

Reference Correlation for the Viscosity of Ammonia from the Triple Point to 725 K and up to 50 MPa^{a)}

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This paper presents a new wide-ranging correlation for the viscosity of ammonia based on critically evaluated experimental data. The correlation is designed to be used with a recently developed equation of state, and it is valid from the triple point to 725 K, at pressures up to 50 MPa. The estimated uncertainty varies depending on the temperature and pressure, from 0.6 % to 5%. The correlation behaves in a physically reasonable manner when extrapolated to 100 MPa, however care should be taken when using the correlations outside of the validated range.

Key words: ammonia; transport properties; viscosity.

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1. Introduction

In a series of recent papers, reference correlations for the viscosity of refrigerants^{1,2} and also selected common hydrocarbon fluids³⁻⁷ have been 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 new reference correlations for the viscosity of ammonia, a fluid very important for agricultural fertilizer production and also as a refrigerant.

There have been several assessments⁸⁻¹¹ of the viscosity of ammonia in the past, but only Fenghour *et al.*,¹² in 1995, proposed a correlation of the viscosity data for both gaseous and liquid ammonia based on a critical assessment of all published experimental measurements at that time. The correlation of Fenghour *et al.*¹² employed the 1993 Tillner-Roth *et al.*¹³ equation of state. Very recently, Gao *et al.*¹⁴ developed a new equation of state for ammonia (including a slightly different critical point than the values used by Tillner-Roth *et al.*¹³) The present work employs this new equation of state, and a) a new set of measurements of viscosity published by Laesecke *et al.*¹⁵ in 1999, b) a new set of kinematic viscosity data published by Estrada-Alexanders and Hurlly in 2008,¹⁶ and c) a low-temperature set of measurements by Wong and Tobias,¹⁷ that were not considered in the previous correlation.

The analysis that will be described follows the procedure also adopted by Fenghour *et al.*,¹² applied to the best available experimental data for the viscosity. Thus, a prerequisite to the analysis is a critical assessment of the experimental data. For this purpose, two categories of experimental data are defined: 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.¹⁸ 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^{1, 4-7} as the sum of four independent contributions, as

$$\eta(\rho, T) = \eta_0(T) + \eta_1(T)\rho + \Delta\eta(\rho, T) + \Delta\eta_c(\rho, T), \quad (1)$$

where ρ is the molar 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 development of the Rainwater-Friend theory¹⁹⁻²¹ for the transport properties of moderately dense gases. The critical enhancement term, $\Delta\eta_c(\rho, T)$, 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.*²² and Hendl *et al.*²³ Since we do not have data very close to the critical point, $\Delta\eta_c(\rho, T)$ will be set to zero in Eq. (1) and not discussed further in this work. 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.

The identification of these four separate contributions to the viscosity and to transport properties in general is useful because it is possible, to some extent, to treat $\eta_0(T)$, $\eta_1(T)$, and $\Delta\eta_c(\rho, T)$ 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 therefore its evaluation is based entirely on an empirical equation obtained by fitting experimental data.

Table 1 summarizes, to the best of our knowledge, the experimental measurements^{15-17,24-60} of the viscosity of ammonia reported in the literature. The majority of these measurements were employed by Fenghour *et al.*¹² in their 1996 reference correlation for ammonia, with 11 of them denoted as primary data. We adopted these same data sets as primary data in this work, and also included

TABLE 1. Viscosity measurements of ammonia.

1 st author	Year Publ.	Technique employed ^a	Purity ^b (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Primary Data							
Estrada-Alexanders ¹⁶	2008	GRAC	na	0.6	71	300–375	0.22–3.4
Laesecke ¹⁵	1999	CAP	99.90	3.3	51	285–335	0.66–2.74
Sun ²⁴	1979	CAP	na	2.0	38	448–598	0.65–12.16
Hongo ²⁵	1977	OD	purified	0.5	62	298–373	0.117–6
Golubev ²⁶	1974	CAP	99.9	2.0–3.0	132	273–444	0.1–14.81
Bhattacharyya ²⁷	1970	OD	na	0.6	5	217–308	0.0066
Makhija ²⁸	1970	CAP	na	1.0	20	208–298	0.101–0.102
Iwasaki ²⁹	1968	OD	purified	0.5	106	298–408	0.101–9.3
Burch ³⁰	1967	CAP	99.99	1.5	11	273–673	0.101
Wong ¹⁷	1966	UCAP	na	0.5	8	203–238	0.09–0.10
Iwasaki ³¹	1964	OD	purified	0.5	20	294–303	0.106–0.62
Carmichael ³²	1963	RC	99.999	2.0–4.0	177	294–444	0.101–39.6
Trautz ^{33,c}	1931	CAP	na	2.0–10.0	28	291–990	0.101
Braune ³⁴	1930	OD	na	1.5	9	293–700	0.101–0.101
Vogel ³⁵	1914	OD	na	2.0–3.0	2	196–273	0.000078–0.0069
Secondary Data							
Rakshit ³⁶	1974	OD	99.5	1.0	5	237–307	0.08–1.34
Rakshit ³⁷	1973	OD	99.5	1.0	4	238–308	0.09–1.34
Golubev ^{38,d}	1970	CAP	na	na	28	303–406	1.16–11.36
Golubev ^{38,e}	1970	CAP	na	na	121	303–523	0.1–81
Pal ³⁹	1969	OD	99.5	1.0	5	297–473	0.013–0.013
Pal ⁴⁰	1967	OD	99.5	1.0	5	306–479	0.013–0.013
Pal ⁴¹	1967	OD	na	1.0	5	306–469	0.013–0.013
Pal ⁴²	1967	OD	na	1.0	4	298–372	0.013–0.013
Chakraborti ⁴³	1965	CAP	na	1.0	3	299–353	0.101–0.101
Krynicky ⁴⁴	1963	CAP	na	4.0	8	273–343	0.432–3.31
Shimotake ⁴⁵	1963	CAP	99.99	0.5	27	373–473	1.7–35
Carmichael ⁴⁶	1952	RB	na	0.7	30	277–378	0.94–42
Kiyama ⁴⁷	1952	CAP	99.9	na	47	323–573	0.101–9.5
Shatenshtein ⁴⁸	1949	CAP	na	3.0	3	288–298	0.7–1.0
Pinevich ⁴⁹	1948	CAP	na	na	6	247–323	0.144–2
Wobser ⁵⁰	1941	RB	na	na	5	293–363	0.101
Planck ⁵¹	1939	CAP	na	na	3	278–298	0.5–1.0
Van Cleave ⁵²	1935	OD	na	1.0	9	201–297	0.01–0.98
Stakelbeck ⁵³	1933	FC	na	na	101	253–353	0.1–2.6
Monoszon ⁵⁴	1932	CAP	na	na	3	223–239	0.0408–0.100
Fredenhagen ⁵⁵	1930	CAP	na	na	2	208–238	0.101
Jung ⁵⁶	1930	CAP	na	2.0	2	287–289	0.71–0.74
Edwards ⁵⁷	1925	CAP	na	1.0	3	288–456	0.101
Rankine ⁵⁸	1921	CAP	na	0.3	1	373	0.101
Elsy ⁵⁹	1920	CAP	na	na	1	239	0.1
Fitzgerald ⁶⁰	1912	CAP	na	na	1	239	0.1

