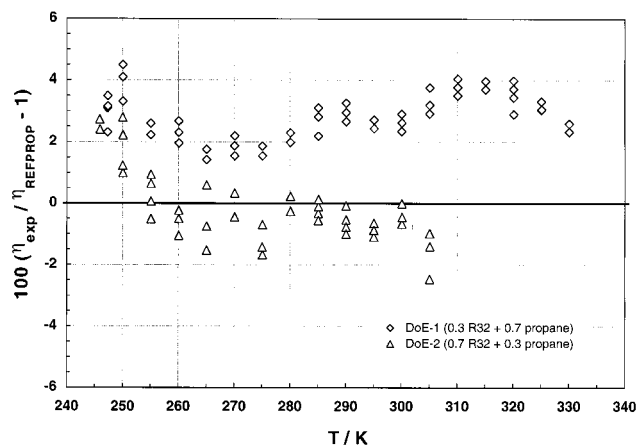
**Figure 5.** Temperature dependence of experimental saturated-liquid viscosities of the mixtures DoE-7 and DoE-8 (propane + R134a) in comparison with those of the pure fluids.



**Figure 6.** Temperature dependence of experimental saturated-liquid viscosities of the mixtures DoE-9 and DoE-10 (R32 + R125 + R134a) in comparison with those of the pure fluids.

well within their mutual estimated uncertainty. The viscosity of the system of propane and R134a shown in Figure 5 displays a nonlinear composition dependence whereby the addition of a mole fraction of 0.35 propane lowers the viscosity of DoE-8 dramatically from that of pure R134a, but the addition of another mole fraction of 0.30 propane to DoE-7 affects the viscosity much less. However, unlike the other nonpolar + polar system R32 + propane, the viscosity of the system propane + R134a does not exhibit a minimum at a composition between the pure fluids. Finally, Figure 6 puts the experimental viscosity data for the ternary mixtures DoE-9 and DoE-10 in context with the viscosities of the pure fluids R32, R125, and R134a. Both mixtures exhibit saturated-liquid viscosities that are close to that of pure R125. Because only two compositions have been measured, it is difficult to discern from Figure 6 whether the viscosity–composition dependence is linear, as in the binary hydrofluorocarbon mixtures, or nonlinear.

**Comparison with Model Predictions.** As pointed out in the preceding section, viscosity measurements of volatile mixtures require a more sophisticated framework for their interpretation than the conventional mixing rules. One successful method is the extended-corresponding-states (ECS) model, which includes the dependence of a system on pressure, temperature, and composition. The present experimental data were compared with values predicted



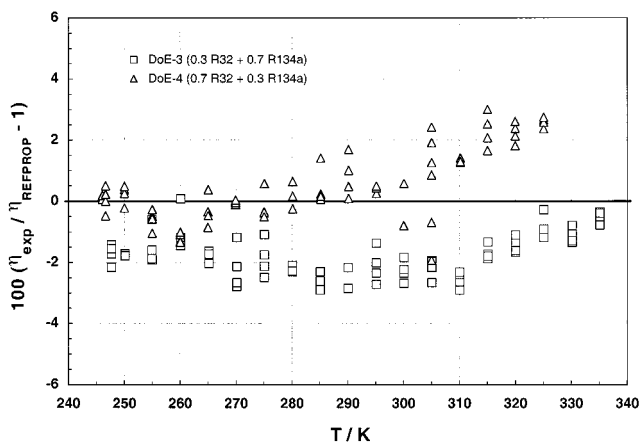
**Figure 7.** Comparison of experimental saturated-liquid viscosities of the mixtures DoE-1 and DoE-2 (R32 + propane) with predicted values from the extended corresponding states model in REFPROP, version 6.01.

with the ECS viscosity model of Klein et al.,<sup>14</sup> as implemented in REFPROP, version 6.01. The NIST ECS model does not account explicitly for electrostatic effects in polar fluids. Instead, *effective* shape factors are used to represent the combined effects of both steric and electrostatic interactions. The properties of a pure fluid or mixture are obtained by scaling with these shape factors from the properties of the polar reference fluid R134a. Consequently, predicted viscosities for the mixtures with the highest R134a concentrations are expected to agree best with the experimental data, e.g., DoE-3 (0.3 R32 + 0.7 R134a), DoE-5 (0.3 R125 + 0.7 R134a), and the ternary mixture DoE-10 (0.3 R32 + 0.1 R125 + 0.6 R134a).

