Reference Correlation for the Viscosity of Difluoromethane (R-32) from the Triple Point to 425 K and up to 70 MPa

Danai Velliadou,[1,a] Konstantinos D. Antoniadis,[1] Marc J. Assael,[1] and Marcia L. Huber[2]

[1] Laboratory of Thermophysical Properties and Environmental Processes, Chemical Engineering Department, Aristotle University, Thessaloniki 54636, Greece

[2] Applied Chemicals and Materials Division, National Institute of Standards and Technology, 325 Broadway, Boulder, CO 80305, USA

We present a wide-ranging correlation expressed in terms of temperature and density for the viscosity of difluoromethane (R-32) based on critically evaluated experimental data. The correlation is designed to be used with an existing equation of state from the triple point to 425 K and at pressures up to 70 MPa. Comparisons with experimental data over the temperature range 220 to 425 K at pressures up to 70 MPa indicate the correlation has an estimated uncertainty (at the 95% confidence level) of 3.4% in this region, and 2% for the vapor at atmospheric pressure. The correlation also behaves in a physically reasonable manner and may be extrapolated up to 350 MPa, but caution is advised in this region since it requires extrapolation of the equation of state.

1 Introduction

Difluoromethane (IUPAC name), also commonly known as R-32 or HFC-32, CAS 75-10-5, is a hydrofluorocarbon (HFC) refrigerant developed to replace ozone-depleting hydrochlorofluorocarbon (HCFC) predecessors, such as chlorodifluoromethane (R-22). Due to its lack of chlorine, it has an ozone-depletion potential of zero, initially making it an attractive fluid on its own and as a component in widely used refrigerant blends such as R-410A and R-407C. However, it has a relatively high global warming potential (GWP, 100-year) of 677 [1], and concerns about climate change may lead to its future use as a component in new blends, such as R-454B, R-454C, R-459A, and R-459B [2] that contain low-GWP hydrofluorinated olefins (HFO's) mixed with R-32.

Although in the literature there are many viscosity correlations that cover the liquid phase, such as the hard-spheres correlation of Assael et al. [3], there are very few that cover the liquid, the vapor, the critical and supercritical regions. The first three such correlations for the viscosity of difluoromethane covering all those regions are summarized below,

-   In 1997, Klein et al. [4] proposed a correlation covering a temperature range 231 to 343 K, and pressures up to 16 MPa, with a 4% uncertainty, based on theory and on the measurements of Assael et al. [5], Oliveira and Wakeham [6], and Ripple and Matar [7] – see also Table 1.
-   Kiselev et al. [8] in 1999, proposed a very similar correlation covering a temperature range 231 to 423 K, and pressures up to 10 MPa, based on the measurements of Takahashi et al. [9], Oliveira and Wakeham [6], and Ripple and Matar [7].
-   The third correlation, which is the viscosity correlation currently employed in REFPROP [10], is an unpublished work based on an extended corresponding-states model described by Huber et al. [11] in 2003 with parameters based in part on the data of Takahashi et al. [9], Bivens et al. [12], Oliveira and Wakeham [6], and Laesecke et al. [13]. The correlation is valid over temperatures from 137 to 435 K and pressures up to 10 MPa.

Chronologically, we note three more correlations:

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[a] Author to whom correspondence should be addressed (danai.velliadou@outlook.com)

- In 2007, Wang et al. [14] proposed a viscosity correlation for difluoromethane using the friction theory, and based upon the measurements of Takahashi et al. [9], Assael et al. [5], Dunlop [15], Oliveira and Wakeham [6], Laesecke et al. [13], and Sun et al. [16]. The correlation is valid in the temperature range 232 to 423 K and up to 15 MPa pressure with an uncertainty of about 2.5%.
- In 2014 a viscosity correlation for "engineering calculations" was published by Boychuk [17]. The correlation was based upon the data of Oliveira and Wakeham [6, 18], Geller et al. [19], Takahashi et al. [9], Assael et al. [5], Dunlop [15], Fröba et al. [20], Laesecke et al. [13], Sun et al. [16], and Ripple and Matar [7]. It employed an equation of state by Vasserman and Fomisky [21], and it is valid from 232 to 423 K and up to 10 MPa with an expanded ($k=2$) uncertainty of 2.5%.
- Finally in 2015, He et al. [22] proposed a viscosity correlation based on the free-volume theory and the measurements of Oliveira and Wakeham [18], Assael et al. [5], Fröba et al. [20], Laesecke et al. [13], and Sun et al. [16]. The correlation is valid in the temperature range 232 to 423 K and up to a pressure of 15 MPa.

All six aforementioned correlations are restricted to pressures up to 16 MPa, a limit imposed by the availability of viscosity measurements up to that pressure. However, since the development of these correlations, two additional sets of measurements have been published at higher pressures. In 2017, Bair and Laesecke [23] published viscosity measurements at significantly higher pressures, up to 350 MPa. These measurements were performed in a falling-cylinder viscometer with an uncertainty of 3-6% (at the 95% confidence level). Furthermore, Meng et al. [24] in 2018, published viscosity measurements up to 30 MPa, in a vibrating-wire instrument with a 2% uncertainty (at the 95% confidence level). Hence, there is currently a need for an up-to-date, wide-ranging reference correlation for the viscosity of difluoromethane.

In a series of recent papers, reference correlations for the viscosity of common fluids [25-36] were developed that cover a wide range of temperature and pressure conditions, including the gas, liquid, and supercritical phases. In this paper, the methodology adopted in the aforementioned papers is extended to developing a new reference correlation for the viscosity of difluoromethane. The analysis we use is based on the best available experimental viscosity data. A prerequisite to the analysis is a critical assessment of the experimental data. Here we define two categories of experimental data: primary data, employed in the development of the correlation, and secondary data, used simply for comparison purposes. According to the recommendation adopted by the Subcommittee on Transport Properties (now known as The International Association for Transport Properties) of the International Union of Pure and Applied Chemistry, the primary data are identified by a well-established set of criteria [37]. These criteria have been successfully employed to establish standard reference values for the viscosity and thermal conductivity of fluids over wide ranges of conditions, with uncertainties in the range of 1%. However, in many cases, such a narrow definition unacceptably limits the range of the data representation. Consequently, within the primary data set, it is also necessary to include results that extend over a wide range of conditions, albeit with a higher uncertainty, provided they are consistent with other lower uncertainty data or with theory. In all cases, the uncertainty claimed for the final recommended data must reflect the estimated uncertainty in the primary information.