^a CAP, Capillary; FC, Falling Cylinder; GRAC, Greenspan Acoustic; OD, Oscillating Disc; RB, Rolling Ball; RC, Rotating Cylinder; UCAP, Ubbelohde Capillary.

^b na. not available.

^c Measurements above 700 K were not considered because decomposition of ammonia into hydrogen and nitrogen was observed

^d Attributed to Golubev, Table 46

^e Attributed to Golubev and Petrov, Table 21

several additional sets. The measurements of Bhattacharyya *et al.*²⁷ were performed in an oscillating-disk viscometer with an uncertainty of 0.6%, and were here considered as primary data to extend the temperature range down to 217 K. Furthermore, in addition to these measurements, three more investigators are considered as primary data. In 2008 Estrada-Alexanders and Hurly¹⁶ performed very accurate measurements of the kinematic viscosity of ammonia in a Greenspan acoustic viscometer, calibrated with a series of vapors, with an uncertainty of 0.6%. These measurements were included in the primary data set. The saturated liquid measurements of Laesecke *et al.*¹⁵ were performed in a sealed gravitational viscometer with a straight vertical capillary and an uncertainty of 3.3%. These were included in the primary data set. Finally, also included in the primary data set were the low-temperature measurements of Wong and Tobias¹⁷ performed in a modified Ubbelohde capillary with a 0.5% uncertainty. This set, although dated 1966, was not included in the Fenghour *et al.*¹² paper.

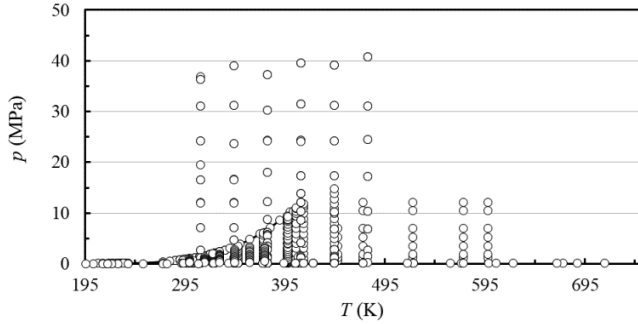


FIG. 1. Temperature-pressure ranges of the primary experimental viscosity data for ammonia. (—) saturation curve.

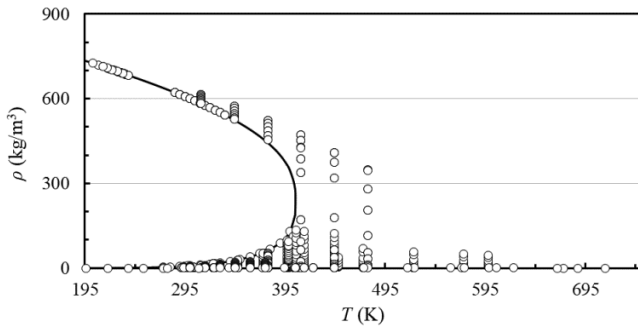


FIG. 2. Temperature-density ranges of the primary experimental viscosity data for ammonia. (—) saturation curve.

Figures 1 and 2 show the ranges of the primary measurements outlined in Table 1, and the phase boundary may be seen as well. The development of the correlation requires densities; Gao *et al.*¹⁴ very recently developed an accurate, wide-ranging equation of state that is valid from the triple point up to 725 K and 1000 MPa. In the vapor phase, the uncertainties in density are 0.1 % at temperatures between 410 K and 580 K with pressures

below 100 MPa, and 0.05 % at temperatures between 220 K and 400 K with pressures below 10 MPa. In the liquid phase, the uncertainty in density is 0.05 % at temperatures between 190 K and 400 K with pressures below 200 MPa. The uncertainty in density is 1.5 % at pressures between 200 MPa and 1000 MPa. In the critical region, the uncertainty in density is estimated to be 1 %. The uncertainty in saturated liquid density is 0.1 % at temperatures between 195 K and 400 K. The uncertainty in saturated vapor density is 2 % at temperatures between 220 K and 395 K. 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 405.56 K and 233.250 kg m⁻³, respectively.¹⁴ The triple-point temperature employed is 195.49 K.¹⁴ Note that the value of the critical density of 233.25 kg m⁻³ proposed by the new correlation of Gao *et al.*,¹⁴ is different from the value of 225.00 kg m⁻³ employed by the previous Tillner-Roth *et al.*¹³ equation of state.

