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The Viscosity of Carbon Dioxide

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When representative equations for the viscosity of carbon dioxide were published in 1990, it was recognized that, owing to inconsistencies among the available experimental liquid viscosity data which could not be resolved, new measurements were necessary. Since then, two new sets of measurements have been performed and it is appropriate to revise the published equations in order to improve their performance in the liquid region. In the previous work, the excess viscosity was represented by two separate equations, one for the gas phase and the other, a provisional one, for the liquid phase. Both equations were joined by a blending function. In the present work, the excess viscosity for the whole thermodynamic surface is represented by one equation. The resulting overall viscosity representation for carbon dioxide covers the temperature range $200\ \text{K} \leqslant T \leqslant 1500\ \text{K}$ and densities up to $1400\ \text{kg m}^{-3}$. In terms of pressure, the viscosity representation is valid up to 300 MPa for temperatures below 1000 K, whereas for higher temperatures and owing to the limitation of the equation of state used, the upper pressure limit is restricted to 30 MPa. The uncertainties associated with the proposed representation vary from $\pm 0.3\%$ for the viscosity of the dilute gas near room temperature to $\pm 5.0\%$ at the highest pressures. Tables of viscosity generated by the representative equations are included for easy reference and to assist validation of computer coding. © *1998 American Institute of Physics and American Chemical Society.* [S0047-2689(98)00101-9]

Key words: carbon dioxide; correlation; liquid viscosity; saturation line; viscosity.

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1. Introduction

The unresolved inconsistencies among the various literature sources of viscosity measurements available at the time of the development of the representative equations of the viscosity of carbon dioxide[1] caused the adoption of a representation of the liquid-phase data by a separate equation from that for the gas phase which was joined to the latter by a blending function. The resulting equations were rather awkward to implement and the representation of the liquid region was necessarily less satisfactory than desirable. In the light of new measurements by van der Gulik[2] and Padua et al.[3] from two independent laboratories, which were prompted by the work reported in Ref. 1, it was decided to refit the excess viscosity in order to improve the representation of the liquid region. The object of the present work is therefore to remedy the deficiency of the earlier representation of the liquid region and to present a unified treatment of the excess viscosity of carbon dioxide. The work, however, relies on the assessment of the experimental data carried out earlier[1] and makes it the basis for the overall excess viscosity representation developed here.

2. Equation of State

The proposed representation of the viscosity of carbon dioxide expresses the viscosity in the customary way as a function of density and temperature. In practice, the majority of the experimental viscosity measurements are reported at specified pressures and temperatures, thus making the use of an accurate equation of state essential. The thermodynamic formulation for carbon dioxide used here was the analytic equation proposed by Ely et al.[4] The equation is applicable in the range extending from 217 K to 1000 K for pressures up to 300 MPa. The equation of Ely et al.[4] is used partly because it provides good representation of the thermodynamic surface and partly because it is consistent with the scaled fundamental equation proposed by Albright et al.[5] which was used for the treatment of the critical region.

3. Methodology

Following a method which has now become standard, the viscosity is decomposed into three separate contributions:

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

or

$$\eta(\rho,T) = \overline{\eta}(\rho,T) + \Delta\eta_c(\rho,T), \qquad (2)$$

where $\eta_0(T)$ is the viscosity in the zero-density limit, $\Delta\eta(\rho,T)$ an excess viscosity which represents the increase in the viscosity at elevated density over the dilute gas value at the same temperature, and $\Delta\eta_c(\rho,T)$ a critical enhancement accounting for the increase in viscosity in the immediate vicinity of the critical point. The first two terms on the right hand side of Eq. (1) are sometimes grouped together and the resulting quantity is termed the background contribution $\overline{\eta}(\rho,T)$.

The treatment of the viscosity in the zero-density limit, $\eta_0(T)$, and the critical enhancement contribution have been dealt with by Vesovic et al.[1] and will not be repeated here because no new information is available. However, we reproduce below the equation of the viscosity in the zero-density limit in order to facilitate the calculation of the background viscosity contribution. The equations provided in this paper will therefore be sufficient to calculate the viscosity of carbon dioxide across most of the thermodynamic surface except in a region bounded approximately by 300 K $<T<$ 310 K and 300 kg m$^{-3}<\rho<$ 600 kg m^{-3}, where the relative critical viscosity enhancement $\Delta\eta_c/\eta$ is higher than 1%. Therefore, in the proximity of the critical region, the equations of this paper should be supplemented by Eq. (39) of Ref. 1.

3.1. Viscosity in the Zero-Density Limit

The representation of the viscosity of carbon dioxide in the limit of zero density has been investigated by Trengove and Wakeham[6] and has been further studied by Vesovic et al.[1] when the data of Vogel and Barkow[7] became available. Since the work of Vesovic et al.,[1] two sets of measurements have been reported by Hunter et al.[8] and Hendl et al.[9] Hunter et al.[8] measured the viscosity of carbon dioxide in the dilute region with a capillary flow viscometer in the temperature range (213–353) K with an accuracy of ±0.7%. Hendl et al.[9] reported experimental data in the temperature range (297–685) K at molar densities between 0.005 and 0.062 mol l^{-1} with an estimated accuracy of ±0.15% at the lowest temperature and ±0.2%–±0.3% at the high temperature range. They also extrapolated their data along isotherms to zero-density in order to determine the zero-density viscosity values. The deviations of the resulting zero-viscosity values and those of the dilute-gas viscosity data of Hunter et al.[8] from the zero-viscosity representation of Vesovic et al.[1] are within experimental uncertainties as shown by Fig. 1. In view of the agreement of the zero-density viscosity equation of Vesovic et al.[1] with the two recent sets of data, it was decided to adopt this equation without further refinement in the present work. For completeness we quote the expression for the zero-density viscosity,
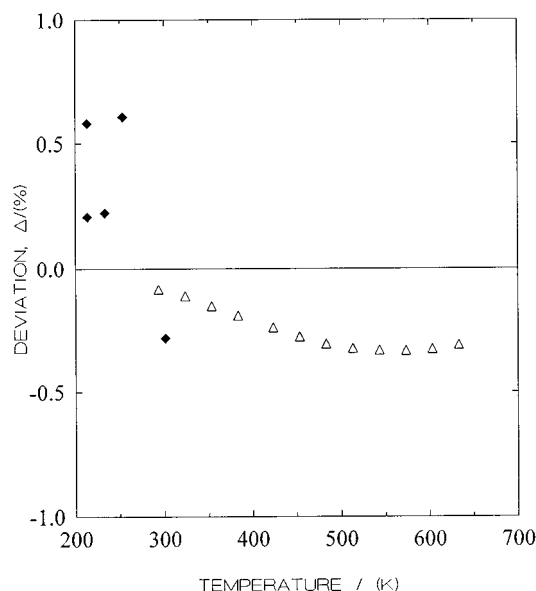
FIG. 1. Deviations of the data of Hunter et al. (Ref. 8) and Hendl et al. (Ref. 9) from the zero-density viscosity representation as a function of temperature. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc})/\eta_{exp}$. ($\blacklozenge$) Hunter et al. (Ref. 8), ($\triangle$) Hendl et al. (Ref. 9).