2   The Correlation

The viscosity η can be expressed [25-36] as the sum of four independent contributions, as

$$\eta(\rho,T)=\eta_0(T)+\eta_1(T)\rho+\Delta\eta(\rho,T)+\Delta\eta_{\mathrm{c}}(\rho,T),\qquad(1)$$

where ρ is the density, T is the absolute temperature, and the first term, $\eta_0(T) = \eta(0,T)$, is the contribution to the viscosity in the dilute-gas limit, where only two-body molecular interactions occur. The linear-in-density term, $\eta_1(T)\rho$, known as the initial-density dependence term, can be separately established with the use of the Rainwater-Friend theory [38-40] for the transport properties of moderately dense gases. The critical enhancement term, $\Delta\eta_{\mathrm{c}}(\rho,T)$, arises from the long-range density fluctuations that occur in a fluid near its critical point, which contribute to divergence of the viscosity at the critical point.

This term for viscosity is significant only in the region very near the critical point, as shown in Vesovic et al. [41] and Hendl et al. [42]. For CO_2, Vesovic et al. [41] showed that the enhancement contributes greater than 1% to the viscosity only in the small region bounded by $0.986 < T_r < 1.019$ and $0.642 < \rho_r < 1.283$. Since data close to the critical point are unavailable, $\Delta\eta_c(\rho,T)$ will be set to zero in Eq. 1 and not discussed further. Finally, the term $\Delta\eta(\rho,T)$, the residual term, represents the contribution of all other effects to the viscosity of the fluid at elevated densities including many-body collisions, molecular-velocity correlations, and collisional transfer.

Identification of these four separate contributions to the viscosity allows one, to some extent, to treat $\eta_0(T)$, and $\eta_1(T)$ theoretically. In addition, it is possible to derive information about both $\eta_0(T)$ and $\eta_1(T)$ from experiment. In contrast, there is little theoretical guidance concerning the residual contribution, $\Delta\eta(\rho,T)$, and its evaluation is usually based entirely on an empirical equation obtained by fitting experimental data.

Table 1 summarizes, to the best of our knowledge, all the available experimental measurements of the viscosity of difluoromethane reported in the literature. Table 1 displays the experimental technique, purity, uncertainty as reported by the original authors, number of measurements, as well as the range of temperatures and pressures investigated. Furthermore, measurements employed in the aforementioned six viscosity correlations are indicated by a corresponding superscript, indicated in the table footnote.

In the primary data set we included the oscillating-disk measurements of Takahashi et al. [9] and the capillary measurement of Dunlop [15], as they were obtained with an uncertainty of 0.3%. Furthermore we included the vibrating-wire measurements of Assael et al. [5] and Oliveira and Wakeham [6], both obtained with an uncertainty of 0.5%. It should however be noted, that from the measurements of Oliveira and Wakeham [6] we excluded the near-saturated-vapor phase data as the conditions of temperature and pressure quoted do not correspond to the vapor phase (even with their recalculation in their 1999 publication [18]). These four sets of measurements have successfully been employed in many of the aforementioned reference correlations; similar data from these authors were used in the latest viscosity reference correlation for 1,1,1,2-tetrafluoroethane [35].

The falling-body measurements of Heide [44] obtained with an uncertainty of 2%, and extending to lower temperatures, were also included in the primary data set, as they were also successfully included in a recent viscosity correlation for R-134a [35]. The falling-cylinder measurements of Grebenkov et al. [43] performed with an uncertainty of 2.8%, were also included in the primary data set, as they extend to 16 MPa.

The measurements of Geller et al. [19], as well as the earlier ones of Bivens et al. [12], both performed in the same capillary instrument with a stated uncertainty of 1.2% were not included in the primary data set, as other data measured with this instrument did not agree well with the other investigators in our previous reference correlation for 1,1,1,2- tetrafluoroethane [35]. However, there are very few vapor-phase measurements at 0.1 MPa, and we did include in the primary data set the 0.1 MPa vapor-phase measurements of Geller et al. [19].

Finally, in the primary data set, we included the recent measurements at high pressures:
-   of Meng et al. [24] obtained in a vibrating-wire instrument, up to a pressure of 30 MPa, with a 2% uncertainty (at the 95% confidence level), and
-   of Bair and Laesecke [23] obtained in a falling-cylinder instrument, up to a pressure of 350 MPa, with a 3 to 6% uncertainty (at the 95% confidence level).

Following our above discussion on the measurements of Geller et al. [19] and Bivens et al. [12], we did not also include in the primary data set
-   the 1999 measurements of Laesecke et al. [13] as they were performed with a sealed gravitational capillary viscometer with straight vertical capillary, for which doubts have been expressed about its proper operation [51], and
-   remaining sets along the liquid saturation line, that quote uncertainty of over 3%, or do not quote the uncertainty.
-   Finally, we became aware of a new dataset [45], from the same group as Ref. [24], after the development of the correlation was completed and have included it in the secondary data for completeness. Figs. 1 and 2 show the ranges of the primary measurements outlined in Table 1, and the phase boundary.