2.1. The dilute-gas limit and the initial-density dependence terms

The dilute-gas limit viscosity, $\eta_0(T)$ in $\mu\text{Pa s}$, can be analyzed independently of all other contributions in Eq. (1). According to the kinetic theory, the viscosity of a pure polyatomic gas may be related to an effective collision cross section, which contains all the dynamic and statistical information about the binary collision. For practical purposes, this relation is formally identical to that of monatomic gases and can be written as:⁶¹

$$\eta_0(T) = 0.021357 \frac{\sqrt{MT}}{\sigma^2 S_\eta^*(T^*)}, \quad (2)$$

where $S_\eta^* = S(2000)/(\pi\sigma^2 f_\eta)$ is a reduced effective cross section, M is the molar mass in g mol⁻¹, σ is the length scaling parameter in nm, f_η is a dimensionless higher-order correction factor according to Chapman and Cowling,^{62, 63} and $S(2000)$ is a generalized cross section that includes all of the information about the dynamics of the binary collisions that govern transport properties, and in turn are governed by the intermolecular potential energy surface.⁶¹ The reduced effective cross section is usually expressed in the functional form

$$\ln S_\eta^*(T^*) = \sum_{i=0}^4 \alpha_i (\ln T^*)^i, \quad (3)$$

$$T^* = k_B T / \varepsilon, \quad (4)$$

where T^* is the reduced temperature, ε/k_B is an energy scaling parameter in K, and k_B is Boltzmann's constant.

The temperature dependence of the linear-in-density coefficient of the viscosity $\eta_1(T)$ in Eq. (1) is very large at

subcritical temperatures and must be taken into account to obtain an accurate representation of the behavior of the viscosity in the vapor phase. It changes sign from positive to negative, as the temperature decreases. Therefore, the viscosity along an isotherm should first decrease in the vapor phase and subsequently increase with increasing density.⁶¹ Vogel *et al.*⁶⁴ have shown that fluids exhibit the same general behavior of the initial density dependence of viscosity, which can also be expressed by means of the second viscosity virial coefficient $B_\eta(T)$ in $\text{m}^3 \text{kg}^{-1}$, as

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

Note that in the above equation, if the dilute-gas limit viscosity, $\eta_0(T)$, is expressed in $\mu\text{Pa s}$, then the initial-density viscosity, $\eta_1(T)$, will be expressed in $\mu\text{Pa s m}^3 \text{kg}^{-1}$. The second viscosity virial coefficient can be obtained according to the theory of Rainwater and Friend^{19, 20} as a function of a reduced second viscosity virial coefficient, $B_\eta^*(T^*)$, as

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

where²⁰

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

In Eq. (6), M is the molar mass in g mol^{-1} given in Table 2 and N_A is the Avogadro number. The coefficients b_i from Ref.⁶¹ are given in Table 2.

TABLE 2. Coefficients and parameters for Eqs. (2), (3), and (7).

Molar mass		
17.03052 g mol^{-1}		
Scaling parameters		
$\varepsilon/k_B = 386.0 \text{ K}$	$\sigma = 0.2957 \text{ nm}$	
Coefficients a_i for Eq. (3)		
$a_0 = 0.39175$	$a_1 = -0.59918$	$a_4 = -0.06942$
$a_2 = -0.00022$	$a_3 = 0.19871$	
Coefficients b_i for Eq. (7) ⁶¹		
$b_0 = -19.572881$	$b_1 = 219.73999$	$b_2 = -1015.3226$
$b_3 = 2471.0125$	$b_4 = -3375.1717$	$b_5 = 2491.6597$
$b_6 = -787.26086$	$b_7 = 14.085455$	$b_8 = -0.34664158$

Eqs. (2)–(7) present a consistent scheme for the correlation of the dilute-gas limit viscosity, $\eta_0(T)$ and the initial-density dependence term, $\eta_1(T)$. In order to calculate the dilute-gas limit viscosity we employed all

measurements considered by Fenghour *et al.*¹² and followed their adopted procedure. As already mentioned, we in addition included the measurements of Estrada-Alexanders and Hurly¹⁶ and Bhattacharyya *et al.*²⁷ Furthermore, for the scaling parameters σ and ε/k_B we employed the values proposed by Fenghour *et al.*,¹² shown in Table 2. Hence, Eqs. (2) – (7) were optimized to obtain the coefficients a_i of Eq. (3). These are also shown in Table 2.

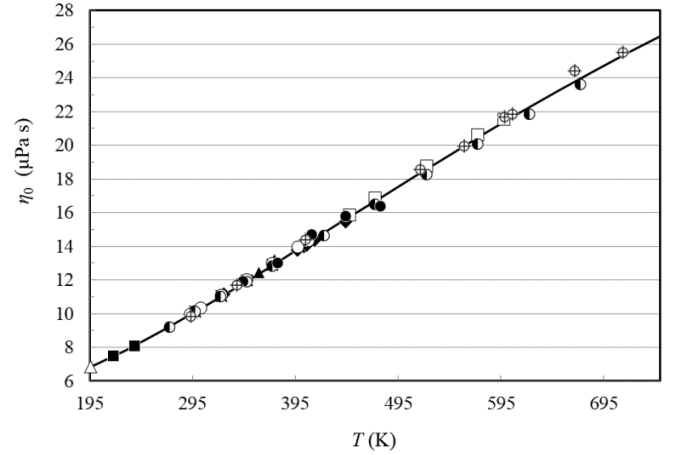


FIG. 3. The dilute-gas viscosity of ammonia as a function of the temperature. Estrada-Alexanders and Hurly¹⁶ (▲), Sun and Storvick²⁴ (□), Hongo and Iwasaki²⁵ (×), Golubev and Likhachev²⁶ (◆), Bhattacharyya *et al.*²⁷ (●), Iwasaki and Takahashi²⁹ (○), Burch and Raw³⁰ (⊕), Carmichael *et al.*³² (●), Braune and Linke³⁴ (⊕), Vogel³⁵ (△), values calculated from scheme of Eqs. (2) – (4) and parameters from Table 2 (—)

Figure 3 shows the dilute-gas viscosity as a function of the temperature, while Fig. 4, shows the percentage deviations between the dilute-gas viscosity, η_0 , (calculated with Eqs. (2) – (4) and the parameters in Table 2) and the experimental viscosity values, as a function of temperature. Of the data sets used in the regression, the highest quality sets are those of Iwasaki and coworkers,^{25, 29, 31} with estimated uncertainties ranging from 0.5-1.5%. Based on comparisons with these data, the estimated expanded relative uncertainty of the dilute gas at temperatures from 293 K to 408 K at a 95% confidence level is 0.6%. All uncertainties stated concerning the correlations presented in this work are expanded uncertainties at the 95% confidence level. Below 293 K, the underlying data have larger uncertainties, and we estimate the expanded uncertainty of the correlation is on the order of 2%. For temperatures above 408 K, again the underlying data have larger uncertainties and we estimate the expanded uncertainty of the correlation is also 2%. In Fig. 4 the correlation proposed by Fenghour *et al.*¹² is also shown. As expected, since they are primarily based on the same measurements, their agreement is within 1% up to 600 K.

