The Viscosity and Thermal Conductivity of Ethane in the Limit of Zero Density

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New representations of the viscosity and thermal conductivity of ethane in the limit of zero density are provided. The correlation for the viscosity extends over the temperature range 200 to 1000 K, whereas that for thermal conductivity extends from 225 to 725 K. The behavior of each property is represented by an independent correlation of the appropriate effective collision cross section as a function of temperature. The final results are compared with experimental data as well as with earlier correlations. The accuracy of the viscosity correlation is estimated to be $\pm 0.5\%$ in the temperature range $300\,\mathrm{K} \leqslant T \leqslant 600\,\mathrm{K}$, increasing to ± 1.5 and $\pm 2.5\%$ at 200 and 1000 K, respectively. The uncertainty associated with the thermal conductivity correlation is $\pm 2\%$ in the temperature range $300\,\mathrm{K} \leqslant T \leqslant 500\,\mathrm{K}$, increasing to $\pm 3\%$ at either end. The results of this study indicate that there is an urgent need for additional high-precision measurements of thermal conductivity especially for temperatures above 400 K.

1. INTRODUCTION

The increasing importance of accurate values of the thermophysical properties in general and for the transport properties in particular has been demonstrated in a number of studies [1–5]. In this context the viscosity and thermal conductivity in the limit of zero density provide the essential bases for the correlation and for the prediction of the properties over a wider range of thermodynamic states.

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In the zero density limit the viscosity is almost independent of the existence of internal degrees of freedom and, therefore, is nearly unaffected by inelastic collisions, whereas the thermal conductivity is greatly influenced by both [6]. This is of importance because the ethane molecule is characterized by a significant number of excited vibrational degrees of freedom including one for hindered rotation. This complicates the theoretically based analysis of thermal conductivity which was outlined in earlier papers [1–5].

The aim of this paper is to extend these earlier studies on the viscosity and thermal conductivity of gases to the near-linear but highly vibrationally excited ethane molecule. The methodology of our correlation for the viscosity is a version of that outlined in Ref. 7. For thermal conductivity the available information—including that for auxiliary data—is not sufficient to carry out the complete theoretical analysis as given in Refs. 1–5. Therefore, we provide a provisional temperature function for this property based on the available experimental data and seek to demonstrate that the general properties of this function show no obvious contradictions with the basic theory.

2. DATA SELECTION

As in the proceeding papers, the first step in our analysis is the division of all the data into categories of primary and secondary data [1, 7–9]. The entire set of published viscosity and thermal conductivity data of ethane [10–44] has been reviewed. Tables I and II list those data selected for inclusion in the primary data sets for viscosity [10–17] and thermal conductivity [30–35], respectively. These data form the basis for our

Table I.   Primary Experimental Data for the Viscosity of Ethane

Reference	Technique[a]	Temperature range (K)	Ascribed uncertainty (%)
Hunter and Smith [10]	C	212–393	1.25–0.75
Hendl and Vogel [11]	OD	293–633	0.2–0.3
Kestin et al. [12]	OD	296–304	0.2
Kestin et al. [13]	OD	301–476	0.2–0.45
Abe et al. [14]	OD	298–468	0.2–0.45
Iwasaki and Takahashi [15]	OD	298–348	0.3
Golubev and Mesheryakov [16]	C	258–523	1.5
Carmichael and Sage [17]	RC	305	1.0

[a] OD, oscillating disk; C, capillary; RC, rotating cylinder.

Table II. Primary Experimental Data for the Thermal Conductivity of Ethane

Reference	Technique[a]	Temperature range (K)	Ascribed uncertainty (%)
Millat et al. [30]	THW	308–426	0.5
Fleeter et al. [31]	THW	301	0.5
Clifford et al. [32]	THW	303	1.0
Roder and Castro [33]	THW	225–305	1.6
Prasad and Venart [34]	THW	295–600	2.0
Le Neindre et al. [35]	CC	308–724	1.5

[a] THW, transient hot wire; CC, concentric cylinders.

correlations. The tables also include information about the experimental techniques used, as well as our estimate of the uncertainty assigned to the data.