$$\eta_0(T) = \frac{1.00697 T^{1/2}}{\mathfrak{S}_\eta^*(T^*)}, \tag{3}$$

where the zero-density viscosity, $\eta_0(T)$, is in units of μPa s and the temperature, T, in Kelvin. The reduced effective cross section, $\mathfrak{S}_\eta^*(T^*)$, is represented by the empirical equation,

$$\ln \mathfrak{S}_\eta^*(T^*) = \sum_{i=0}^{4} a_i (\ln T^*)^i, \tag{4}$$

where the reduced temperature T^* is given by

$$T^* = kT/\varepsilon, \tag{5}$$

and the energy scaling parameter is $\varepsilon/k = 251.196$ K. The coefficients a_i of Eq. (4) are listed in Table 1.

3.2. Viscosity in the Critical Region

Theoretical and experimental evidence indicates that the viscosity of fluids diverges at the critical point[10] ($\rho_c = 467.69$ kg/m^{-3}, $T_c = 304.107$ K, $P_c = 7.3721$ MPa). However, unlike the critical enhancement in thermal conductivity, that in vis-

TABLE 1. Coefficients of the representation of the zero-density viscosity of carbon dioxide (T in K; η_0 in μPa s)

i	a_i
0	0.235156
1	−0.491266
2	5.211155×10^{-2}
3	5.347906×10^{-2}
4	-1.537102×10^{-2}

cosity is much less pronounced and is restricted to a much narrower range of conditions around the critical point. For a number of fluids, it has been found that the ratio of $\Delta \eta_c(\rho,T)/\eta(\rho,T)$ is greater than 0.01 only within 1% ($\sim$5 K) of the critical temperature. The critical enhancement has been amply dealt with by Vesovic et al.[1] For its calculation, the reader is referred to their work, the results of which are used unchanged here.

3.3. Excess Viscosity

The excess viscosity contribution describes how the viscosity of the fluid behaves as a function of density outside of the critical region. The current state of knowledge does not provide a satisfactory theory for the calculation of the excess viscosity over the whole phase space. However, in practice, a power series expansion in density for the representation of the excess viscosity has often been found to be adequate.[1,11–13] The series adopted here is of the following form

$$\Delta \eta(\rho,T) = \sum_{i=1}^{n} b_i(T)\rho^i. \tag{6}$$

It is customary to represent the temperature dependence of the density coefficients, $b_i(T)$, by a functional relationship of the form

$$b_i = \sum_{j=1}^{m} d_{ij}/T^{*(j-1)}, \tag{7}$$

where the reduced temperature, T^*, is defined by Eq. (5) and the coefficients d_{ij} are to be determined by fitting the primary experimental data for the excess viscosity to Eqs. (6) and (7).

4. Data Selection and Fitting Procedure

A comprehensive critical analysis of the experimental data has been carried out by Vesovic et al.[1] In what follows we confine our analysis to the liquid region where the earlier analysis revealed inconsistencies among the available sources of data that could not be resolved. It is now accepted[1] that old liquid viscosity data are not accurate by comparison with recent data owing to either the nonavailability of accurate instruments or the lack of full working equations for the instruments employed. For this reason, the experimental data predating 1957 were not considered. The first accurate measurements of the viscosity of liquid carbon dioxide were made by Michels et al.[14] who carried out measurements along two isotherms using a capillary viscometer in the van der Waals laboratory in 1957. Since then five extensive measurement sets have been reported. Golubev and Shepeleva[15] performed measurements in the temperature range 242 K$\leq T \leq$294 K and the pressure range 5 MPa $\leq P \leq$50 MPa. However, the measurements were not carried out along isotherms. They used a capillary viscometer and claimed an experimental accuracy of $\pm$1%. Ulybin and

TABLE 2. List of recent experimental sources of data on the viscosity of liquid carbon dioxide

Author(s)	Method	T/K	P/MPa	Ascribed accuracy (%)
Primary set				
van der Gulik[2]	vibrating wire	220–303	0.6–453	±2
Padua et al.[3a]	vibrating wire	260–300	6–100	±2
Michels et al.[14]	capillary	298–303	6.6–134	±1
Golubev and Shepeleva[15]	capillary	242–294	5.0–50	±3
Secondary set				
Ulybin and Makarushkin[16–18]	capillary	223–293	6.0–55	
Diller and Ball[19]	oscillating quartz	220–300	2.5–28	
Herreman et al.[20]	oscillating quartz	219–303	1.0–19	

[a]Data along the 300 K isotherm have not been used as primary data.

Makarushkin[16–18] used the same type of instrument to make measurements in the temperature range 223 K $\leq T \leq$ 293 K and the pressure range 6 MPa $\leq P \leq$ 55 MPa with a reported accuracy of ±1.7%. Diller and Ball[19] and Herreman et al.[20] employed an oscillating quartz-crystal viscometer to perform measurements over a similar range of conditions. Diller and Ball[19] claimed an accuracy of ±2%. Padua et al.[3] carried out measurements along three isotherms, 260 K, 280 K, and 300 K at pressures from 6 MPa to 100 MPa by using a vibrating-wire viscometer. The accuracy was stated to be ±0.5%. Finally, van der Gulik,[2] using a similar type of instrument measured extensively the viscosity of carbon dioxide in the temperature range 220 K $\leq T \leq$ 303 K and the pressure range 0.6 MPa $\leq P \leq$ 453 MPa with an estimated accuracy of ±1%. The above mentioned sources of experimental data are summarized in Table 2, together with our estimates of their accuracy. The estimates of accuracy were based primarily on the authors' reported values with some modifications where consistency checks or examinations of previous measurements with the same apparatus on other fluids implied larger uncertainty bands.

Since the current state of the kinetic theory does not provide us with an effective means of discriminating between data, an empirical fitting approach, which combines careful scrutiny of the experimental sources of data with statistical confidence tests for selecting the significant coefficients of the fit, remains the only viable approach for the development of a representative equation for the viscosity. It is this approach which has been employed in order to select a primary data set on which to base the development of a new empirical equation for the excess viscosity of carbon dioxide.