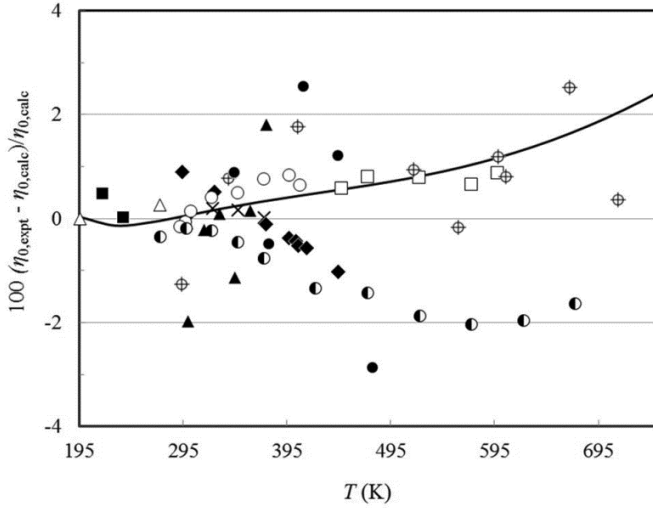


FIG. 4. Percentage deviations between the dilute-gas viscosity, η_0 , (calculated with Eqs. (2) – (4) and the parameters in Table 2) and the experimental viscosity values, as a function of temperature. Estrada-Alexanders and Hurly¹⁶ (\blacktriangle), Sun and Storvick²⁴ (\square), Hongo and Iwasaki²⁵ (\times), Golubev and Likhachev²⁶ (\blacklozenge), Bhattacharyya *et al.*²⁷ (\blacksquare), Iwasaki and Takahashi²⁹ (\circ), Burch and Raw³⁰ (\bullet), Carmichael *et al.*³² (\bullet), Braune and Linke³⁴ (\oplus), Vogel³⁵ (\triangle), Fenghour *et al.*¹² 1995 correlation (—)

Figure 5 shows the experimental and calculated values of the initial-density viscosity. Having obtained the dilute-gas viscosity, the initial-density viscosity is calculated from Eqs. (5) – (7), and the low-density primary measurements of Table 1, extrapolated to zero density. Only those measurements that could be extrapolated to zero density were employed.

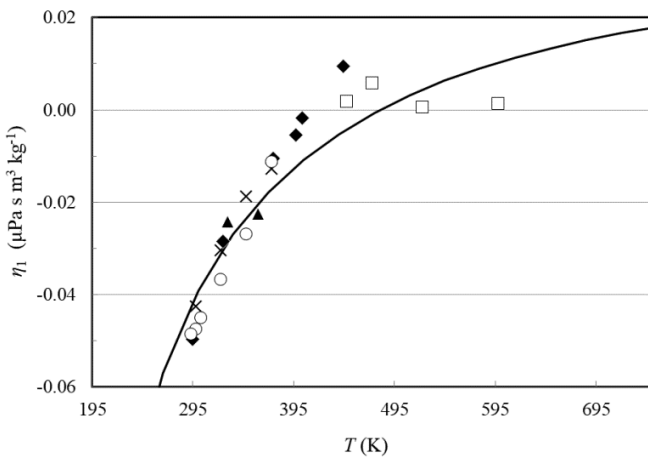


FIG. 5. Experimental and calculated values of the initial-density viscosity. Estrada-Alexanders and Hurly¹⁶ (\blacktriangle), Sun and Storvick²⁴ (\square), Hongo and Iwasaki²⁵ (\times), Golubev and Likhachev²⁶ (\blacklozenge), Iwasaki and Takahashi²⁹ (\circ), values calculated from scheme of Eqs. (2) – (7) and parameters from Table 2 (—).

2.2. The residual term

As stated in Section 2, 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. Because there is little theoretical guidance concerning this term, its evaluation here is based entirely on experimentally obtained data.

The procedure adopted during this analysis used symbolic regression software⁶⁵ to fit all the primary data to the residual viscosity. Symbolic regression is a type of genetic programming that allows the exploration of arbitrary functional forms to regress data. The functional form is obtained by use of a set of operators, parameters, and variables as building blocks. Most recently this method has been used to obtain correlations for the viscosity of R161,¹ n-undecane,³ and R1234yf and R1234ze(E).² In the present work, we restricted the operators to the set (+, -, *, /) and the operands (constant, T_r , ρ_r), with $T_r = T/T_c$ and $\rho_r = \rho/\rho_c$. In addition, we adopted a form suggested from the hard-sphere model employed by Assael *et al.*,⁶⁶ $\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3} T_r^{1/2}) F(\rho_r, T_r)$, where the symbolic regression method 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. (2)–(7), and subtracted from the experimental viscosity to obtain the residual term, $\Delta\eta(\rho_r, T_r)$. The density values employed were obtained by the equation of state of Gao *et al.*¹⁴ The final equation obtained was

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

Coefficients c_i are given in Table 3, and $\Delta\eta$ is in $\mu\text{Pa s}$.

TABLE 3. Coefficients c_i for Eq. (8).

i	c_i
0	0.039 330 8
1	16.724 735 0
2	1.197 593 4
3	0.001 699 5
4	-4.239 979 4

Table 4 summarizes comparisons of the primary data with the correlation. We have defined the percent deviation as $\text{PCTDEV} = 100 * (\eta_{\text{exp}} - \eta_{\text{fit}}) / \eta_{\text{fit}}$, where η_{exp} is the experimental value of the viscosity and η_{fit} is the value calculated from the correlation. Thus, the average absolute percent deviation (AAD) is found with the