2.1. Viscosity

The selected primary data for this property originated in six laboratories including the very recent measurements by Hunter and Smith [10] and by Hendl and Vogel [11]. Whereas the former used a capillary viscometer, the latter employed an oscillating-disk viscometer. A similar technique has been used by Kestin and his collaborators [12–14] and by Iwasaki and Takahashi [15]. For all the instruments, there exist complete working equations and the measurements, especially of monatomic systems, have proved mutually consistent with independent results derived from intermolecular potentials [6, 45] within certain limits. Since a number of other data sources will be of importance for the subsequent analysis of the density dependence of viscosity, we also included results by Golubev and Mesheryakov [16] and by Carmichael and Sage [17] in the primary data set.

The remaining data sets [18–29] have been classified as secondary data, because these measurements have been carried out in instruments for which either the complete working equation was unavailable or not all corrections have been applied.

2.2. Thermal Conductivity

We have carried out a survey of the available data and assigned them to one of the above-mentioned categories. From a comparison of the entire

set of published data it is clear that there are substantial discrepancies amounting in some cases to 15%.

The most accurate of the thermal conductivity measurements have been carried out with transient hot-wire instruments at Imperial College [30] and Brown University [31]. The uncertainty we ascribe to the zero-density values described in these papers[3] are given in Table II. Other measurements made with the transient hot-wire technique are those of Clifford et al. [32], Roder and Castro [33], and Prasad and Venart [34]. These data are estimated to have the slightly larger uncertainty as stated in Table II. The remaining primary data are those of Le Neindre et al. [35] measured with a steady-state coaxial cylinder method. Although these results have a greater uncertainty than those measured using transient hot-wire techniques with a complete working equation, the temperature range covered is substantially expanded by their inclusion.

By definition, all other experimental data are included in the category of secondary data and employed for comparison purposes only.

3. METHODOLOGY

3.1. Viscosity

The viscosity of a pure gas in the limit of zero density is given by

$$\eta_0 = \frac{kT}{\langle v \rangle_0} \frac{1}{\mathfrak{S}(2000)} f_\eta = \frac{kT}{\langle v \rangle_0} \frac{1}{\mathfrak{S}_\eta} \tag{1}$$

where

$$\langle v \rangle_0 = 4(kT/\pi m)^{1/2} \tag{2}$$

$\mathfrak{S}(2000)$ is the effective collision cross section that includes all the information about binary molecular collisions, m is the molecular mass, T is the temperature, k is Boltzmann's constant, and f_η is the correction factor that accounts for higher-order terms of the kinetic theory [6]. In line with our previous work it has been found convenient to introduce the notation $\mathfrak{S}_\eta$ that incorporates $\mathfrak{S}(2000)$ as well as the higher-order correction factor, f_η. $\mathfrak{S}_\eta$ is related to the frequently used collision integral Ω_η, by $\Omega_\eta = (5/4) \, \mathfrak{S}_\eta$.

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[3] A new analysis of the results for the 425.65 K isotherm in Ref. 30 conducted during the present work indicates that the value of the zero-density thermal conductivity should be replaced by the value $\lambda_0 = 42.01$ mW·m^{-1}·K^{-1} and that the first density coefficient should be $\lambda_1 = 42.0$ μW·m^2·kg^{-1}·K^{-1}.

In a more practical form including the recently published values of the fundamental constants [46], Eq. (1) can be written as

$$\eta_0 = \frac{0.021357(TM)^{1/2}}{\sigma^2 \mathfrak{S}_\eta^*} \tag{3}$$

where η_0 is in units of μPas, the scaling parameter σ is in nm, M is the relative molecular mass, and $\mathfrak{S}_\eta^*$ is a reduced form of $\mathfrak{S}_\eta$ given by

$$\mathfrak{S}_\eta^* = \mathfrak{S}_\eta / (\pi \sigma^2) \tag{4}$$

3.2. Thermal Conductivity

The available set of experimental data has been found not sufficient to carry out the complete kinetic theory analysis as given in Refs. 1–5. This is due largely to the fact that no experimental information is available on either rotational or vibrational collision numbers covering a sufficient range of temperature. Moreover, the kinetic theory is still not sufficiently developed to deal with fluids with different modes of internal motion. In particular, the assumption of mechanically independent modes of internal motion, that is universally applied, is an unproved hypothesis in this context. Because of this situation it has been necessary to apply another scheme with regard to the correlation of the thermal conductivity of ethane. As shown earlier [47], the thermal conductivity of a gas at zero density can be expressed in the form

$$\lambda_0 = \frac{5k^2 T}{2m \langle v \rangle_0} \frac{(1 + r^2)}{\mathfrak{S}(10E)} F_\lambda \tag{5}$$