Table 1 Viscosity measurements of difluoromethane

Investigators/reference	Publ. Year	Technique employed[a]	Purity (%)	Uncer-tainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Primary data							
Meng et al. [24]	2018	VW	99.80	2.0*	36	263-350	2.0-30
Bair and Laesecke [23]	2017	FCyl	Lab[b]	3.0-6.0*	18	313-393	10.0-350
Geller et al.[vap] [19][B]	1996	Cap	99.98	1.2	7	253-363	0.1
Grebenkov et al. [43]	1996	FCyl	99.99	2.8	EQ[c]	290-345	0.1-16
Heide [44]	1996	FBod	99.40	2.0	12	223-333	0.1-4.0
Takahashi et al. [9][K,H,W,B]	1995	OD	99.972	0.3	114	298-423	0.1-10
Assael et al. [5][L,W,B,M]	1994	VW	99.98	0.5	26	273-313	1.6-16
Dunlop [15][W,B]	1994	Cap	99.10	0.3	1	298.15	0.1
Oliveira and Wakeham[liq] [6][L,K,H,W,B]	1993	VW	99.90	0.5	19	231-343	0.1-4.9
Secondary data							
Liang et al. [45]	2022	VW	99.96	2.0*	9	273-353	3.0-15.0
Yang et al. [46]	2020	VW	99.99	3.5	13	282-363	4.1-6.3
Cui et al. [47]	2016	SLS (kin)	99.96	2.0-6.0	7	293-348	1.4-5.4
Fröba et al. [20][B,M]	2000	SLS (kin)	99.90	4.0	12	233-343	0.1-4.9
Laesecke et al. [13][H,W,B,M]	1999	Cap	99.93	2.4*	60	290-315	0.3-2.6
Geller et al. [19][B]	1996	Cap	99.98	1.2	44	253-363	0.4-5.5
Sun et al. [16][W,B,M]	1996	Cap	99.95	3.0	21	233-333	0.1-3.9
Burke et al. [48]	1994	Cap	99.10	na[d]	4	235-294	0.1-1.5
Bivens et al. [12][H]	1993	Cap	99.98	1.2	7	253-348	0.1-5.3
Ripple and Matar [7][L,K,B]	1993	Cap	99.98	3.0-5.0	10	250-294	0.3-1.5
Kochubey and Moin [49]	1978	Cap	na[d]	na[d]	1	273	0.1
Phillips and Murphy [50]	1970	SSLCap	99.90	na[d]	10	200-287	0.03-1.3

[M] Measurements employed in the development of the 2015 correlation of He et al. [22].

[B] Measurements employed in the development of the 2014 correlation of Boychuk [17].

[W] Measurements employed in the development of the 2007 correlation of Wang et al. [14].

[H] Measurements employed in the development of the 2003 correlation of Huber et al. [11].

[K] Measurements employed in the development of the 1999 correlation of Kiselev et al. [8].

[L] Measurements employed in the development of the 1997 correlation of Klein et al. [4]

[liq] Only liquid phase measurements – see text.

[vap] Only 0.1 MPa vapor phase measurements – see text.

[a] Cap, Capillary; FBod, Falling Body; FCyl, Falling Cylinder; kin, kinematic viscosity; OD, Oscillating Disc; SLS, Surface Light Scaterring; SSLCap, Sealed Suspended Level Capillary; VW, Vibrating Wire.

[b] Lab, electronic grade further purified in the Laboratory with freezing and pumping cycles but specific purity level not provided.

[c] EQ, Equation form

[d] na, not available

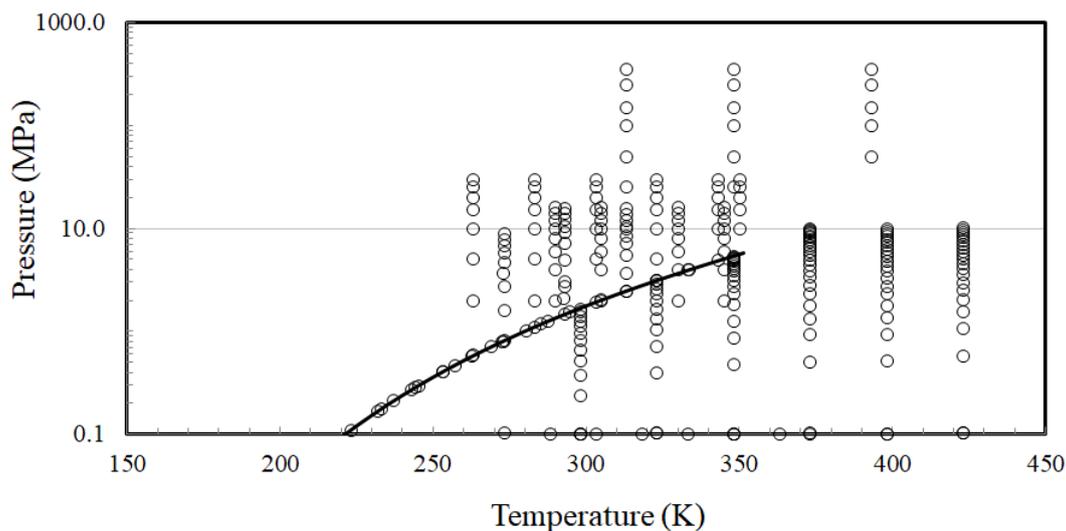* Uncertainty quoted at the 95% confidence level. Others are not specified.

FIG. 1 Temperature-pressure ranges of the primary experimental viscosity data for difluoromethane. (-) saturation curve.
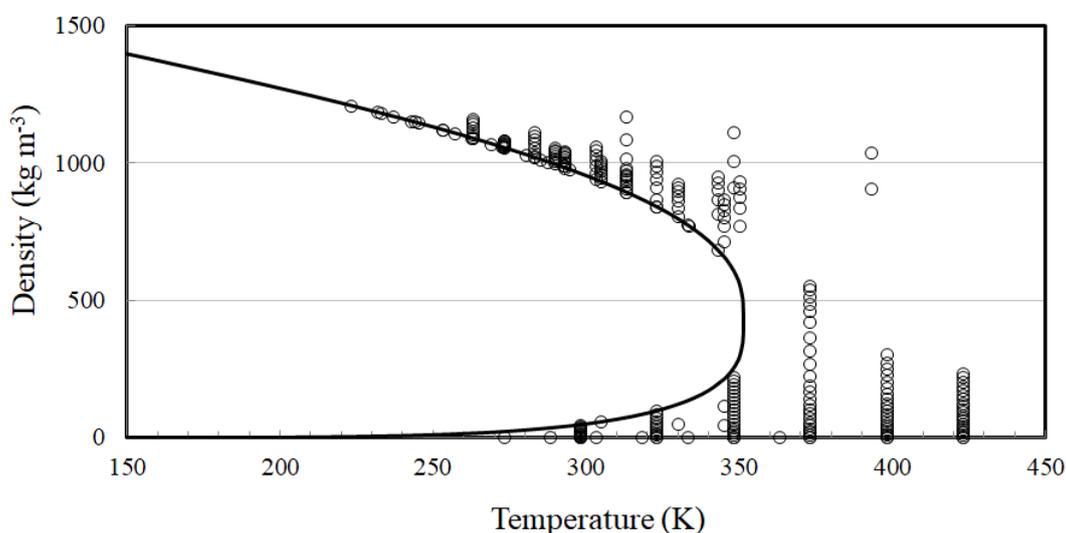

FIG. 2 Temperature-density ranges of the primary experimental viscosity data for difluoromethane. (-) saturation curve.