Large discrepancies between the data of Golubev et al.,[15] Ulybin and Makarushkin,[16–18] and Diller and Ball[19] were observed and reported by Vesovic et al.[1] In the initial course of development of the present excess viscosity representation, all of the liquid viscosity data contained in Table 2 except those of Herreman et al.[20] were used to generate preliminary fits. The data of Herreman et al.[20] were excluded from the analysis because both the type of instrument used and the data analysis were incompletely described.[1] Each liquid data set was initially ascribed the uncertainty reported by the authors. In order to investigate the compatibility of the liquid data sets with the viscosity in the supercritical region,

the primary gas-phase viscosity data selected by Vesovic et al.[1] were added to the body of the liquid viscosity data. Preliminary fits based on all the excess viscosity data referred to above were generated using Eqs. (6) and (7). The deviation plot of each data source was scrutinized for systematic departure from the overall data trend. The data sets reported along similar isotherms by different authors were also compared with each other for consistency. Figure 2 depicts the deviations of all the employed liquid data sets from one of such preliminary representations in which the data were weighted according to their reported uncertainties. The extent of the deviations of a particular liquid data set is an indication of its departure from the consensus of other sets. Thus, the data of Diller and Ball[19] were found to lie systematically above the representations and to depart from them by up to 6%. Furthermore, all the isotherms exhibited different density behaviors with respect to corresponding isotherms of

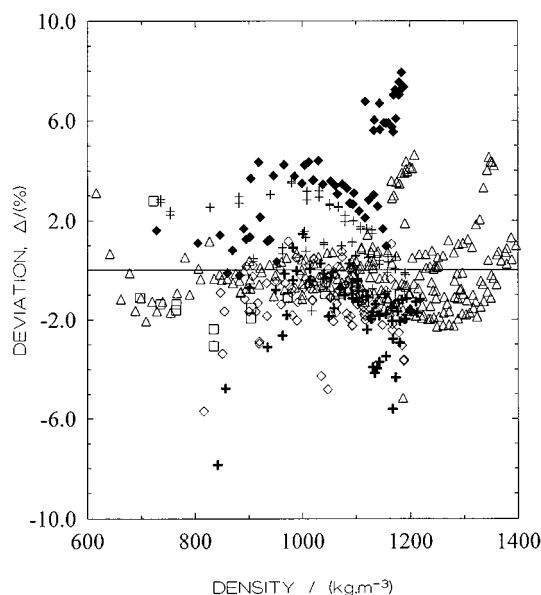FIG. 2. Deviations of all the liquid data from a preliminary representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc})/\eta_{exp}$. (+) Padua et al. (Ref. 3), ($\triangle$) van der Gulik (Ref. 2), ($\blacklozenge$) Diller and Ball (Ref. 19), (+) Ulybin and Makarushkin (Refs. 16–18), ($\diamond$) Golubev and Shepeleva (Ref. 15), ($\square$) Michels et al. (Ref. 14).

TABLE 3. Coefficients of the representation of the excess viscosity of carbon dioxide, Eq. (7). [T in K; $\Delta\eta(\rho,T)$ in μ Pa s and ρ in kg m^{-3}]. The rest of the d_{ij} coefficients are equal to zero

d_{11}	0.4071119×10^{-2}
d_{21}	0.7198037×10^{-4}
d_{64}	0.2411697×10^{-16}
d_{81}	0.2971072×10^{-22}
d_{82}	-0.1627888×10^{-22}

other workers. The data of Ulybin and Makarushkin[16–18] were also found to have systematic deviations, with the (223–243) K isotherms showing deviations 1%–8% below the representations. Consequently, both sets of data were excluded from the primary set.

The data sets of Padua et al.[3] and van der Gulik[2] along the 300 K isotherm were also found to be inconsistent with each other within their reported uncertainty. At the preliminary analysis stage, it was not possible to identify which set was more accurate or consistent with the supercritical viscosity data. Nevertheless, subsequent analysis indicated that the 300 K isotherm of van der Gulik[2] was consistent with the high density supercritical data of Ref. 1. Therefore, both the 300 K and the 303 K isotherms of van der Gulik[2] were included in the primary data set. However, the measurements reported by van der Gulik[2] in metastable regions (superheated and undercooled liquid) were excluded from the fit because of some concern about the accuracy of the thermodynamic data in these regions. The remaining data identified in Table 2 as primary set, together with the gas-phase data selected by Vesovic et al.,[1] were employed to generate a general excess viscosity correlation for the whole phase space for which experimental data are available. In this process each datum was given a weight dependent on our estimated uncertainty and not necessarily on the reported uncertainty by the authors. The ascribed uncertainty for each primary data set is given in Table 2. The fitting was performed by means of the SEEQ technique,[21] which is described in Ref. 1. The resulting coefficients, as well as the optimum values of i and j, of Eq. (6) and (7) are given in Table 3.

The excess viscosity correlation can be written explicitly as:

$$\Delta\eta(\rho,T)=d_{11}\rho+d_{21}\rho^2+\frac{d_{64}\rho^6}{T^{*3}}+d_{81}\rho^8+\frac{d_{82}\rho^8}{T^*}. \quad (8)$$

5. Results and Discussion

In order to assess the accuracy and to determine the region of validity of the proposed representation of the viscosity of carbon dioxide, deviation plots of all the analyzed experimental data were generated and studied. Figure 3 illustrates the deviations of the data of van der Gulik[2] from the proposed representation. The majority of the primary data of van der Gulik[2] are represented within ±3% with the exception of the high density data along the 260 K isotherm which

FIG. 3. Deviations of the data of van der Gulik (Ref. 2) from the representation. The deviations are defined as: $\Delta=100\times(\eta_{\mathrm{exp}}-\eta_{\mathrm{calc}})/\eta_{\mathrm{exp}}$. ($+$) 220 K isotherm, ($\triangle$) 230 K isotherm, ($\bigcirc$) 240 K isotherm, ($+$) 260 K isotherm, ($\blacktriangledown$) 280 K isotherm, ($\diamond$) 300 K isotherm, ($\square$) 303 K isotherm.

exhibit a systematic upward trend, reaching about 7% at the high density end. In fact, at densities above 1300 kg m^{-3} the experimental data along the (260–303 K) isotherms show similar systematic trends, albeit remaining within the ±2% ascribed uncertainty. The steep increase in viscosity at high densities has been observed by a number of workers[22] for some fluids. Indeed, it has been suggested that an excess viscosity representation based on a power series in density such as Eqs. (6) and (7) may not represent adequately the observed behavior, and consequently a number of representations containing exponential terms have been proposed. We tried to incorporate such exponential terms in the preliminary chosen density functions in order to improve the representation of the high density data of van der Gulik but no noticeable improvement was achieved. The data of van der Gulik[2] along the 260 K isotherm were still found to have high deviations. There are no other independent data at such high densities to confirm the behavior of this particular isotherm at densities higher than 1300 kg m^{-3}. On the basis of the available experimental data, it is not therefore possible to conclude that a representation based on a density power series may not be suitable. Until additional information becomes available, we propose to keep the present viscosity representation but to increase its uncertainty at densities above 1300 kg m^{-3} and temperatures above 260 K to ±5%.