expression $AAD = (\sum |PCTDEV|)/n$, where the summation is over all n points, the bias percent is found with the expression $BIAS = (\sum PCTDEV)/n$. The average absolute percentage deviation of the fit for all primary data is 0.98%, with a bias of 0.07%. The uncertainty varies depending on the region of the phase diagram the data are located in. As mentioned earlier, the dilute-gas region has an estimated expanded uncertainty ranging from 0.6% to 2% depending on the temperature. The estimated expanded uncertainty for the liquid at pressures near atmospheric or saturation pressure from the triple point to 285 K is 2%. Along the saturated liquid boundary over the temperature range from 285 K to 335 K, the estimated expanded uncertainty is the same as the data of Laesecke *et al.*,¹⁵ namely 3.3%. At higher pressures, the primary data are rather limited, especially above 10 MPa, where only the data of Carmichael *et al.*³² exist. We estimate the expanded uncertainty at a 95% confidence level for pressures up to 40 MPa over the temperature range from 310 K to 480 K is about 4%, and also is 4% for the supercritical fluid up to 600 K and 12 MPa. Outside of this range, we estimate the expanded uncertainty of the liquid viscosity is 5% at pressures to 50 MPa. The correlation behaves in a physically realistic manner at pressures up to 100 MPa and we feel the correlation may be extrapolated to this limit, although the uncertainty will be larger, especially at lower temperatures, and may be on the order of 25% near the triple-point temperature at 100 MPa. Additional experimental data at high pressures are necessary to validate the correlation or make improved correlations possible in the future. The equation of state is valid up to extremely high pressures, 1000 MPa, but we do not recommend the use of the correlation at these conditions.

TABLE 4. Evaluation of the ammonia viscosity correlation for the primary data.

I st Author	Year Publ.	AAD (%)	BIAS (%)
Estrada-Alexanders ¹⁶	2008	0.78	0.40
Laesecke ¹⁵	1999	0.70	-0.18
Sun ²⁴	1979	1.60	-1.27
Hongo ²⁵	1977	0.36	-0.31
Golubev ²⁶	1974	0.85	-0.36
Bhattacharyya ²⁷	1970	0.65	-0.40
Makhija ²⁸	1970	0.86	0.53
Iwasaki ²⁹	1968	0.42	0.15
Burch ³⁰	1967	1.06	-1.03
Wong ¹⁷	1966	1.19	-1.01
Iwasaki ³¹	1964	0.24	-0.17
Carmichael ³²	1963	1.35	0.71
Trautz ^{33,a}	1931	1.41	-1.41
Braune ³⁴	1930	1.07	0.79
Vogel ³⁵	1914	0.15	0.15
Entire data set		0.98	0.07

^a Measurements above 700 K were not considered because decomposition of ammonia into hydrogen and nitrogen was observed

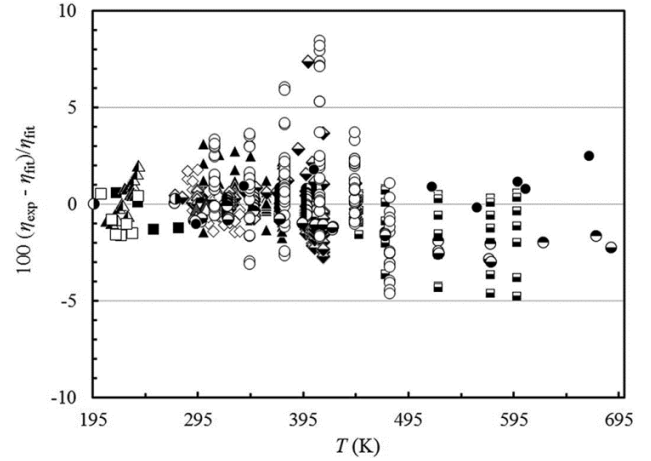


FIG. 6. Percentage deviations of primary experimental data of ammonia from the values calculated by the present model as a function of temperature. Estrada-Alexanders and Hurly¹⁶ (\blacktriangle), Laesecke *et al.*¹⁵ (\diamond), Sun and Storvick²⁴ (\blacksquare), Hongo and Iwasaki²⁵ (\blacklozenge), Golubev and Likhachev²⁶ (\blacklozenge), Bhattacharyya *et al.*²⁷ (\blacksquare), Makhija and Stairs²⁸ (\blacktriangle), Iwasaki and Takahashi²⁹ (\bullet), Burch and Raw³⁰ (\circ), Wong and Tobias¹⁷ (\square), Iwasaki *et al.*³¹ (\triangle), Carmichael *et al.*³² (\circ), Trautz and Heberling³³ (\bullet), Braune and Linke³⁴ (\bullet), and Vogel³⁵ (\bullet).

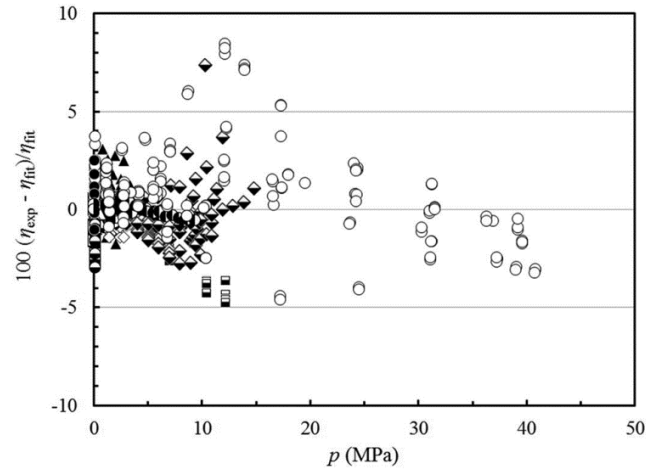


FIG. 7. Percentage deviations of primary experimental data of ammonia from the values calculated by the present model as a function of pressure. Estrada-Alexanders and Hurly¹⁶ (\blacktriangle), Laesecke *et al.*¹⁵ (\diamond), Sun and Storvick²⁴ (\blacksquare), Hongo and Iwasaki²⁵ (\blacklozenge), Golubev and Likhachev²⁶ (\blacklozenge), Bhattacharyya *et al.*²⁷ (\blacksquare), Makhija and Stairs²⁸ (\blacktriangle), Iwasaki and Takahashi²⁹ (\bullet), Burch and Raw³⁰ (\circ), Wong and Tobias¹⁷ (\square), Iwasaki *et al.*³¹ (\triangle), Carmichael *et al.*³² (\circ), Trautz and Heberling³³ (\bullet), Braune and Linke³⁴ (\bullet), and Vogel³⁵ (\bullet).