The correction factor F_λ has been found close to unity so that the rather simpler formula

$$\lambda_0 = \frac{5k^2 T}{2m \langle v \rangle_0} \frac{(1 + r^2)}{\mathfrak{S}_\lambda} \tag{6}$$

has proved useful in representing the thermal conductivity [1–5]. Here

$$r = (2C_{\text{int}} / 5k)^{1/2} \tag{7}$$

where $(C_{\text{int}} / k = C_v / k - 3/2)$ is the internal part of the isochoric heat capacity. Again, we apply a more practical form,

$$\lambda_0 = \frac{0.44392(T/M)^{1/2}(1 + r^2)}{\sigma^2 \mathfrak{S}_\lambda^*} \tag{8}$$

with

$$\mathfrak{S}_\lambda^* = \mathfrak{S}_\lambda/(\pi\sigma^2) \tag{9}$$

3.3. Correlation

In developing the correlations of the experimental data for ethane, experimental values of $\mathfrak{S}_\eta$ and $\mathfrak{S}_\lambda$ have been derived from each data source for viscosity and thermal conductivity, respectively, and then the complete set was fitted to the functional form

$$\ln \mathfrak{S}_\eta^* = \sum_{i=0}^{n} a_i(\ln T^*)^i \tag{10}$$

and

$$\mathfrak{S}_\lambda^* = \sum_{j=0}^{m} b_j(1/T^*)^j \tag{11}$$

as a function of reduced temperature

$$T^* = kT/\varepsilon \tag{12}$$

in which ε is an energy scaling parameter.

For thermal conductivity we made use of our earlier result that a simple series in $1/T^*$ (often an almost linear form) [1] provides an accurate basis for the representation of $\mathfrak{S}_\lambda^*$.

In the fitting procedure a statistical weight is assigned to each datum according to our estimate of the uncertainty of that point [7]. To apply Eq. (8) it is necessary to have a subsidiary representation of the heat capacity. In order to be able to analyze thermal conductivity data over a large temperature range, we used the representation of heat capacity given in Refs. 48 and 49 as

$$C_{\mathrm{p}}/k = C_{\mathrm{int}}/k + 5/2 = u\left(\sum_{i=1}^{7} f_i X^{(4-i)}\right) + f_8 \tag{13}$$

with

$$u = \exp(-f_9/X) \tag{13a}$$

and

$$X = 100.0/T \tag{13b}$$

The coefficients f_i are summarized in Table III.

Table III. Coefficients for the Representation
of the Reduced Heat Capacity of Ethane (C_p/k)

i	f_i
1	0.003 689 009 6
2	$-0.171\ 969\ 07$
3	3.159 226
4	$-8.045\ 994\ 2$
5	7.423 767 3
6	0.0
7	$-2.072\ 457\ 2$
8	4.0
9	0.02

3.4. Internal Consistency

Although it has been found impossible to carry out a full kinetic theory analysis, the kinetic theory provides a relationship for a consistency check.

Using Eqs. (3) and (5), the Prandtl number for a fluid can be expressed in terms of effective collision cross sections to a high degree of accuracy, as

$$Pr = \frac{C_p \eta}{m\lambda} = \frac{\mathfrak{S}(10E)}{\mathfrak{S}(2000)} \tag{14}$$

Making use of arguments by van den Oord and Korving [50], it is possible to show that in a first approximation

$$Pr \approx \frac{2}{3} + \frac{1}{3} \frac{\mathfrak{S}(0010)}{\mathfrak{S}(2000)} \tag{15}$$

where the effective cross section $\mathfrak{S}(0010)$ is related to the bulk viscosity.

At high temperatures the available experimental evidence [51, 52], supported by a few numerical calculations [53], shows that for all polyatomic gases the ratio of the effective cross sections $\mathfrak{S}(0010)/\mathfrak{S}(2000)$ is a monotonically decreasing and weak function of temperature. Therefore, it is expected that the Prandtl number of a gas should be a monotonically decreasing function of a temperature with an almost constant limiting value greater than 2/3. It is this behavior that makes the Prandtl number

potentially more useful as a check of consistency of a representation than the Eucken factor

$$f_E = \frac{m\lambda}{\eta C_v} = \frac{C_p \mathfrak{S}(2000)}{C_v \mathfrak{S}(10E)} \tag{16}$$

which has been used more frequently but for which it is more difficult to make general statements about limiting behavior.