Figure 4 shows the deviations of the data of Padua et al.[3] from the present representation. The data along the 260 K isotherm is represented within ±0.5% and is in good agreement with the data of van der Gulik[2] in their region of overlap. The primary data along the 280 K isotherm deviate systematically from the representation by up to 3%, with the deviations decreasing with increasing density. However the

FIG. 4. Deviations of the data of Padua *et al.* (Ref. 3) from the representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc})/\eta_{exp}$. ($\bigcirc$) 260 K isotherm, ($\blacklozenge$) 280 K isotherm, ($\Diamond$) 300 K isotherm.
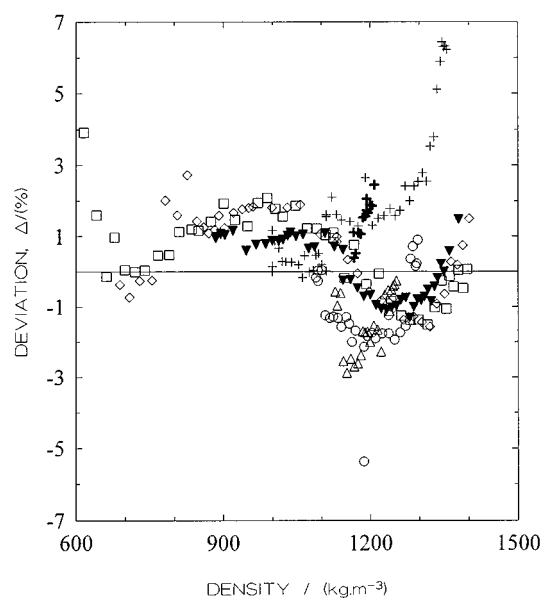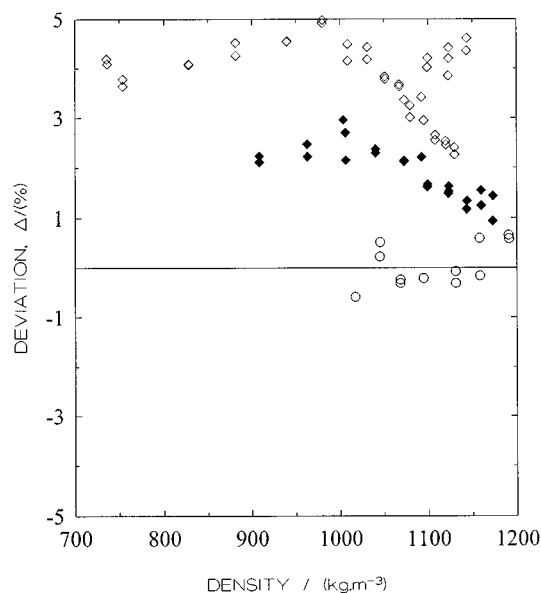

FIG. 6. Deviations of the data of Golubev and Shepeleva (Ref. 15) from the representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc})/\eta_{exp}$. ($\bigcirc$) 242–294 K

majority of the data fall within the ascribed uncertainty of $\pm 2\%$. The secondary data along the 300 K isotherm lie outside the ascribed uncertainty and deviate by up to 5% from the representation. They show a similar behavior to those along the 280 K isotherm in terms of falling deviations with increase in density.

The deviations of the data of Michels *et al.*[14] from the representation are illustrated in Fig. 5 which shows that their data are represented by the correlation within $\pm 1\%$ except

FIG. 5. Deviations of the data of Michels *et al.* (Ref. 14) from the representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc})/\eta_{exp}$. ($\blacklozenge$) 298 K isotherm, ($\bigcirc$) 303 K isotherm.

for one datum. The deviations of the data of Golubev and Shepeleva[15] are shown in Fig. 6. The majority of their data is represented within $\pm 3\%$, with the highest and lowest temperature isotherms showing the highest deviations.

The excess viscosity correlation developed in this work results in an improvement of the liquid viscosity region with respect to the provisional liquid representation proposed by Vesovic *et al.*[1] as illustrated by Fig. 7 for four isotherms (220, 260, 280, and 300) K along which experimental data are available for comparison. It is, however, entirely consistent with that developed for the gas-phase by Vesovic *et al.*[1] within its uncertainty. The deviations of the gas-phase primary data selected by Vesovic *et al.*[1] have been calculated using the present representation and compared to the results of the earlier representation.[1] The corresponding deviation plots have been found to be identical to those reported by Vesovic *et al.*[1] and are therefore not reproduced here.

The data not used in the fit were classed as secondary data and are shown in Table 2. They were used only for comparison purposes with respect to the final correlation. The deviations from the representation for each data set have been calculated. The deviations of the data of Ulybin and Makarushkin[16–18] are shown in Fig. 8. The data along the 273 K, 253 K, and 243 K isotherms are represented within $\pm 2.5\%$, while the rest of the data exhibit systematic deviations from the representation reaching up to -8%. The deviations of the data of Diller *et al.*[19] are shown in Fig. 9 which clearly demonstrates systematic deviations ranging from $+1\%$ to $+7\%$ from the present representation.