The development of the correlation requires densities; in 1997, Tillner-Roth and Yokozeki [52] developed an accurate, wide-ranging equation of state that is valid from the triple-point temperature of 136.34 K up to 435 K and pressures of 70 MPa, with an uncertainty in density of 0.05%. It was selected by Annex 18 of the International Energy Agency as an international standard for the thermodynamic properties of R-32 [52]. We also mention that in 2003, Span and Wagner [53] also published an equation of state for difluoromethane, but with a higher uncertainty in density (0.2 to 0.5%). Hence, it was decided to employ in this work the Tillner-Roth and Yokozeki [52] equation of state (also adopted in REFPROP [10]). We also adopt the values for the critical point from their equation of state; the critical temperature, T_c, and the critical density, ρ_c, are 351.255 K and 424.00 kg m^{-3}, respectively, while the triple-point temperature is 136.34 K [52]. Furthermore, we note that the measurements of Bair and Laesecke [23] extend to pressures well above the recommended limit of the EOS. The EOS behaves in a physically reasonable manner up to 350 MPa, the limit of the Bair and Laesecke [23] data, however the uncertainty in density is unknown in the extrapolated region.

2.1 The zero-density limit viscosity term

The zero-density viscosity, $\eta_0(T)$ is a function only of temperature and can be analyzed independently of all other contributions in Eq. 1. Assuming that the Lennard-Jones potential is applicable, one can use Chapman-Enskog theory [54] to express the zero-density viscosity as [55]

$$\eta_0(T) = 0.02669 \frac{\sqrt{MT}}{\sigma^2 \Omega^{(2,2)}} \tag{2}$$

where M (52.024 g mol^{-1}), is the molar mass, and σ is the Lennard-Jones collision diameter in nm, T is the temperature in K, and the resulting viscosity is in µPa s. $\Omega^{(2,2)}$ is the Lennard-Jones collision integral, which can be calculated by the empirical correlation developed by Neufeld et al. [56] as a function of dimensionless temperature $T^* = k_B T / \varepsilon$ (where k_B is Boltzmann's constant and ε is the Lennard-Jones energy parameter), as

$$\begin{aligned} \Omega^{(2,2)}(T^*) = & 1.16145(T^*)^{-0.14874} + 0.52487 e^{-0.7732 T^*} + 2.16178 e^{-2.43787 T^*} \\ & - 6.435 \times 10^{-4} (T^*)^{0.14874} \sin\left[18.0323(T^*)^{-0.7683} - 7.27371\right] . \end{aligned} \tag{3}$$

We used the estimation method in Chung et. al.[55] to provide starting estimates of the Lennard-Jones parameters σ (m) and ε/k_B (K) and adjusted those values to fit the atmospheric pressure data of Geller et al. [19], Takahashi et al. [9], and Dunlop [15] using Eqs. 2 and 3. Although Eqs. 2 and 3 represent the viscosity in the limit of zero density, employing atmospheric-pressure vapor-phase viscosity measurements introduces little error, as the difference in the viscosity values between $p = 0$ and $p = 0.1$ MPa is less than 0.1%. The resulting values obtained from the atmospheric-pressure data are $\sigma = 0.411$ nm and $\varepsilon/k_B = 290$ K. Figure 3 shows the gas-phase atmospheric-pressure data and the calculated results for η_0 using Eqs. 2 and 3. Note that the range of experimental data is limited and having theoretical guidance provides physically reasonable behavior upon extrapolation outside of the range of data.
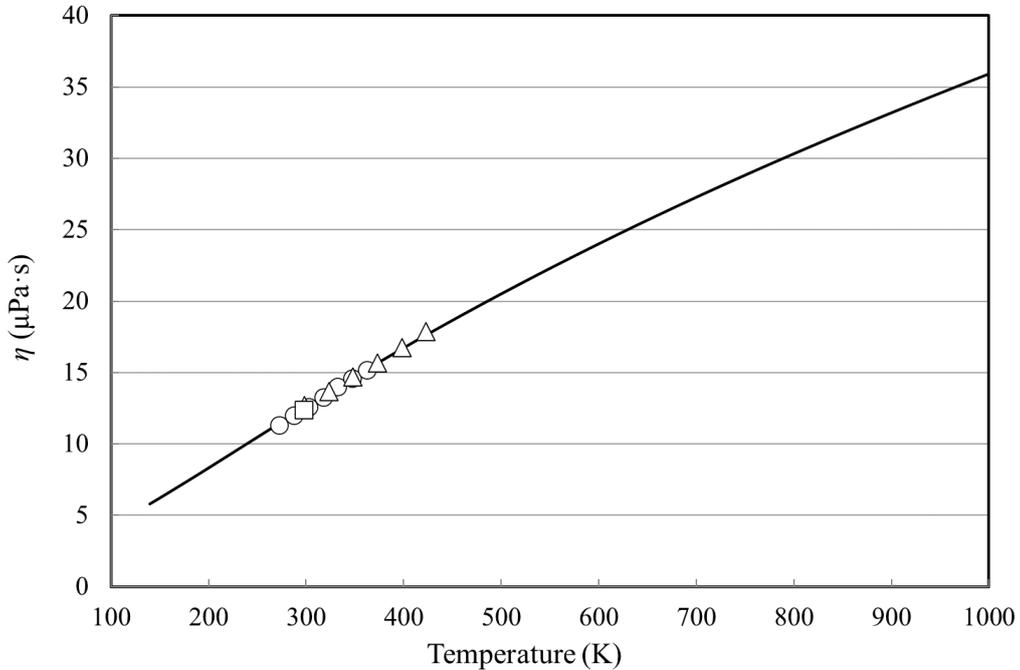

FIG. 3 Dilute-gas viscosity as a function of the temperature. η_0 calculated by Eqs. 2 and 3, Geller et al. [19] (○), Takahashi et al. [9] (△), Dunlop [15] (□)