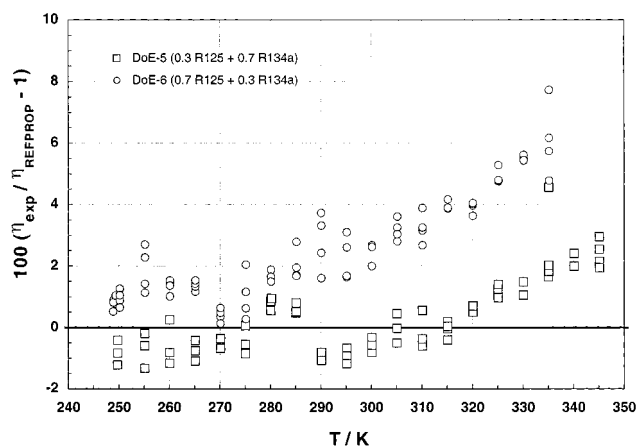
Fractional deviations between experimental and predicted viscosities for the five systems are illustrated in Figures 7–11. For the R32 + propane mixtures (DoE-1 and DoE-2), the agreement between experimental and predicted results is mostly within the estimated uncertainty of the measurements of  $\pm 2.8\%$  (Figure 7). However, there is a difference between the deviations for DoE-1 and DoE-2 that increases to about 5% at 305 K. The experimental viscosity data for the propane-rich mixture DoE-1 are systematically 3% higher than the predicted values, which can be rationalized considering that this mixture is most dissimilar (nonpolar) from the reference fluid R134a.

Similar deviation trends are observed for the mixtures of R32 + R134a (DoE-3 and DoE-4) (Figure 8). Most of the predicted viscosities agree with the experimental data within the estimated uncertainty of the latter (2.4%). However, the deviations for the two compositions DoE-3 and DoE-4 differ increasingly with temperature, up to about 3% at 325 K.

Surprisingly large and systematic deviations were obtained for the mixtures DoE-5 and DoE-6 consisting of R125 and R134a (Figure 9), although these molecules are very similar. In addition, R134a is the reference fluid for the ECS model. Although the data for both mixtures are predicted within their experimental uncertainty of  $\pm 2.2\%$  up to 280 K, a systematic increase in the deviations can be seen at higher temperatures, with a distinct difference between the two measured compositions. As before, the viscosity of the R134a-rich mixture DoE-5 is better predicted than that of the mixture DoE-6. This difference occurred also in the results for the two previously discussed mixtures, R32 + propane and R32 + R134a (Figures 7 and 8, respectively). This suggests that the ECS model does not correctly predict the composition dependence of the mixture



**Figure 8.** Comparison of experimental saturated-liquid viscosities of the mixtures DoE-3 and DoE-4 (R32 + R134a) with predicted values from the extended corresponding states model in REFPROP, version 6.01.



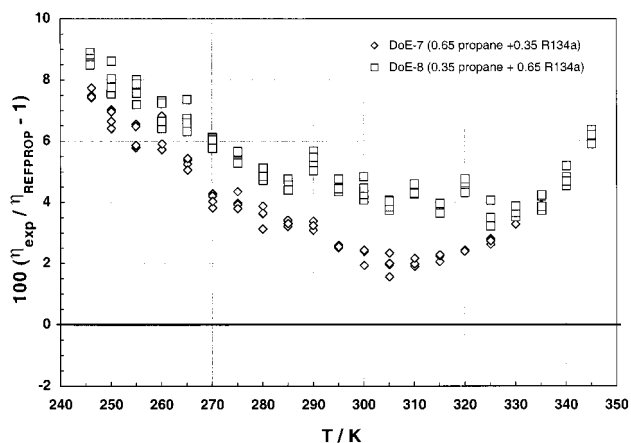
**Figure 9.** Comparison of experimental saturated-liquid viscosities of the mixtures DoE-5 and DoE-6 (R125 + R134a) with predicted values from the extended corresponding states model in REFPROP, version 6.01.

viscosity. The consistently higher deviations at higher temperatures could be caused by inadequacies in the viscosity correlation for the reference fluid R134a.<sup>14</sup>