The new general excess viscosity [Eq. (8)] is entirely consistent with the gas phase excess correlation of Vesovic *et al.*[1] which has three temperature-independent coefficients. In fact, the deviations of the gas-phase data from the present
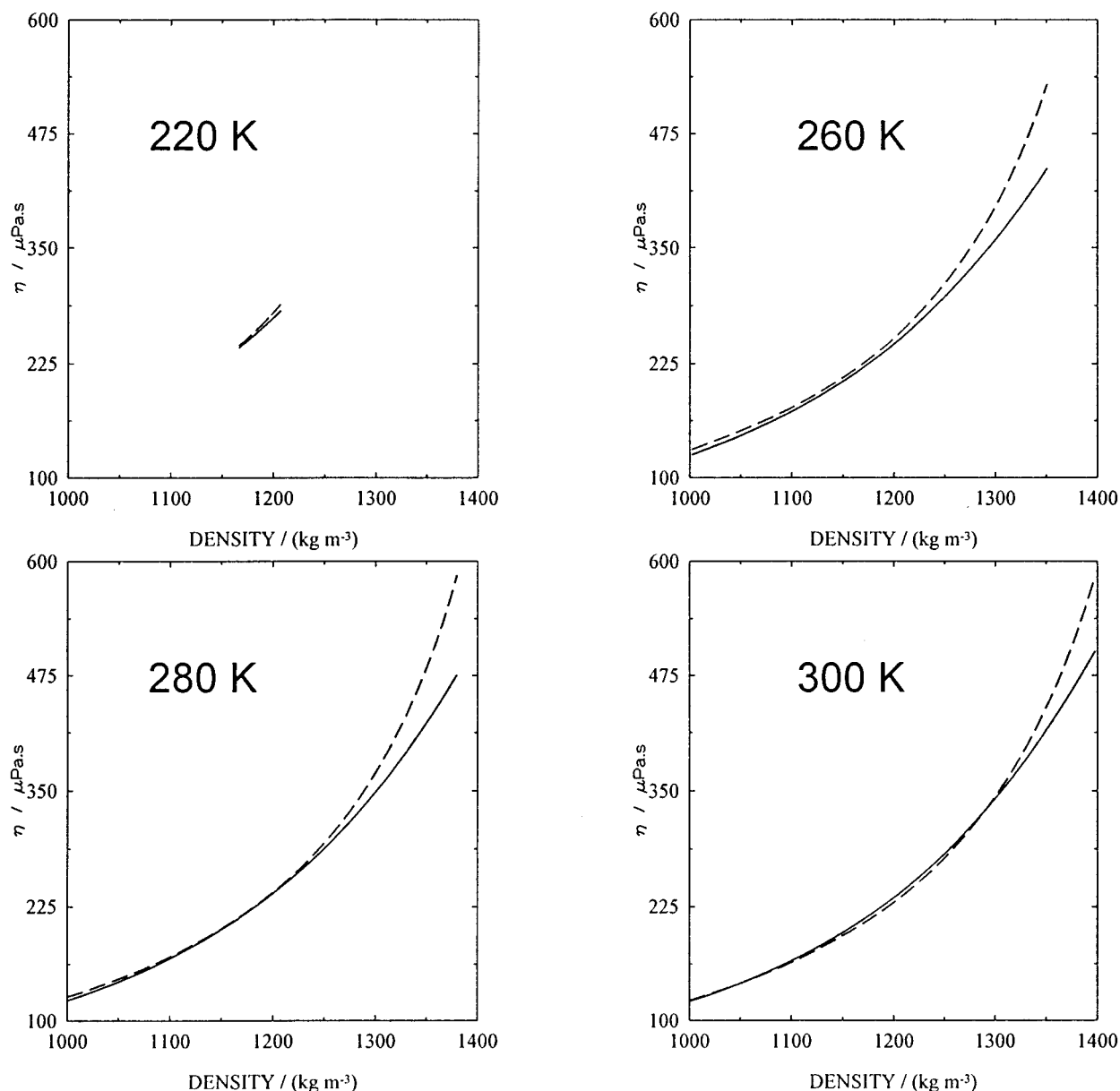
FIG. 7. Comparison of liquid viscosity values calculated along four isotherms from the present representation with corresponding values obtained from that of Vesovic et al. (Ref. 1) (−) present representation, (−−−) representation of Vesovic et al. (Ref. 1).

representation and that of Vesovic et al.[1] are nearly identical. The new correlation has, however, five coefficients among which two are temperature-dependent. The temperature-dependent coefficients have been found essential for an optimal representation of the liquid viscosity data, although it still remains true that the excess viscosity of carbon dioxide is practically temperature-independent within the supercritical region. This temperature-independence of the excess viscosity can be used as described in Ref. 1, together with the zero-density equation, to generate the viscosity up to 1500 K, which is the upper limit of the zero-density representation, and densities up to 1400 kg m^{-3}. However, the equation of state of Ely et al.[4] is valid only up to 1000 K; therefore, it is

not possible to extend the validity of the viscosity representation to pressures corresponding to densities of 1400 kg m^{-3}. For temperatures above 1000 K, the upper pressure limit has been set to 30 MPa. Up to this pressure value, the calculated density using an ideal gas behavior will not produce an error in viscosity greater than 0.5%. This error is an order of magnitude smaller than the ascribed uncertainty of the representation in this region.

6. Tabulations

Tabulations of the viscosity of carbon dioxide over the temperature range of the representation, including the satu-
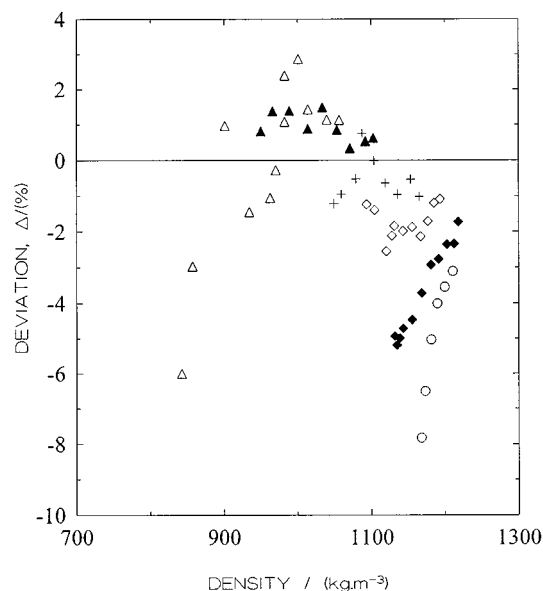
FIG. 8. Deviations of the data of Ulybin and Makarushkin (Refs. 16–18) from the representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{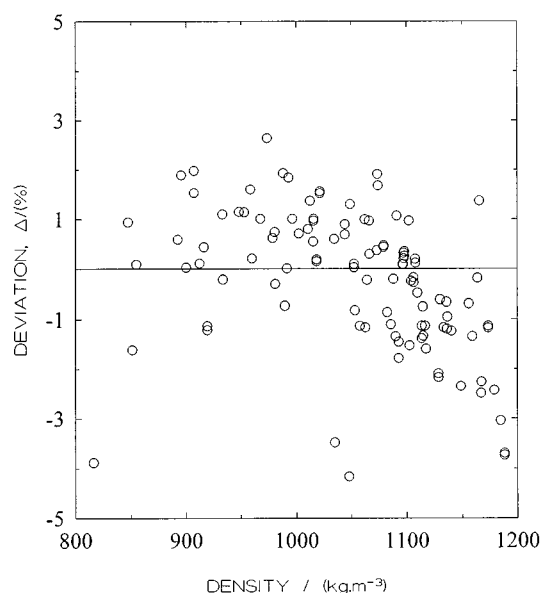calc}) / \eta_{exp}$. (○) 223 K isotherm, (◆) 233 K isotherm, (◇) 243 K isotherm, (+) 253 K isotherm, (▲) 273 K isotherm, (△) 293 K isotherm.