For ease of use in calculations, η_0 was fitted as:

$$\eta_0(T_r) = \frac{\sum_{i=0}^{4} \alpha_i T_r^i}{\sum_{i=0}^{2} \beta_i T_r^i}, \qquad (4)$$

where the units for η_0 are μPa s, the reduced temperature is $T_r = (T/T_c)$, and the coefficients α_i and β_i are in Table 2. Eq. 4 reproduces the values calculated by Eqs. 2 and 3 to within 0.05 % up to 1000 K, and thus it will be employed hereafter. Figure 4 shows the deviations of the measurements, Eq. 4, and the previous dilute-gas limit viscosity correlations from the values calculated by Chapman-Enskog theory, η_{CE}, as given in Eq. 2 and 3. As shown in Fig. 4, Eq. 4 represents the experimental data to 2%, at a level of $k = 2$. There is an offset of about 0.7 % from the values of REFPROP v10.0, due to the fact that the model implemented in REFPROP v10.0 was based solely on the data of Takahashi et al. [9]. The limited experimental data do not agree to within their mutual estimated uncertainties as given in Table 1, and the availability of better data would allow improved representation of the dilute gas in the future.

Table 2 Coefficients and parameters for Eqs. 4 - 7

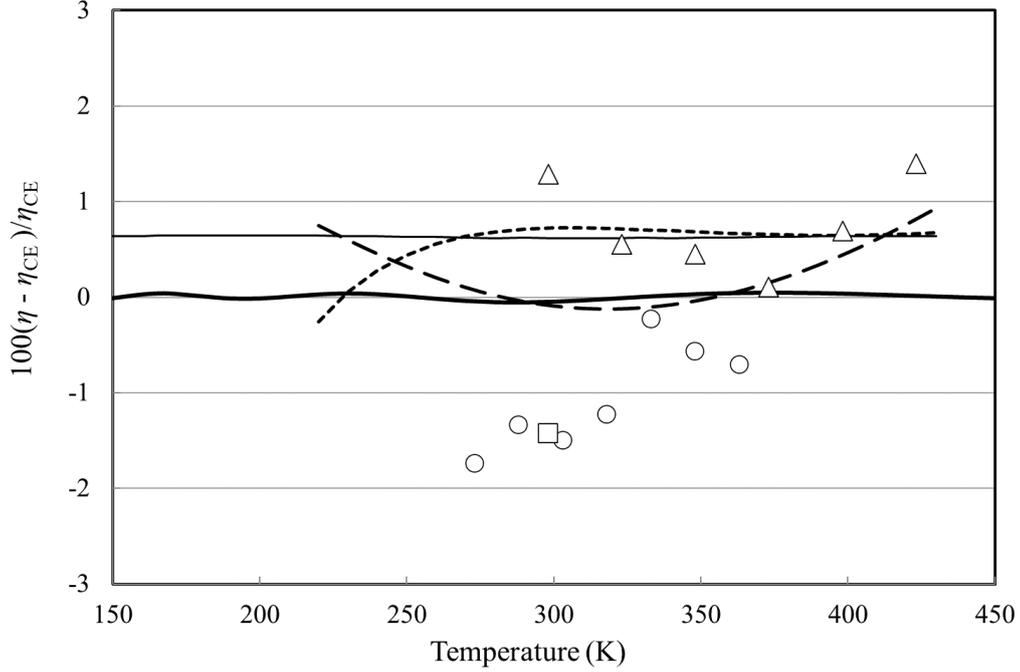|	
ε/k_B (K)	290.0
σ (nm)	0.411
M (g mol^{-1})	52.024
T_c (K)	351.255
ρ_c (kg m^{-3})	424.00
Coefficients α_i (μPa s) for Eq. 4	
0	0.577885
1	1.02498×10^1
2	−4.95882
3	1.41485×10^1
4	−0.816434
Coefficients β_i (-) for Eq. 4	
0	0.896478
1	−0.595706
2	1.0
Coefficients b_i (-) for Eq. 7 [57]	
0	-1.9572881×10^1
1	2.1973999×10^2
2	-1.0153226×10^3
3	2.4710125×10^3
4	-3.3751717×10^3
5	2.4916597×10^3
6	-7.8726086×10^2
7	1.4085455×10^1
8	-3.4664158×10^{-1}

FIG. 4 Dilute-gas viscosity deviations from the values obtained by Chapman-Enskog theory, η_{CE}, as a function of the temperature. Geller et al. [19] ($\circ$), Takahashi et al. [9] ($\triangle$), Dunlop [15] ($\square$), Eq. 4 (━), He et al. [22] and Wang et al. [14] (---), Boychuk [17] (— —), REFPROP 10 [10] (——).

2.2 The initial-density dependence viscosity term

To represent the initial density dependence of viscosity term, $\eta_1\rho$, in Eq. 1, we use the model proposed by Vogel et al. [58] which expresses the second viscosity virial coefficient $B_\eta(T)$ in m^3 kg^{-1}, as

$$B_\eta(T) = \frac{\eta_1(T)}{\eta_0(T)} . \tag{5}$$

In Eq. 5, if the dilute-gas limit viscosity, $\eta_0(T)$, is expressed in µPa s, then the initial-density dependence viscosity term, $\eta_1(T)$, will be expressed in µPa s m^3 kg^{-1}. The second viscosity virial coefficient can be obtained according to the theory of Rainwater and Friend [38, 39] as a function of a reduced second viscosity virial coefficient, $B_\eta^*(T^*)$, as

$$B_\eta^*(T^*) = \frac{B_\eta(T)\, M}{N_A\, \sigma^3} , \tag{6}$$

where [39]

$$B_\eta^*(T^*) = \sum_{i=0}^{6} b_i \left(T^*\right)^{-0.25i} + b_7 \left(T^*\right)^{-2.5} + b_8 \left(T^*\right)^{-5.5} . \tag{7}$$

In Eq. 6, M is the molar mass 52.024 g mol^{-1}, the scaled temperature is $T^* = T/(\varepsilon/k_B)$, and N_A is the Avogadro constant. The coefficients b_i from Ref. [57] are given in Table 2.