4. RESULTS

4.1. Viscosity

The viscosity of ethane is represented by Eqs. (3) and (4) with the aid of the coefficients and parameters listed in Table IV. The coefficients used have been taken from a "universal correlation" derived for the reduced temperature range $1.2 \leqslant T^* \leqslant 10$ given by Bich et al. [54]. It has been found that, by chance, this particular correlation represents the experimental viscosity data for ethane down to about $T^* = 0.75$ ($T = 200$ K). The use of this representation has the advantage that it is possible to apply it to "extrapolate" the representation of the viscosity of ethane to a practical upper temperature limit of $T = 1000$ K ($T^* = 4$) as has been shown by corresponding-states analysis [54, 55]. It should be stressed that the fact that the low-temperature behavior of ethane can be represented by the particular universal function selected is accidental and not typical of other gases [5, 7, 54, 56, 57].

Figure 1 contains a plot of the deviations of the primary data from our representation. Most of the data are represented within the experimental uncertainty. From the comparison we conclude that we can assign an

Table IV.   Coefficients for the Representation of the Effective
Collision Cross Sections of Ethane[a]

| i	$\mathfrak{S}_\eta^*$ (a_i)	Ω_η (a_i')	$\mathfrak{S}_\lambda^*$ (b_i)
0	0.221 882	0.445 025 1	0.444 358
1	−0.507 932 2	−0.507 932 2	0.327 867
2	0.128 577 6	0.128 577 6	0.193 683 5
3	−0.008 328 17	−0.008 328 17	—
4	−0.002 713 17	−0.002 713 17	—

[a] $\varepsilon/k = 264.70$ K; $\sigma = 0.43075$ nm.
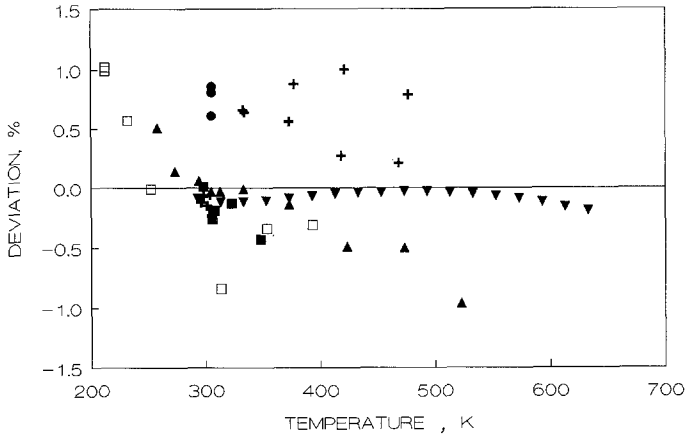
Fig. 1. Deviations of primary viscosity data from the present correlation. (□) Hunter and Smith [10]. (▼) Hendl and Vogel [11]. (+) Kestin et al. [12–14]. (■) Iwasaki and Takahashi [15]. (▲) Golubev and Mesheryakov [16]. (●) Carmichael and Sage [17].

uncertainty to the representation of ±0.5% in the temperature range 300 ≤ *T* ≤ 600 K but must increase it to ±1.5% at the lower temperature extreme and to ±2.5% at 1000 K where the data are obtained by extrapolation. Figure 2 contains a comparison with selected secondary data. The deviations from the present correlation are obviously larger, amounting to 3% in some cases.
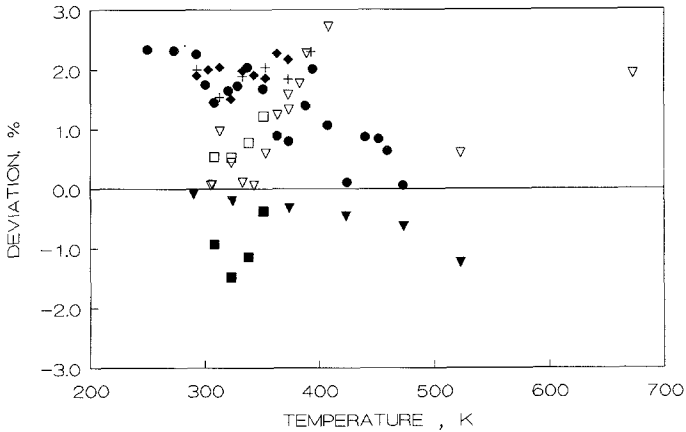
Fig. 2. Deviations of selected secondary viscosity data from the present correlation. (+) Titani [20]. (▼) Trautz and Sorg [21]. (◆) Adzumi [22]. (■) Craven and Lambert [23]. (□) Lambert et al. [25]. (●) De Rocco and Halford [26]. (▽) Diaz Pena and Cheda [28].
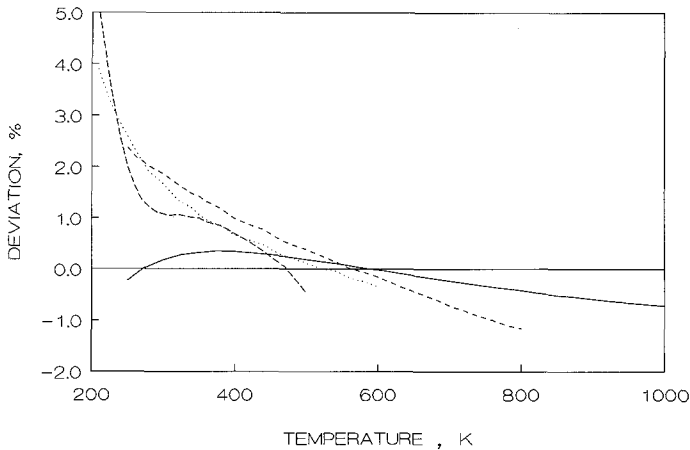
Fig. 3. Deviations of previously proposed correlations from the present correlation. (——) Boushehri et al. [55]. (——) Hanley et al. [58]. (---) Tarzimanov et al. [59]. (······) Younglove and Ely [48].