ration line are provided in Appendices I–III. The tabulations have been generated directly from Eqs. (3), (8), and Eq. (39) of Ref. 1 as a function of pressure and temperature using the equations of state of Ely *et al.*[4] and Albright *et al.*[5] In order to assist those programming the representative equations with the checking of the code, a small table in Appendix IV giving viscosity as a function of temperature and density is included.
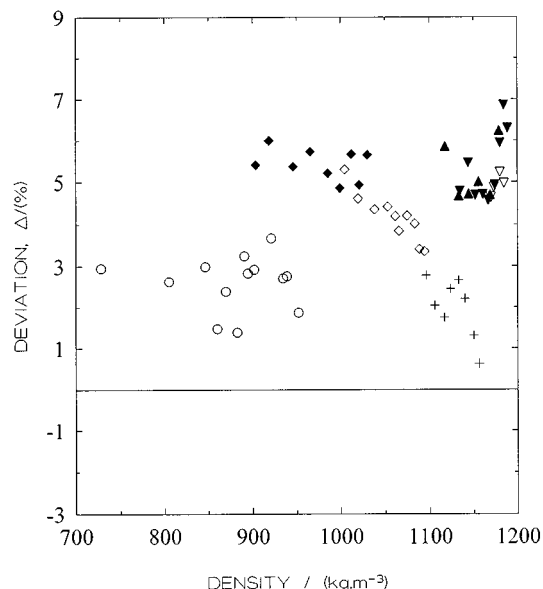
FIG. 9. Deviations of the data of Diller and Ball (Ref. 19) from the representation. The deviations are defined as: $\Delta = 100 \times (\eta_{exp} - \eta_{calc}) / \eta_{exp}$. (○) 300 K isotherm, (◆) 280 K isotherm, (◇) 260 K isotherm, (+) 240 K isotherm, (▲) 233 K isotherm, (▼) 230 K isotherm, (▽) 220 K isotherm.
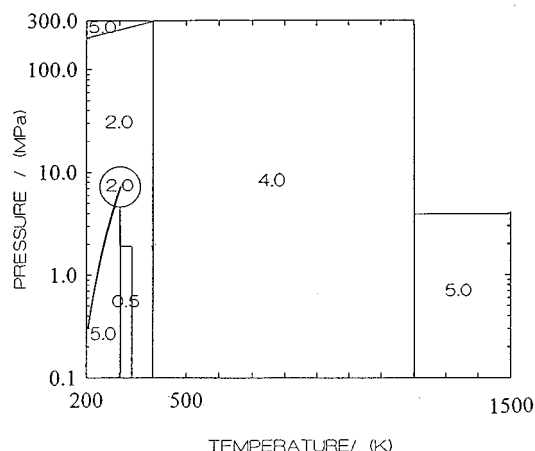
FIG. 10. The extent of the viscosity representation and its estimated uncertainty.

7. Conclusion

A critical assessment has been made of the available experimental data for the dynamic viscosity of liquid carbon dioxide. An excess viscosity equation has been developed to represent the excess viscosity of carbon dioxide over a wide range of temperatures and pressures. The new correlation is consistent with the earlier gas-phase correlation developed by Vesovic *et al.*[1] Tables of values of the viscosity have been prepared together with an assessment of accuracy. The representative equations reproduce all of the data within their ascribed uncertainties. We note here that the new measurements of van der Gulik[2] and Padua *et al.*,[3] while not entirely consistent, are nevertheless in sufficient agreement to be able to resolve the difficulties reported in Ref. 1.

The comparison of the viscosity values calculated from the proposed representation with the corresponding experimental data provides the basis for estimating the accuracy of the overall representation over the validity range of thermodynamic states. For the liquid region, the accuracy of the representation is estimated to be ±2%, except at temperatures above 260 K for pressures above 250 MPa where the uncertainty increases to ±5%. The accuracy of the representation in the gas-phase remains as quoted in Ref. 1. Comparison of liquid viscosity values up to pressures of 453 MPa calculated from the present representation with the experimental data of van der Gulik[2] suggests that the representation can be extrapolated up to such conditions. In this case, the values yielded have an uncertainty of ±2%. A summary of the extent of the viscosity representation and its estimated uncertainty are presented in the form of a tolerance diagram shown in Fig. 10.

8. Acknowledgments

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10. Appendix I. Tabulations of the Viscosity of Carbon Dioxide η/μ Pa s

T/K (P/MPa)	200	220	240	260	280	300	320	340	360	380
0.1	10.06	11.06	12.07	13.06	14.05	15.02	15.98	16.93	17.87	18.79
0.5		11.12	12.11	13.10	14.09	15.06	16.02	16.96	17.90	18.82
1.0		242.46	12.20	13.18	14.15	15.11	16.07	17.01	17.94	18.86
2.5		245.44	175.03	124.07	14.51	15.41	16.32	17.23	18.13	19.03
5.0		250.36	179.79	129.96	90.41	16.72	17.24	17.95	18.73	19.54
7.5		255.20	184.41	134.85	96.86	60.47	19.78	19.48	19.85	20.44
10.0		259.98	188.91	139.48	102.42	71.13	32.58	22.80	21.8	21.86
12.5		264.70	193.30	143.90	107.27	78.09	51.13	30.10	25.19	24.02
15.0		269.37	197.61	148.15	111.94	83.74	60.11	40.23	30.29	27.05
17.5		273.99	201.85	152.26	116.38	88.64	66.51	48.43	36.47	30.93
20.0		278.57	206.01	156.25	120.07	93.06	71.74	54.76	42.46	35.29
22.5		283.11	210.11	160.14	124.12	97.07	76.29	59.97	47.74	39.71
25.0		287.61	214.16	163.95	127.85	101.08	80.39	64.45	52.36	43.90
27.5			218.16	167.68	131.47	104.66	84.16	68.45	56.47	47.77
30.0			222.11	171.34	134.98	108.29	87.68	72.09	60.18	51.34
35.0			229.90	178.50	141.76	114.88	94.25	78.67	66.76	57.72
40.0			237.54	185.47	148.28	120.65	100.20	84.55	72.56	63.35
45.0			245.07	192.29	154.59	126.81	105.87	90.06	77.85	68.44
50.0			252.50	198.97	160.73	132.55	111.64	95.21	82.77	73.15
55.0			259.84	205.55	166.74	138.12	116.53	100.07	87.45	77.57
60.0			267.11	212.03	172.63	143.55	121.62	104.80	91.94	81.79
65.0			274.30	218.43	178.44	148.87	126.55	109.31	96.15	85.80
70.0			281.43	224.76	184.16	154.10	131.37	113.99	100.27	89.71
75.0			288.51	231.03	189.81	159.25	136.10	118.66	104.31	93.49
80.0			295.53	237.24	195.40	164.34	140.76	122.58	108.38	97.20
85.0			302.51	243.41	200.93	169.37	145.36	126.81	112.39	100.86
90.0			309.45	249.53	206.42	174.35	149.91	130.99	115.92	104.31
95.0			316.35	255.61	211.87	179.28	154.41	135.12	119.88	107.85
100.0			323.21	261.65	217.28	184.18	158.87	139.21	123.72	111.48
125.0				291.42	243.90	208.22	180.75	159.21	142.09	128.34
150.0				320.61	269.97	231.75	202.14	178.75	160.02	144.84
175.0				349.37	295.68	254.98	223.26	198.06	177.73	161.15
200.0				377.80	321.13	277.99	244.22	217.24	195.35	177.38
225.0				405.96	346.37	300.86	265.08	236.35	212.94	193.61
250.0				433.90	371.46	323.62	285.88	255.45	230.53	209.87
275.0				461.64	396.41	346.30	306.64	274.55	248.16	226.19
300.0				489.21	421.26	368.93	327.39	293.67	265.83	242.57