2.3 The residual term

The residual viscosity term, $\Delta\eta(\rho,T)$, represents the contribution of all other effects to the viscosity of the fluid at elevated densities including many-body collisions, molecular-velocity correlations, and collisional transfer. An attempt to employ the hard-sphere model proposed by Assael et al. [59] was not very successful, as the present data cover the gas, liquid, and supercritical regions. Hence, it was preferred to evaluate this term almost entirely on experimentally-obtained data, as discussed in the next paragraph.

The procedure adopted during this analysis used an in-house-developed least-squares software optimized using the Fisher score [60] to fit all the primary data to the residual viscosity as a function of the reduced temperature, $T_r = T/T_c$ and reduced density, $\rho_r = \rho/\rho_c$. In addition, we adopted a form suggested by the hard-sphere model employed by Assael *et al.* [59] $\Delta\eta(\rho_r,T_r)=(\rho_r^{2/3}T_r^{1/2})F(\rho_r,T_r)$, where the fitting software was used to determine the functional form for $F(\rho_r,T_r)$. For this task, the dilute-gas limit and the initial density dependence term were calculated for each experimental point, employing Eqs. 4-7, and subtracted from the experimental viscosity to obtain the residual term, $\Delta\eta(\rho_r,T_r)$. The density values were obtained by the Tillner-Roth and Yokozeki [52] equation of state. The final equation was

$$\Delta\eta(\rho,T) = (\rho_r^{2/3}T_r^{1/2})\left\{c_0 + c_1\rho_r + c_2\frac{\rho_r^4}{T_r} + c_3\frac{\rho_r^{14}}{T_r} + c_4\frac{\rho_r^2}{T_r^2}\right\} \quad (8)$$

Coefficients c_i are given in Table 3, and $\Delta\eta$ is in μPa·s.

Table 3 Coefficients c_i for Eq. 8.

i	c_i
0	1.24655×10^0
1	8.85264×10^0
2	5.87282×10^{-1}
3	2.81507×10^{-6}
4	4.41060×10^0

2.4 Comparison with data

Table 4 summarizes comparisons of the primary data with the correlation. We define the percent deviation as PCTDEV = $100(\eta_{exp}-\eta_{fit})/\eta_{fit}$, where η_{exp} is the experimental value of the viscosity and η_{fit} is the value calculated from the correlation. The average absolute percent deviation (AAD) is found with the expression AAD = $(\sum|\text{PCTDEV}|)/n$, where the summation is over all n points, the bias percent is found with the expression BIAS = $(\sum\text{PCTDEV})/n$. The average absolute percentage deviation of the fit for all primary data is 1.32 %, with a bias of -0.33 %. The estimated uncertainty of the correlation in the temperature range 220 to 425 K and up to 350 MPa is 3.4 % (at the 95% confidence level). Although the recommended upper pressure limit for the EOS of Tillner-Roth and Yokozeki [52] is 70 MPa, the viscosity correlation and the EOS behave in a physically realistic manner at pressures up to 350 MPa and we included all of the Laesecke and Bair data [61] in the primary data and analysis.

Table 4 Evaluation of the difluoromethane viscosity correlation for the primary data.

1st Author	Year Publ.	AAD (%)	BIAS (%)
Meng et al. [24]	2018	2.79	-2.79
Bair and Laesecke [23]	2017	1.07	0.20
Geller et al.[vap] [19]	1996	1.18	-1.18
Grebenkov et al. [43]	1996	1.72	-0.76
Heide [44]	1996	2.00	-1.49
Takahashi et al. [9]	1995	1.06	-0.35
Assael et al. [5]	1994	1.23	1.23
Dunlop [15]	1994	1.53	-1.53
Oliveira and Wakeham[liq] [6]	1993	1.57	0.06
Entire data set		**1.32**	**-0.33**

Fig. 5 shows the relative deviations of all primary viscosity data of difluoromethane from the values calculated by Eqs. 1,4 - 8, as a function of temperature, while Figs. 6 and 7 show the same deviations but as a function of the pressure and the density. As shown in Fig. 7, the viscosity correlation well represents the data at pressures up to 350 MPa, even though the pressures are above the recommended limit of the EOS. The correlation also behaves in a physically realistic manner and may be safely used down to the triple-point temperature of 136.34 K [52]. Near the triple-point temperature, we expect the uncertainty to be larger than 3.4%, possibly much larger, although the uncertainty is difficult to assess due to lack of data, as there are no primary data below 223 K. Additional experimental data for the liquid at temperatures below 220 K are necessary to further validate the correlation and make improved correlations possible in the future, especially if there is interest in very-low-temperature applications. As mentioned earlier, additional vapor-phase measurements at pressures up to atmospheric pressure also would enable a more accurate correlation in the future.
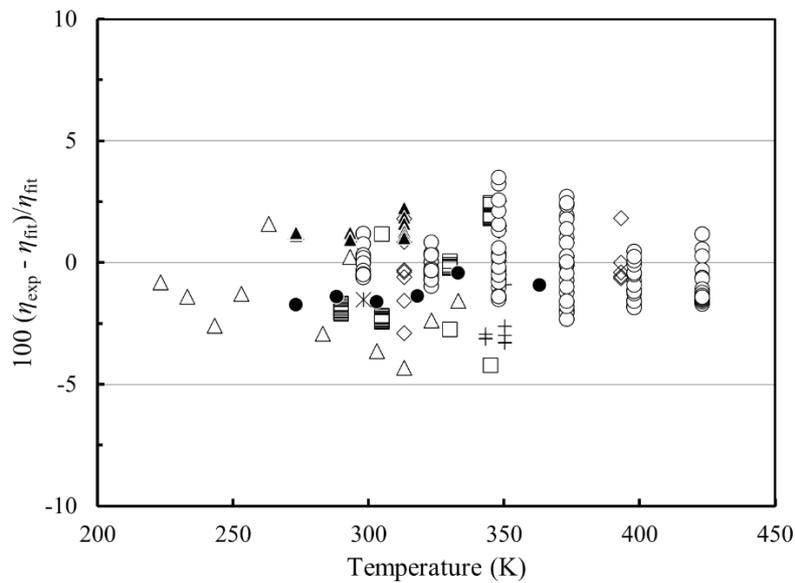

FIG. 5 Relative deviations of primary experimental data of difluoromethane from the values calculated by the present model, Eqs. 1, 4-8, as a function of temperature. Meng et al. [24] (+), Bair and Laesecke [23] (◇), Grebenkov et al. [43] (□), Heide [44] (△), Geller et al.[vap] [19] (●), Takahashi et al. [9] (O), Assael et al. [5] (▲), Dunlop [15] (✱), and Oliveira and Wakeham[liq] [6] (×).