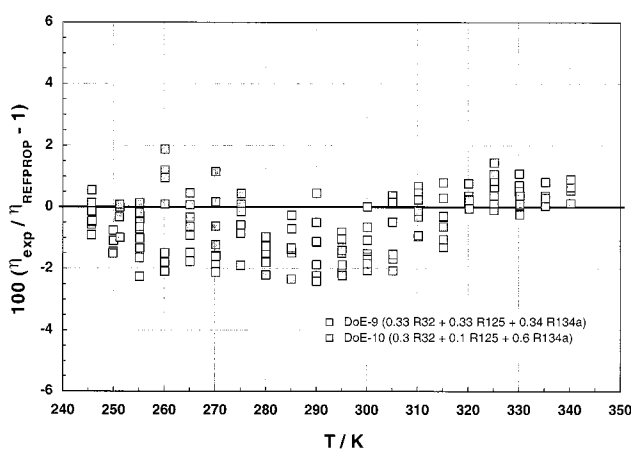
By far the highest deviations between experimental and predicted viscosities were found in the two mixtures of propane with R134a, DoE-7 and DoE-8 (Figure 10). They range from about 8.5% for DoE-8 at 245 K to 2% for DoE-7 at 305 K, whereas the experimental uncertainty was estimated to be 2.6% at 345 K. In addition, the deviations show a pronounced upturn at this temperature, which might be caused by the reference fluid's viscosity correlation. The mutual agreement between the results for the two compositions is within the experimental uncertainty. Interestingly, the viscosity of the R134a-rich mixture DoE-8 is predicted worse than that of the propane-rich mixture DoE-7.

The best agreement between measured and predicted viscosities, with respect to both composition and temperature dependence, was obtained for the ternary mixtures DoE-9 and DoE-10 (Figure 11). The deviations are within the experimental uncertainty of  $\pm 2.4\%$  over the entire temperature range that was measured, and systematic deviations are minor.

To put these results in perspective, it should be mentioned that the ECS model in REFPROP evolved during the measurements. Although the underlying physical



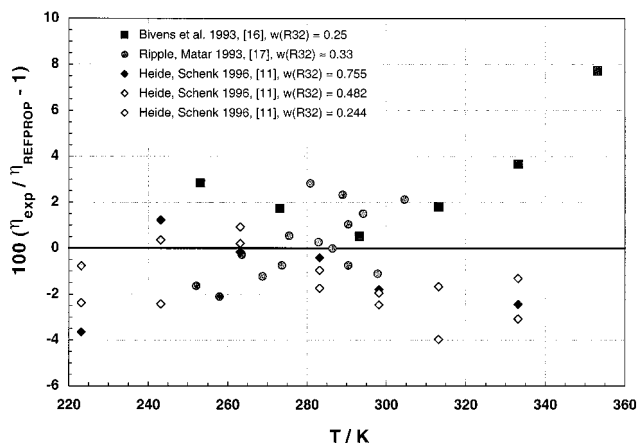
**Figure 10.** Comparison of experimental saturated-liquid viscosities of the mixtures DoE-7 and DoE-8 (propane + R134a) with predicted values from the extended corresponding states model in REFPROP, version 6.01.



**Figure 11.** Comparison of experimental saturated-liquid viscosities of the mixtures DoE-9 and DoE-10 (R32 + R125 + R134a) with predicted values from the extended corresponding states model in REFPROP, version 6.01.

model remained unchanged, significant improvements resulted from more reliable density predictions due to advances in the thermodynamic mixture models and from revised correlations for the viscosity of the reference fluid R134a. Comparisons of the measured viscosities of the propane mixtures with earlier versions of the ECS model showed predicted viscosities up to 20% higher. The model improvements are indicated by the fact that the deviations for these mixtures were reduced to a maximum of 8.5% with the current version (Figure 10). This is another reason that suggests that the latest version of REFPROP should be used for accurate predictions of the transport properties of refrigerant mixtures.