Figure 3 shows a comparison with some recent correlations including the "universal" correlations of Boushehri et al. [55] and those of Hanley et al. [58], Tarzimanov et al. [59], and Younglove and Ely [48]. The agreement above room temperature is generally within the expected range and indeed very good above 400 K. The differences at lower temperatures are undoubtedly caused by the inaccurate data that were used before the measurements of Refs. 10 and 11 became available.

4.2. Thermal Conductivity

The thermal conductivity of ethane is represented by Eqs. (7)–(9) with the aid of the coefficients and parameters listed in Table IV over the temperature range $225 \leqslant T \leqslant 725$ K.

Figure 4 shows a deviation plot of the primary data set from the proposed representation. It becomes evident that the discrepancies between the different data sets are rather large and often outside the range that has been claimed by the authors for the experimental uncertainty or indeed that ascribed to them in this work. Therefore, it is necessary to mention that the proposed correlation must remain provisional in the sense that further high-precision measurements are necessary especially at higher temperatures.

Figure 5 shows a deviation plot for selected secondary data that confirms the large scatters mentioned earlier and the fact that the uncertainty in the thermal conductivity is significantly higher than that of the viscosity.
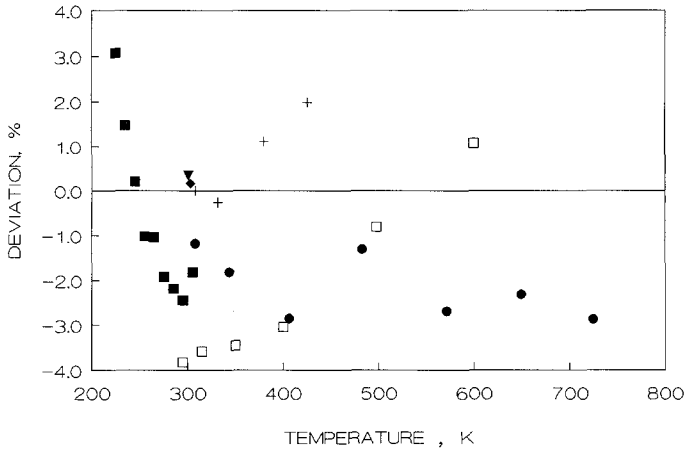
Fig. 4. Deviations of primary thermal conductivity data from the present correlation. (+) Millat et al. [30]. (▼) Fleeter et al. [31]. (◆) Clifford et al. [32]. (■) Roder and Nieto de Castro [33]. (□) Prasad and Venart [34]. (●) Le Neindre et al. [35].