FIG. 6 Relative deviations of primary experimental data of difluoromethane from the values calculated by the present model, Eqs. 1, 4-8, as a function of pressure. Meng et al. [24] (+), Bair and Laesecke [23] (◇), Grebenkov et al. [43] (□), Heide [44] (△), Geller et al.[vap] [19] (●), Takahashi et al. [9] (O), Assael et al. [5] (▲), Dunlop [15] (✱), and Oliveira and Wakeham[liq] [6] (×).
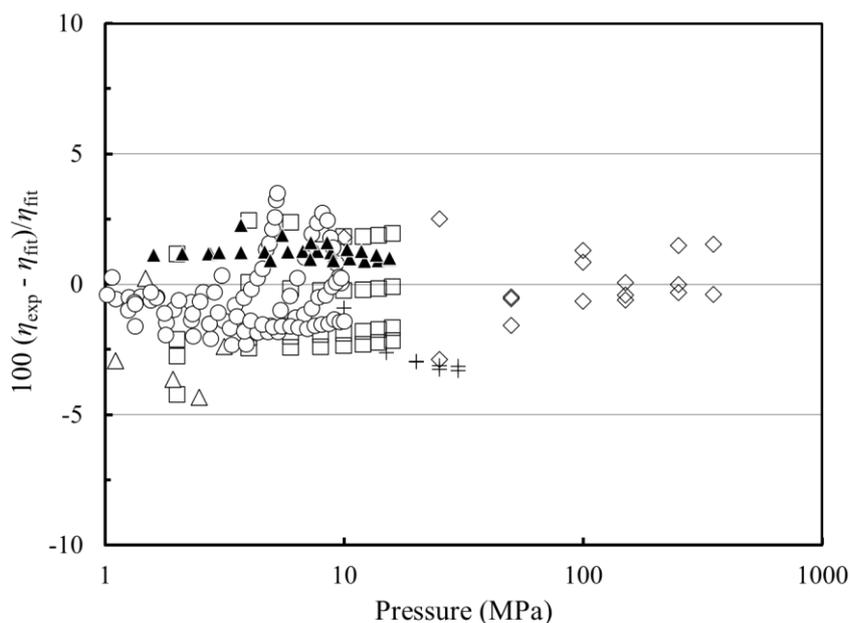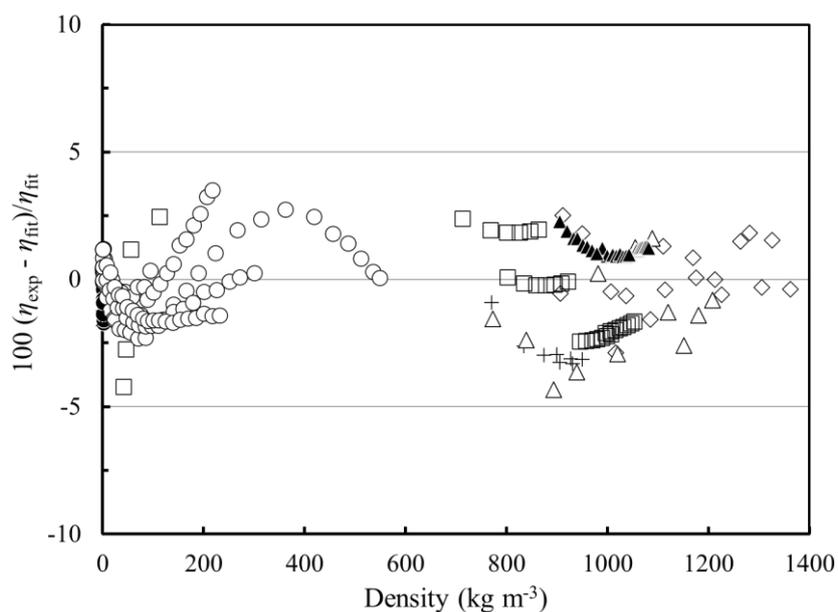

FIG. 7 Relative deviations of primary experimental data of difluoromethane from the values calculated by the present model, Eqs. 1, 4-8, as a function of density. Meng et al. [24] (+), Bair and Laesecke [23] (◇), Grebenkov et al. [43] (□), Heide [44] (△), Geller et al.[vap] [19] (●), Takahashi et al. [9] (O), Assael et al. [5] (▲), Dunlop [15] (✱), and Oliveira and Wakeham[liq] [6] (×).

Table 5 shows the average absolute percent deviation (AAD) and the bias for the secondary data. The measurements of Liang et al. [45] show deviations consistent with the earlier measurements of that same group (Meng et al. [24]) that were included in the primary data set. Finally, Fig. 8 shows a plot of the viscosity of difluoromethane as a function of the temperature for different pressures. The plot demonstrates the extrapolation behavior at pressures higher than 70 MPa (limit imposed by the equation of state), and at temperatures that exceed the 423 K limit of the current measurements.

Table 5 Evaluation of the difluoromethane viscosity correlation for the secondary data.