**Comparison with Literature Data.** The viscosity of some of the present refrigerant mixtures has been studied in three other published reports. Bivens et al.<sup>16</sup> used a constant-flow-rate capillary viscometer with a mercury pump to measure pure hydrofluorocarbons, as well as binary and ternary mixtures. An uncertainty of  $\pm 1.2\%$  was quoted for these experimental results. Ripple and Matar<sup>17</sup> measured pure hydrofluorocarbons and binary mixtures in the precursor of the sealed gravitational capillary viscometer that was used in the present work. The earlier instrument had a coiled capillary of 0.508 mm internal diameter vs a straight vertical capillary with 0.236 mm internal diameter in the newer one, so that the liquid flow was faster and subject to a radial acceleration in the coil.



**Figure 12.** Deviations of experimental literature viscosities for mixtures of R32 + R134a from values predicted with REFPROP, version 6.01. Compositions in the legend are mass fractions.

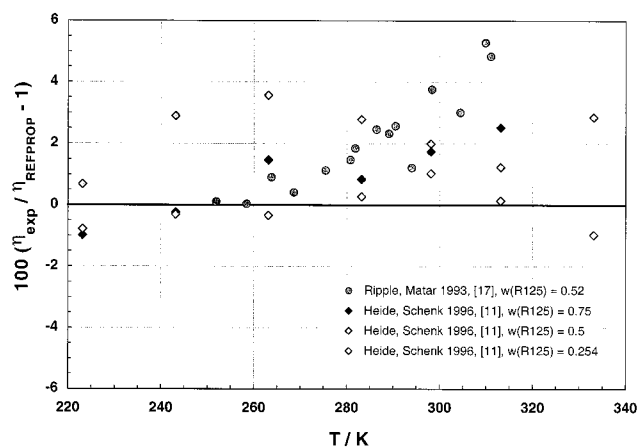
The correction for this radial acceleration was suspected to be inadequate by Ripple and Matar, which prompted them to quote an uncertainty of  $\pm 5\%$  for the measured viscosities. The inadequacy of the literature radial acceleration correction has been confirmed subsequently by new measurements of R32 and R134a in the viscometer with the straight capillary.<sup>7</sup> It was found that the literature correction was only about 95% effective, resulting in viscosities about 5% too high. This should be kept in mind in the following comparisons.

The most recent study was carried out by Heide and Schenk<sup>11</sup> within a project sponsored by the German Research Council "Refrigeration Technology", a nonprofit research organization of refrigeration companies. The measurements of Heide and Schenk were carried out in a rolling-ball viscometer and included five pure HFCs, and seven binary and two ternary mixtures. The uncertainty of the measurements was quoted as  $\pm 2\%$  at a viscosity of 0.2 mPa s. The mixture compositions of the literature data are shown in Figure 1, together with those that were measured in this work.

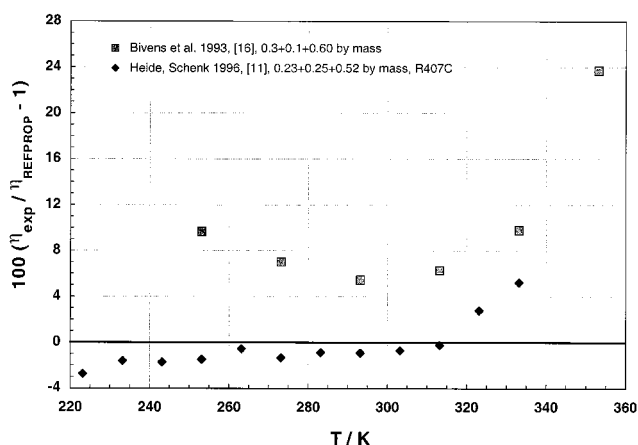
Fractional deviations between measurements and predictions with REFPROP 6.01 are illustrated in Figure 12 for binary mixtures of R32 and R134a. The data of Bivens et al.<sup>16</sup> are higher than the predicted viscosities by 2–8%, with the deviations systematically increasing with temperature. The data of Ripple and Matar<sup>17</sup> are represented within their quoted experimental uncertainty. However, the deviations increase systematically from  $-2\%$  at 252 K to 2% at 304 K. This trend is likely to be due to the inadequate radial acceleration correction for the flow in a curved tube that was applied to these measurements. The experimental results of Heide and Schenk<sup>11</sup> for three binary mixture concentrations are on the average 1.4% lower than the predicted viscosities. These data, as well as those of Ripple and Matar, agree with the present results within their estimated experimental uncertainties.