Tabulations of the Viscosity of Carbon Dioxide (η/μPa s)—Continued

T/K (P/MPa)	400	420	440	460	480	500	520	540	560	580
0.1	19.70	20.59	21.47	22.33	23.18	24.02	24.84	25.65	26.44	27.23
0.5	19.72	20.61	21.49	22.35	23.20	24.04	24.86	25.67	26.46	27.24
1.0	19.76	20.65	21.52	22.38	23.23	24.06	24.88	25.69	26.49	27.27
2.5	19.92	20.79	21.66	22.51	23.35	24.17	24.99	25.79	26.58	27.36
5.0	20.37	21.19	22.02	22.84	23.65	24.45	25.25	26.03	26.80	27.57
7.5	21.11	21.83	22.57	23.33	24.09	24.86	25.62	26.37	27.12	27.86
10.0	22.23	22.75	23.36	24.01	24.69	25.39	26.10	26.81	27.53	28.24
12.5	23.81	24.01	24.40	24.90	25.46	26.07	26.71	27.36	28.03	28.70
15.0	25.94	25.63	25.71	25.99	26.41	26.90	27.44	28.02	28.62	29.24
17.5	28.61	27.63	27.30	27.31	27.52	27.86	28.29	28.78	29.31	29.86
20.0	31.71	29.97	29.14	28.82	28.80	28.97	29.26	29.64	30.08	30.56
22.5	35.07	32.55	31.20	30.51	30.23	30.19	30.34	30.59	30.94	31.34
25.0	38.50	35.29	33.41	32.34	31.78	31.53	31.50	31.63	31.86	32.18
27.5	41.86	38.08	35.72	34.28	33.42	32.96	32.75	32.74	32.86	33.08
30.0	45.08	40.85	38.07	36.28	35.14	34.45	34.07	33.91	33.91	34.03
35.0	51.02	46.16	42.73	40.33	38.69	37.57	36.84	36.39	36.14	36.06
40.0	56.34	51.08	47.19	44.33	42.26	40.77	39.71	38.98	38.50	38.21
45.0	61.19	55.63	51.39	48.18	45.76	43.95	42.61	41.62	40.92	40.43
50.0	65.67	59.85	55.34	51.84	49.14	47.06	45.47	44.26	43.35	42.68
55.0	69.86	63.82	59.06	55.33	52.39	50.08	48.27	46.86	45.77	44.93
60.0	73.83	67.57	62.60	58.65	55.51	53.00	51.00	49.42	48.16	47.17
65.0	77.65	71.16	65.99	61.84	58.51	55.82	53.66	51.91	50.50	49.38
70.0	81.33	74.61	69.24	64.91	61.40	58.55	56.23	54.34	52.80	51.54
75.0	84.87	77.95	72.38	67.87	64.20	61.20	58.74	56.71	55.04	53.67
80.0	88.36	81.19	75.43	70.75	66.92	63.77	61.17	59.02	57.24	55.76
85.0	91.66	84.36	78.41	73.56	69.57	66.28	63.55	61.28	59.38	57.80
90.0	95.04	87.46	81.32	76.30	72.16	68.73	65.88	63.49	61.49	59.81
95.0	98.35	90.52	84.18	78.99	74.70	71.13	68.15	65.65	63.55	61.77
100.0	101.41	93.52	86.99	81.63	77.19	73.49	70.39	67.78	65.58	63.71
125.0	117.43	108.09	100.58	94.36	89.16	84.79	81.10	77.95	75.27	72.96
150.0	132.44	122.21	113.73	106.64	100.67	95.62	91.32	87.65	84.48	81.75
175.0	147.49	136.16	126.69	118.73	111.98	106.24	101.33	97.11	93.45	90.28
200.0	162.50	150.07	139.62	130.77	123.25	116.81	111.27	106.48	102.32	98.70
225.0	177.51	163.99	152.57	142.85	134.54	127.39	121.22	115.86	111.19	107.10
250.0	192.58	177.99	165.59	155.00	145.90	138.04	131.23	125.29	120.10	115.54
275.0	207.72	192.06	178.71	167.25	157.36	148.79	141.33	134.81	129.09	124.05
300.0	222.94	206.24	191.93	179.61	168.94	159.66	151.55	144.44	138.18	132.65

Tabulations of the Viscosity of Carbon Dioxide (η/μPa s)—Continued

T/K (P/MPa)	600	620	640	660	680
0.1	28.00	28.76	29.50	30.24	30.96
0.5	28.01	28.77	29.52	30.25	30.98
1.0	28.04	28.79	29.54	30.27	31.00
2.5	28.12	28.87	29.62	30.35	31.07
5.0	28.32	29.06	29.79	30.52	31.23
7.5	28.59	29.32	30.04	30.74	31.44
10.0	28.94	29.65	30.34	31.03	31.71
12.5	29.37	30.04	30.71	31.38	32.04
15.0	29.87	30.50	31.14	31.78	32.41
17.5	30.44	31.03	31.63	32.23	32.84
20.0	31.08	31.62	32.17	32.73	33.31
22.5	31.78	32.26	32.77	33.29	33.82
25.0	32.55	32.96	33.41	33.89	34.38
27.5	33.37	33.71	34.11	34.53	34.98
30.0	34.23	34.51	34.84	35.21	35.61
35.0	36.09	36.21	36.41	36.67	36.97
40.0	38.06	38.03	38.10	38.23	38.43
45.0	40.11	39.93	39.86	39.88	39.97
50.0	42.20	41.87	41.67	41.58	41.56
55.0	44.30	43.84	43.52	43.31	43.19
60.0	46.40	45.81	45.37	45.05	44.84
65.0	48.48	47.77	47.22	46.81	46.50
70.0	50.53	49.71	49.06	48.55	48.16
75.0	52.55	51.63	50.88	50.28	49.81
80.0	54.53	53.52	52.68	52.00	51.45
85.0	56.48	55.37	54.45	53.69	53.07
90.0	58.39	57.20	56.20	55.37	54.67
95.0	60.27	59.00	57.93	57.02	56.26
100.0	62.12	60.77	59.62	58.65	57.82
125.0	70.98	69.26	67.77	66.49	65.37
150.0	79.37	77.30	75.50	73.91	72.52
175.0	87.51	85.09	82.96	81.08	79.42
200.0	95.52	92.74	90.27	88.10	86.16
225.0	103.51	100.34	97.54	95.05	92.83
250.0	111.52	107.96	104.81	102.00	99.49
275.0	119.59	115.63	112.11	108.97	106.16
300.0	127.75	123.38	119.49	116.01	112.89