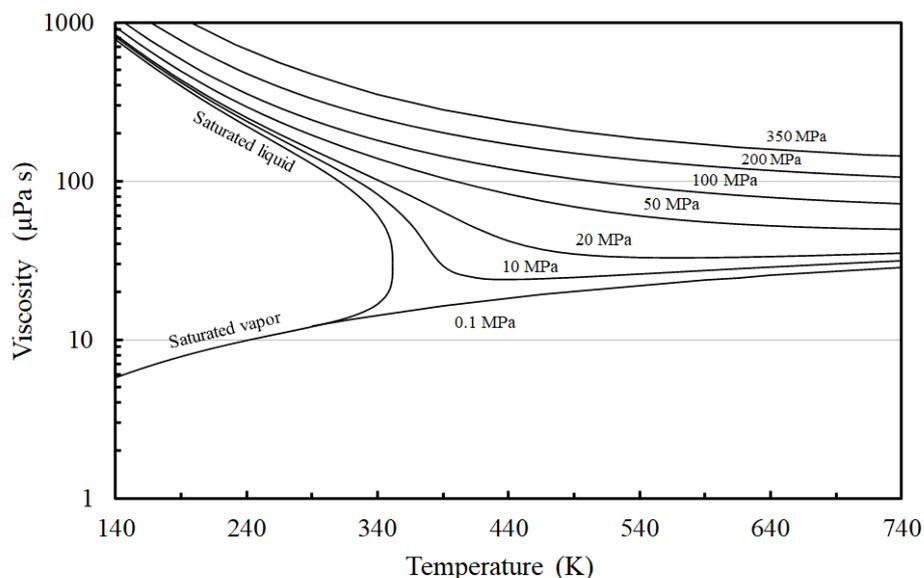| 1st author	Year Publ.	AAD (%)	BIAS (%)
Liang et al. [45]	2022	2.00	-2.00
Yang et al. [46]	2020	2.71	-2.07
Cui et al. [47]	2016	1.37	0.46
Fröba et al. [20]	2000	4.04	0.46
Laesecke et al. [13]	1999	2.88	-2.88
Geller et al. [19]	1996	5.43	-3.53
Sun et al. [16]	1996	1.84	-1.23
Burke et al. [48]	1994	4.41	-4.41
Bivens et al. [12]	1993	5.83	3.06
Ripple and Matar [7]	1993	1.27	1.27
Kochubey and Moin [49]	1978	1.03	1.03
Phillips and Murphy [50]	1970	34.3	34.3

FIG. 8 Viscosity of difluoromethane as a function of the temperature for different pressures.

3   Recommended Values

In Table 6, viscosity values are given along the saturated liquid and vapor curves, calculated from the present proposed correlations between 220 and 350 K, while in Table 7 viscosity values are given for temperatures between 220 and 420 K at selected pressures. Saturation pressure and saturation density values for selected temperatures, as well as the density values for the selected temperature and pressure, are obtained from the equation of state of Tillner-Roth and Yokozeki [52]. The calculations are performed at the given temperatures and densities. For computer verification of values, the following points may be used for the given T, ρ conditions: $T = 300$ K, $\rho = 0$ kg m^{-3}, $\eta = 12.6170$ μPa s; $T = 300$ K, $\rho = 10.0$ kg m^{-3}, $\eta = 12.6333$ μPa s; $T = 300$ K, $\rho = 1100.0$ kg m^{-3}, $\eta = 173.431$ μPa s.

Table 6   Viscosity values of difluoromethane along the saturation curve, calculated by the present scheme.

T (K)	p (MPa)	ρ_{liq} (kg m^{-3})	ρ_{vap} (kg m^{-3})	η_{liq} (μPa s)	η_{vap} (μPa s)
220	0.093818	1217.1	2.7796	278.2	9.117
240	0.23965	1160.3	6.7367	222.4	9.938
260	0.52157	1098.9	14.239	178.8	10.77
280	1.0069	1031.1	27.430	143.6	11.65
300	1.7749	953.2	49.97	113.9	12.67
320	2.9194	857.2	89.65	87.37	14.08
340	4.5614	714.8	172.8	60.35	16.88
350	5.6311	558.3	297.4	40.77	22.01

4   Conclusions

A new wide-ranging correlation for the viscosity of difluoromethane was developed based on critically evaluated experimental data and theoretical results. The average absolute percentage deviation of the fit for all primary data is 1.32 %, with a bias of -0.33 %. The estimated uncertainty of the correlation in the temperature range 220 to 425 K and up to 350 MPa is 3.4 % (at the 95% confidence level), however the recommended upper limit of the EOS is 70 MPa, and densities used in the development and analysis of the viscosity correlation above 70 MPa are extrapolated values. The correlation behaves in a physically reasonable manner at temperatures between the triple-point temperature, 136.34 K [52], and 220 K, however there are no reliable experimental data in this region and uncertainty assessment is difficult. Additional experimental measurements in the liquid phase at temperatures below 220 K would allow validation of the correlation in this region. The uncertainty in the atmospheric-pressure gas phase is 2%; the lack of high-quality experimental data in this region also hinders the development of a more accurate correlation.

Supporting Information Available

A text file containing the parameters for the calculation of the thermophysical properties of R-32 including the viscosity correlation in this work is available for use with the REFPROP computer program. It must be named R32.fld to be viewed properly by the REFPROP program.

Table 7 Viscosity values of difluoromethane at selected temperatures and pressures, calculated by the present scheme.

p (MPa)	T (K)	ρ (kg m^{-3})	η (μPa s)	p (MPa)	T (K)	ρ (kg m^{-3})	η (μPa s)
0.1	220	1217.1	278.2	50	220	1285.4	355.2
	240	2.685	10.01		240	1243.0	293.1
	260	2.457	10.82		260	1200.3	245.9
	280	2.270	11.77		280	1157.1	209.3
	300	2.111	12.63		300	1113.5	180.4
	320	1.974	13.49		320	1069.5	157.2
	340	1.854	14.33		340	1025.1	138.3
	360	1.749	15.15		360	980.43	122.8
	380	1.655	15.97		380	935.62	109.9
	400	1.571	16.77		400	890.93	99.1
	420	1.495	17.55		420	846.62	90.1
10	220	1233.4	294.4	70	220	1306.1	384.3
	240	1180.8	237.8		240	1266.7	318.6
	260	1125.0	193.8		260	1227.5	269.0
	280	1064.9	158.7		280	1188.3	230.6
	300	998.6	129.7		300	1149.2	200.4
	320	922.7	104.8		320	1110.2	176.3
	340	829.8	82.11		340	1071.5	156.8
	360	699.6	59.60		360	1033.1	140.8
	380	478.5	36.01		380	995.1	127.6
	400	305.8	26.15		400	957.7	116.6
	420	239.0	24.30		420	921.0	107.3
30	220	1261.7	325.6				
	240	1215.1	266.4				
	260	1167.4	221.3				
	280	1118.3	186.0				
	300	1067.4	157.9				
	320	1014.5	134.9				
	340	959.4	116.0				
	360	901.8	100.2				
	380	841.7	86.9				
	400	779.6	75.8				
	420	716.4	66.6				

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