The second binary system for which literature data are available involves the pure components R125 and R134a, corresponding to the mixtures DoE-5 and DoE-6 in this work. Figure 13 shows that the measurements of Ripple and Matar differ from the predicted viscosities systematically with temperature, with the difference reaching 5.3% at 310 K. The measurements of Heide and Schenk<sup>11</sup> do not show this trend with increasing temperature but again exhibit considerable internal scatter. However, it is believed that the systematic deviation trend of the data of Ripple and Matar is not caused by the inadequate former





**Figure 13.** Deviations of experimental literature viscosities for mixtures of R125 + R134a from values predicted with REFPROP, version 6.01. Compositions in the legend are mass fractions.



**Figure 14.** Deviations of experimental literature viscosities for mixtures of R32 + R125 + R134a from values predicted with REFPROP, version 6.01. Compositions in the legend are mass fractions.

radial acceleration correction. These mixtures have the highest viscosities because the molecules of the pure compounds R125 and R134a are of considerable size and polarity. The measurements of Ripple and Matar, therefore, encompassed lower Reynolds numbers, for which radial acceleration contributes less than 5% to the total viscosity. Furthermore, the measurements with the straight capillary viscometer in this work exhibit similar deviation trends (Figure 9), although they are not influenced by radial acceleration. This supports the previous assumption that these deviations might be due to the viscosity correlation for the reference fluid R134a in the extended-corresponding-states model.

Viscosity measurements for the ternary system R32 + R125 + R134a were reported by Bivens et al.<sup>16</sup> and by Heide and Schenk.<sup>11</sup> The deviations between these results and the predicted viscosities are shown in Figure 14. Substantial deviations between 5 and 24% occur for the data of Bivens et al. As similar deviation patterns have been found in comparisons<sup>7</sup> of other data, which Bivens et al. reported for pure fluids, binary, and ternary mixtures, a systematic error in the experimental work of these authors cannot be excluded. Their results should not be used to fit parameters in predictive mixture viscosity models. The agreement between the experimental data of Heide and Schenk<sup>11</sup> and the predicted viscosities is within their quoted experimental uncertainty. Exceptions occur at the two highest temperatures above 315 K, where the

deviations increase to more than 5%. Thus, these literature data are largely consistent with the results of this work. However, as seen in Figure 11, the present data for ternary mixtures of R32 + R125 + R134a are matched by the predicted viscosities within their experimental uncertainty, even up to 340 K.

## Conclusions

Experimental viscosity data for 10 binary and ternary mixtures of propane and hydrofluorocarbons are presented. Because the mixtures are volatile, a sealed gravitational capillary viscometer was used for the measurements. Measurements in such sealed viscometers require a different methodology than measurements of nonvolatile mixtures in open capillary viscometers. In particular, the conversion of the measured volumetric flow rates into viscosities, kinematic or absolute, is very sensitive to the uncertainty in the saturated vapor and liquid densities of the mixtures. There are large gaps in viscosity data for volatile liquids that usually are small molecules and often first members of homologous series. Viscosity measurements for such liquids are very desirable for theoretical reasons. However, they should always be planned and conducted together with density measurements, unless reliable density data are already available.

The measured viscosity data fall into two groups. Systems that contain only hydrofluorocarbons exhibit linear viscosity–composition dependences. Nonlinear variations of the viscosity with mole fraction were observed in the two systems of propane with hydrofluorocarbons.

The experimental viscosity data are compared with predictions from an extended-corresponding-states model that employs the viscosity of R134a as a reference. The agreement is mostly within the estimated experimental uncertainty ( $\pm 2.8\%$  max.). However, deviations of up to 6% between experiment and prediction occur for the mixture 0.7 R125 + 0.3 R134a. Deviations of up to 8% are noted for both mixtures of propane + R134a. These deviations are presumably caused by inadequacies in the viscosity correlation for the reference fluid R134a and not by experimental errors.

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## Supporting Information Available:

The data of Tables 3–12 are available in electronic format via anonymous ftp at URL [ftp://ftp.boulder.nist.gov/pub/fluids/NIST\\_Data/Viscosity/Capillary](ftp://ftp.boulder.nist.gov/pub/fluids/NIST_Data/Viscosity/Capillary).

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