Tabulations of the Viscosity of Carbon Dioxide (η/μPa s)—Continued

T/K (P/MPa)	700	800	900	1000	1100	1200	1300	1400	1500
0.1	31.68	35.09	38.27	41.26	44.08	46.76	49.32	51.77	54.13
0.5	31.69	35.10	38.28	41.27	44.09	46.77	49.33	51.78	54.14
1.0	31.71	35.12	38.30	41.28	44.10	46.78	49.34	51.79	54.15
2.5	31.78	35.17	38.35	41.33	44.14	46.81	49.37	51.81	54.17
5.0	31.93	35.30	38.45	41.42	44.22	46.88	49.43	51.87	54.23
7.5	32.13	35.46	38.58	41.53	44.32	46.97	49.50	51.94	54.29
10.0	32.39	35.66	38.75	41.66	44.43	47.07	49.59	52.02	54.36
12.5	32.69	35.89	38.93	41.82	44.57	47.18	49.69	52.11	54.44
15.0	33.05	36.16	39.15	42.00	44.71	47.31	49.81	52.21	54.53
17.5	33.44	36.46	39.39	42.19	44.88	47.46	49.93	52.32	54.63
20.0	33.89	36.79	39.65	42.41	45.06	47.61	50.07	52.44	54.73
22.5	34.37	37.15	39.93	42.64	45.26	47.78	50.21	52.57	54.85
25.0	34.89	37.54	40.24	42.89	45.47	47.96	50.37	52.70	54.97
27.5	35.45	37.96	40.57	43.16	45.69	48.15	50.54	52.85	55.10
30.0	36.04	38.40	40.91	43.44	45.93	48.35	50.71	53.00	55.24
35.0	37.31	39.35	41.66	44.05					
40.0	38.67	40.37	42.46	44.71					
45.0	40.12	41.46	43.33	45.41					
50.0	41.62	42.60	44.23	46.16					
55.0	43.15	43.78	45.18	46.93					
60.0	44.72	44.99	46.15	47.74					
65.0	46.29	46.23	47.15	48.57					
70.0	47.87	47.48	48.18	49.43					
75.0	49.44	48.74	49.21	50.30					
80.0	51.00	50.01	50.26	51.18					
85.0	52.56	51.27	51.31	52.07					
90.0	54.10	52.54	52.37	52.97					
95.0	55.62	53.80	53.43	53.88					
100.0	57.12	55.05	54.49	54.79					
125.0	64.39	61.18	59.75	59.35					
150.0	71.30	67.06	64.85	63.85					
175.0	77.95	72.72	69.80	68.24					
200.0	84.45	78.23	74.61	72.52					
225.0	90.86	83.63	79.31	76.70					
250.0	97.24	88.96	83.93	80.79					
275.0	103.64	94.28	88.50	84.83					
300.0	110.08	99.60	93.06	88.84					

11. Appendix II. The Viscosity of Carbon Dioxide Near the Critical Point (η/μPa s)

T/K (P/MPa)	298	300	302	304	306	308	310
0.1	14.92	15.02	15.12	15.21	15.31	15.41	15.50
0.5	14.96	15.06	15.15	15.25	15.35	15.44	15.54
1.0	15.02	15.11	15.21	15.31	15.40	15.50	15.59
1.5	15.09	15.19	15.28	15.38	15.47	15.57	15.66
2.0	15.19	15.29	15.38	15.47	15.57	15.66	15.75
2.5	15.32	15.41	15.50	15.59	15.68	15.77	15.86
3.0	15.47	15.56	15.65	15.73	15.82	15.91	16.00
3.5	15.67	15.75	15.83	15.92	16.00	16.08	16.16
4.0	15.93	16.00	16.07	16.14	16.22	16.29	16.37
4.5	16.26	16.31	16.37	16.43	16.49	16.56	16.62
5.0	16.70	16.72	16.76	16.80	16.84	16.89	16.94
5.5	17.33	17.30	17.28	17.28	17.29	17.31	17.34
6.0	18.37	18.17	18.04	17.96	17.91	17.87	17.86
6.5	58.36	19.83	19.30	19.00	18.79	18.66	18.56
7.0	62.08	56.71	22.97	20.99	20.27	19.85	19.57
7.5	64.85	60.47	55.26	47.33	24.06	22.07	21.21
8.0	67.16	63.27	59.01	54.00	47.07	31.53	24.78
8.5	69.17	65.60	61.81	57.68	52.92	46.81	37.43
9.0	70.99	67.63	64.15	60.47	56.49	51.99	46.57
9.5	72.66	69.46	66.18	62.80	59.24	55.42	51.20
10.0	74.21	71.13	68.01	64.83	61.54	58.11	54.47
10.5	75.67	72.68	69.69	66.65	63.56	60.39	57.09
11.0	77.05	74.15	71.24	68.33	65.38	62.39	59.32
11.5	78.36	75.53	72.70	69.88	67.04	64.19	61.30
12.0	79.62	76.84	74.08	71.34	68.59	65.84	63.08
12.5	80.83	78.09	75.39	72.71	70.04	67.38	64.71
13.0	81.99	79.30	76.65	74.02	71.40	68.82	66.24
13.5	83.10	80.45	77.85	75.27	72.71	70.18	67.67
14.0	84.21	81.59	79.00	76.47	73.95	71.47	69.02

12. Appendix III. The Viscosity of Carbon Dioxide along the Saturation Line

T/K	η_{gas}/μPa s	η_{liquid}/μPa s
205	10.33	
210	10.60	
215	10.87	
220	11.13	241.68
225	11.41	221.72
230	11.69	203.75
235	11.98	187.48
240	12.27	172.67
245	12.58	159.13
250	12.90	146.69
255	13.24	135.20
260	13.61	124.30
265	14.02	114.63
270	14.47	105.21
275	14.99	96.44
280	15.61	87.89
285	16.37	79.64
290	17.36	71.47
295	18.79	63.01
300	21.29	53.33
302	23.52	48.30

13. Appendix IV. Values of η Given at Specified T, P and ρ for Checking Computer Code

T/K	P/MPa	ρ/kg m^{-3}	η/μPa s
220	0.1	2.440	11.06
300	0.1	1.773	15.02
800	0.1	0.662	35.09
304	7	254.320	20.99
220	15	1194.86	269.37
300	50	1029.27	132.55
800	75	407.828	48.74