We also include a comparison with a recent correlation of Younglove and Ely [48] which itself strongly depends on the selected data basis. Therefore, again the differences are rather large compared to viscosity and also compared to our previous work for other gases. As a result of our analysis it is estimated that this uncertainty associated with the present representa-
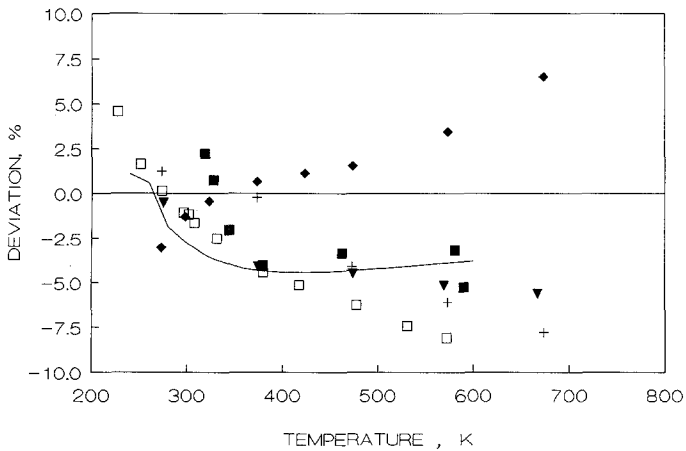
Fig. 5. Deviations of selected secondary thermal conductivity data from the present correlation. (+) Geier and Schäfer [39]. (◆) Senftleben [40]. (▼) Schramm and Schäfer [42]. (□) Rjabcev and Kasarjan [43]. (■) Vargaftik [44]. (———) Younglove and Ely [48].

Table V.   The Viscosity, Thermal Conductivity, Reduced Heat Capacity,
Prandtl Number, and Eucken Factor of Ethane at Selected Temperatures[a]

Temperature (K)	Viscosity (μPas)	Thermal conductivity ($\text{mW} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)	C_p/k	Prandtl number	Eucken factor
250	6.138	—	5.056	—	—
250	7.762	15.48	5.658	0.7845	1.548
300	9.313	21.76	6.366	0.7535	1.575
350	10.79	28.94	7.118	0.7341	1.585
400	12.21	36.86	7.880	0.7216	1.587
450	13.56	45.37	8.631	0.7134	1.585
500	14.86	54.33	9.359	0.7079	1.582
550	16.11	63.62	10.06	0.7043	1.577
600	17.32	73.13	10.72	0.7020	1.571
650	18.48	82.78	11.35	0.7006	1.565
700	19.61	92.49	11.94	0.6999	1.559
750	20.70	(102.2)	12.49	(0.7)	1.554
800	21.76	(111.8)	13.01	(0.7)	1.548
850	22.79	(121.5)	13.50	(0.7)	1.543
900	23.80	(131.1)	13.95	(0.7)	1.537
950	24.78	(140.6)	14.37	(0.7)	1.532
1000	25.73	(150.0)	14.76	(0.7)	1.528

[a] Thermal conductivity data in parentheses are estimated assuming a constant value $Pr = 0.7$.

tion may be as much as $\pm 2\%$ in the temperature range $300 \leqslant T \leqslant 500$ K, rising to $\pm 3\%$ at both 225 and 725 K.

Table V contains a brief listing of the viscosity and thermal conductivity of ethane at selected temperatures to enable users to check their coding of the present correlations. We also present results for the Prandtl number and the Eucken factor. As expected, the Prandtl number is decreasing with increasing temperature and seems to reach an almost constant value of $Pr = 0.7$. Therefore, our results are in qualitative agreement with the results of the kinetic theory embodied in equation (15). The existence of an almost constant limiting value for Pr [50] provides the possibility to estimate values for the thermal conductivity outside of the experimental range. The results of this estimation procedure between 750 and 1000 K are also presented in Table V. To indicate the larger uncertainty, we give these numbers in parentheses.

5. CONCLUSION

Correlations for the viscosity and thermal conductivity of ethane have been presented. The results provide the basis for a detailed analysis of these

transport properties over a wide range of thermodynamic states. For the viscosity the representation extends over the temperature range $200 \leqslant T \leqslant 1000$ K, with the associated uncertainty of $\pm 0.5\%$ in the $300 \leqslant T \leqslant 600$ K range increasing to $\pm 1.5\%$ at 200 K and $\pm 2.5\%$ at 1000 K. The thermal conductivity correlation extends over the temperature range $225 \leqslant T \leqslant 725$ K, with the associated uncertainty of 2% in the $300 \leqslant T \leqslant 500$ K range increasing to $\pm 3\%$ at both 225 and 725 K.

Estimates of the thermal conductivity above 725 K are also given. From this study it becomes evident that further accurate measurements of the thermal conductivity of ethane are required.

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