Figure 6 shows the percentage deviations of all primary viscosity data from the values calculated by Eqs. (1) - (8), as a function of temperature, while Figs. 7 and 8 show the same deviations but as a function of the pressure and the density. Table 5 shows the average absolute percent deviation (AAD) and the bias for the secondary data. Finally, Fig. 9 shows a plot of the viscosity of ammonia as a function of the temperature for different pressures. The plot demonstrates the extrapolation behavior at pressures higher than 50 MPa, and at temperatures that exceed the 725 K limit of the equation of state.

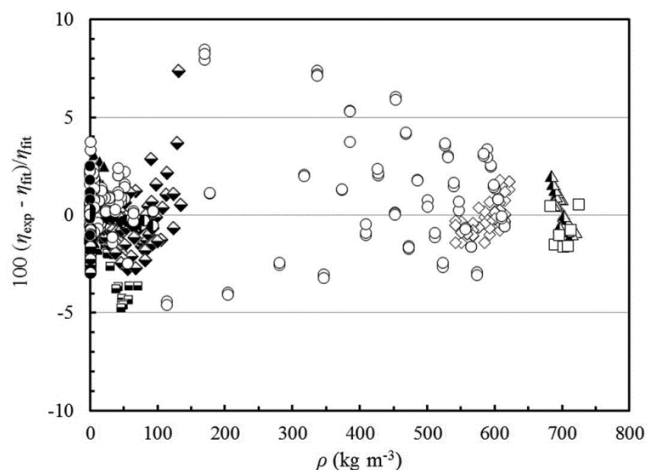


FIG. 8. Percentage deviations of primary experimental data of ammonia from the values calculated by the present model as a function of density. Estrada-Alexanders and Hurly¹⁶ (\blacktriangle), Laesecke *et al.*¹⁵ (\diamond), Sun and Storvik²⁴ (\blacksquare), Hongo and Iwasaki²⁵ (\blacklozenge), Golubev and Likhachev²⁶ (\blacklozenge), Bhattacharyya *et al.*²⁷ (\blacksquare), Makhija and Stairs²⁸ (\blacktriangle), Iwasaki and Takahashi²⁹ (\bullet), Burch and Raw³⁰ (\circ), Wong and Tobias¹⁷ (\square), Iwasaki *et al.*³¹ (\triangle), Carmichael *et al.*³² (\circ), Trautz and Heberling³³ (\bullet), Braune and Linke³⁴ (\bullet), and Vogel³⁵ (\bullet).

TABLE 5. Evaluation of the ammonia viscosity correlation for the secondary data.

1 st author	Year Publ.	AAD (%)	BIAS (%)
Rakshit ³⁶	1974	1.04	1.00
Rakshit ³⁷	1973	0.98	0.98
Golubev ^{38,a}	1970	7.99	7.74
Golubev ^{38,b}	1970	4.14	1.52
Pal ³⁹	1969	1.92	1.92
Pal ⁴⁰	1967	1.26	1.26
Pal ⁴¹	1967	1.33	1.33
Pal ⁴²	1967	4.17	4.17
Chakraborti ⁴³	1965	0.77	-0.04
Krynicky ⁴⁴	1963	3.49	3.49
Shimotake ⁴⁵	1963	11.89	-2.43
Carmichael ⁴⁶	1952	17.30	0.73
Kiyama ⁴⁷	1952	10.38	9.13
Shatenshtein ⁴⁸	1949	6.19	6.19
Pinevich ⁴⁹	1948	7.51	6.71
Wobser ⁵⁰	1941	0.71	0.55
Planck ⁵¹	1939	3.49	3.49
Van Cleave ⁵²	1935	2.25	0.57
Stakelbeck ⁵³	1933	35.58	35.58
Monoszon ⁵⁴	1932	1.80	-1.07
Fredenhagen ⁵⁵	1930	4.91	4.91
Jung ⁵⁶	1930	3.14	3.14
Edwards ⁵⁷	1925	1.30	1.30
Rankine ⁵⁸	1921	1.10	1.10
Elsy ⁵⁹	1920	0.78	0.78
Fitzgerald ⁶⁰	1912	5.42	5.42

^a Attributed to Golubev, Table 46

^b Attributed to Golubev and Petrov, Table 21

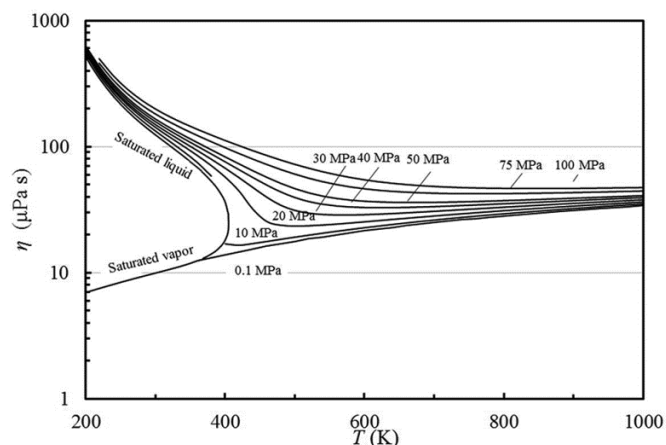


FIG. 9. Viscosity of ammonia as a function of the temperature for different pressures.

3. Recommended Values

In Table 6, viscosity values are given along the saturated liquid and vapor lines, calculated from the present proposed correlations between 200 and 400 K, while in Table 7 viscosity values are given for temperatures between 200 and 700 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 Gao *et al.*¹⁴ 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, $\eta=10.1812$ μ Pa s; $T=300$ K, $\rho=8.0$ kg m⁻³, $\eta=9.9219$ μ Pa s, $T=300$ K, $\rho=609.0$ kg m⁻³, $\eta=133.3937$ μ Pa s.

TABLE 6. Viscosity values of ammonia along the saturation line, calculated by the present scheme.

T (K)	p (MPa)	ρ_{liq} (kg m ⁻³)	ρ_{vap} (kg m ⁻³)	η_{liq} (μ Pa s)	η_{vap} (μ Pa s)
200	0.0086098	728.67	0.088673	516.02	6.95
220	0.033731	705.46	0.31840	347.27	7.49
240	0.10217	681.43	0.89692	251.04	8.07
260	0.25525	656.14	2.1156	192.06	8.67
280	0.55070	629.20	4.3800	153.03	9.28
300	1.0611	600.17	8.2443	125.00	9.92
320	1.8718	568.33	14.500	103.20	10.60
340	3.0792	532.48	24.390	85.03	11.38
360	4.7925	490.34	40.196	68.99	12.41
380	7.1397	436.25	67.327	53.95	14.03
400	10.297	344.01	130.89	37.32	17.94

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

p (MPa)	T (K)	ρ (kg m ⁻³)	η (μ Pa s)
0.1	200	728.7031	516.21
	250	0.8381	8.41
	300	0.6898	10.16
	350	0.5885	12.01
	400	0.5138	13.91
	450	0.4562	15.83
	500	0.4103	17.74
	550	0.3728	19.61
	600	0.3416	21.43
	650	0.3153	23.20
	700	0.2927	24.92
10	200	732.30	536.98
	250	674.27	229.05
	300	608.47	132.88
	350	525.57	83.21
	400	411.47	16.95
	450	61.22	17.37
	500	48.76	19.13
	550	41.67	20.93
	600	36.82	22.70
	650	33.18	24.42
	700	30.31	26.08
30	200	739.19	578.87
	250	684.14	250.50
	300	624.25	149.30
	350	555.69	100.29
	400	470.58	67.23
	450	350.12	43.01
	500	219.00	31.04
	550	156.80	28.63
	600	126.57	28.69
	650	108.37	29.47
	700	95.89	30.52
50	200	745.67	620.80
	250	693.03	271.48
	300	637.43	164.63
	350	576.90	114.68
	400	509.08	82.69
	450	431.63	60.35
	500	347.35	46.35
	550	273.15	39.53
	600	221.15	36.87
	650	186.52	36.06
	700	162.59	36.10

It should be noted that values produced by the proposed correlation for the viscosity of ammonia, are within the mutual uncertainties of the values calculated by the Fenghour *et al.*¹² correlation. However, we do believe that the proposed correlation should be preferred as it describes better the low-pressure liquid range (new measurements of Estrada-Alexanders and Hurly,¹⁶ and Laesecke *et al.*¹⁵), and more specifically the low-temperature region (measurements of Wong and Tobias¹⁷), while it also incorporates the new EoS of Gao *et al.*¹⁴

4. Conclusions

A new wide-ranging correlation for the viscosity of ammonia was developed based on critically evaluated experimental data, including a data set along the saturated liquid boundary¹⁵ not available when the viscosity surface of Fenghour *et al.*¹² was developed. The correlation is expressed in terms of temperature and density, is designed to be used with the very recent equation of state of Gao *et al.*¹⁴ It is valid from the triple point to 725 K, at pressures up to 50 MPa. The correlation behaves in a physically realistic manner at pressures up to 100 MPa and we feel the correlation may be extrapolated to this limit, although the uncertainty will be larger and caution is advised when extrapolating to 100 MPa. The uncertainty in the critical region also will be larger since the critical enhancement term has been omitted. Additional liquid-phase experimental data at pressures, especially from 10- 100 MPa, and also measurements very close to the critical point, are necessary to validate the correlation or make improved correlations possible in the future.

5. References

1. C. M. Tsolakidou, M. J. Assael, M. L. Huber, and R. A. Perkins, *J. Phys. Chem. Ref. Data* **46**, 023103 (2017).
2. M. L. Huber and M. J. Assael, *Int. J. Refrig.* **71**, 39 (2016).
3. M. J. Assael, T. B. Papalas, and M. L. Huber, *J. Phys. Chem. Ref. Data* **46**, 033103 (2017).
4. S. Avgeri, M. J. Assael, M. L. Huber, and R. A. Perkins, *J. Phys. Chem. Ref. Data* **44**, 033101 (2015).
5. S. Avgeri, M. J. Assael, M. L. Huber, and R. A. Perkins, *J. Phys. Chem. Ref. Data* **43**, 033103 (2014).
6. E. K. Michailidou, M. J. Assael, M. L. Huber, I. M. Abdulagatov, and R. A. Perkins, *J. Phys. Chem. Ref. Data* **43**, 023103 (2014).
7. E. K. Michailidou, M. J. Assael, M. L. Huber, and R. A. Perkins, *J. Phys. Chem. Ref. Data* **42**, 033104 (2013).
8. W. G. Grocnier and G. Thodos, *J. Chem. Eng. Data* **6**, 240 (1961).
9. *ASHRAE, Thermophysical Properties of Refrigerants* (American Society of Heating, Refrigeration and Air-Conditioning Engineers, Inc., New York, 1976).
10. F. Golubev, V. P. Kiyashova, I. I. Perel'shtein, and E. B. Parushin, *The thermophysical properties of ammonia (in Russian), Izdatel'stvo standartov, Moscow, English translation of Chapters 4 and 5: Technical Translation No. NEL-TT-2866* (National Engineering Laboratory, East Kilbride, Glasgow, U.K., 1978).

11. K. Stephan and K. Lucas, *Viscosity of Dense Fluids* (Plenum, New York 1979).
12. A. Fenghour, W. A. Wakeham, V. Vesovic, J. T. R. Watson, J. Millat, and E. Vogel, *J. Phys. Chem. Ref. Data* **24**, 1649 (1995).
13. R. Tillner-Roth and F. Harms-Watzenberg, *DKV Tagungsbericht* **20**, 167 (1993).
14. K. Gao, J. Wu, I. H. Bell, and E. W. Lemmon, (to be submitted, 2018).
15. A. Laesecke, T. O. D. Luddecke, R. F. Hafer, and D. J. Morris, *Int. J. Thermophys.* **20**, 401 (1999).
16. A. F. Estrada-Alexanders and J. J. Hurly, *J. Chem. Thermodyn.* **40**, 193 (2008).
17. W. S. Wong and C. W. Tobias, *Viscosity Of Liquid Ammonia in the Temperature Interval of -30°C to -70°C* (Report Number: UCRL-16909, Berkeley California, USA, 1966).
18. M. J. Assael, M. L. V. Ramires, C. A. Nieto de Castro, and W. A. Wakeham, *J. Phys. Chem. Ref. Data* **19**, 113 (1990).
19. D. G. Friend and J. C. Rainwater, *Chem. Phys. Lett.* **107**, 590 (1984).
20. J. C. Rainwater and D. G. Friend, *Phys. Rev. A* **36**, 4062 (1987).
21. E. Bich and E. Vogel, *Chap. 5.2, in Transport Properties of Fluids. Their Correlation, Prediction and Estimation* (Cambridge University Press, Cambridge, 1996).
22. V. Vesovic, W. A. Wakeham, G. A. Olchoway, J. V. Sengers, J. T. R. Watson, and J. Millat, *J. Phys. Chem. Ref. Data* **19**, 763 (1990).
23. S. Hendl, J. Millat, E. Vogel, V. Vesovic, W. A. Wakeham, J. Luettmer-Strathmann, J. V. Sengers, and M. J. Assael, *Int. J. Thermophys.* **15**, 1 (1994).
24. S. B. K. Sun and T. S. Storvick, *J. Chem. Eng. Data* **24**, 88 (1979).
25. M. Hongo and H. Iwasaki, *Rev. Phys. Chem. Jpn.* **47**, 90 (1977).
26. I. F. Golubev and V. F. Likhachev, *Trudy GIAP (Gosud. Inst. Azotn. Prom.)* **6**, 15 (1974).
27. P. K. Bhattacharyya, A. K. Ghosh, and A. K. Barna, *J. Phys. B* **3**, 526 (1970).
28. R. C. Makhija and R. A. Stairs, *Can. J. Chem.* **48**, 1214 (1970).
29. H. Iwasaki and M. Takahashi, *Rev. Phys. Chem. Jpn.* **38**, 18 (1968).
30. J. G. Burch and G. I. G. Raw, *J. Chem. Phys.* **47**, 2798 (1967).
31. H. Iwasaki, J. Kestin, and A. Nagashima, *J. Chem. Phys.* **40**, 2988 (1964).
32. L. T. Carmichael, H. H. Reamer, and B. H. Sage, *J. Chem. Eng. Data* **8**, 400 (1963).
33. M. Trautz and R. Heberling, *Ann. Phys.* **10**, 155 (1931).
34. H. Braune and R. Linke, *Z. Phys. Chem. Leipzig* **A148**, 195 (1930).
35. H. Vogel, *Ann. Phys.* **43**, 1235 (1914).
36. A. B. Rakshit and C. S. Roy, *Physica* **78**, 153 (1974).
37. A. B. Rakshit, C. S. Roy, and A. K. Barna, *J. Chem. Phys.* **59**, 3633 (1973).
38. I. F. Golubev and V. A. Petrov, *in Viscosity of Gases and Gas Mixtures, A Handbook*, edited by I. F. Golubev (English translation published by Israel Program for Scientific Translations, Jerusalem, 1970).
39. A. K. Pal and P. K. Bhattacharyya, *J. Chem. Phys.* **51**, 828 (1969).
40. A. K. Pal and A. K. Barua, *J. Chem. Phys.* **47**, 216 (1967).
41. A. K. Pal and A. K. Barua, *Trans. Faraday Soc.* **63**, 341 (1967).
42. A. K. Pal and A. K. Barua, *Ind. J. Phys.* **41**, 713 (1967).
43. P. K. Chakraborti and P. Gray, *Trans. Faraday Soc.* **61**, 2422 (1965).
44. K. Krynicki and J. W. Hennel, *Acta Phys. Polon.* **24**, 269 (1963).
45. H. Shimotake and G. Thodos, *AIChE J.* **9**, 68 (1963).
46. L. T. Carmichael and B. H. Sage, *Ind. Eng. Chem.* **44**, 2728 (1952).
47. R. Kiyama and T. Makita, *Rev. Phys. Chem. Jpn.* **22**, 49 (1952).
48. A. I. Shatenshtein, E. A. Izrailevich, and N. I. Ladyshnikova, *Zh. Fiz. Khim.* **23**, 497 (1949).
49. G. Pinevich, *Kholod. Tekh.* **20**, 30 (1948).
50. R. Wobser and F. Muller, *Kolloidbeihfte* **52**, 165 (1941).
51. C. J. Planck and H. J. Hunt, *J. Am. Chem. Soc.* **61**, 3590 (1939).
52. A. B. Van Cleave and O. Maass, *Can. J. Res.* **B13**, 140 (1935).
53. H. Stakelbeck, *Z. Gesamte Kälte-Ind.* **40**, 33 (1933).
54. A. M. Monoszon and V. A. Pleskov, *Zh. Fiz. Khim.* **3**, 221 (1932).
55. K. Fredenhagen, *Z. Anorg. Chem.* **186**, 1 (1930).
56. G. Jung and H. Schmick, *Z. Phys. Chem. Leipzig* **7**, 130 (1930).
57. R. S. Edwards and B. Worswick, *Proc. Phys. Soc. London* **38**, 16 (1925).
58. A. O. Rankine and C. J. Smith, *Philos. Mag.* **42**, 601 (1921).
59. H. M. Elsey, *J. Am. Chem. Soc.* **42**, 2454 (1920).
60. F. F. Fitzgerald, *J. Phys. Chem.* **16**, 621 (1912).
61. E. Vogel, C. Küchenmeister, E. Bich, and A. Laesecke, *J. Phys. Chem. Ref. Data* **27**, 947 (1998).
62. S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, London, 1970).
63. G. C. Maitland, M. Rigby, E. B. Smith, and W. A. Wakeham, *Intermolecular Forces: Their Origin and Determination* (Clarendon, Oxford, 1981).

64. E. Vogel, E. Bich, and R. Nimz, *Physica A* **139**, 188 (1986).
65. EUREQA Formulize v.098.1 (Nutionian Inc., Cambridge MA, USA, 2012). Commercial equipment, instruments, or materials are identified only in order to adequately specify certain procedures. In no case does such identification imply

recommendation or endorsement by the National Institute of Standards and technology, no does it imply that the products identified are necessarily the best available for the purpose.

66. M. J. Assael, J. H. Dymond, M. Papadaki, and P. M. Patterson, *Int. J. Thermophys.* **13**, 269 (1992).