International Journal of Thermophysics, Vol. 12, No. 5, 1991

Accurate Transport Properties and Second Virial Coefficients for Helium Based on a State-of-the Art Interatomic Potential

M. J. Slaman[1] **and R. A. Aziz**[1]

Received December 7, 1990

Second virial coefficients and transport properties of helium are presented based on a state-of-the-art interatomic potential which was constructed with the use of a multiproperty fit. The experimental potential employed to produce these properties accurately reproduces a wide range of bulk and microscopic data and agrees well with *ab initio* calculations which were not available at the time of its construction. Virial coefficients of ^{3}He and ^{4}He are presented from 2 to 600 K, and transport properties of pure ^{3}He and ^{4}He gases and ^{3}He–^{4}He mixtures are presented from 5 to 6000 K.

KEY WORDS: diffusion; helium; thermal conductivity; thermal diffusion; transport properties; virials; viscosity.

1. INTRODUCTION

The behavior of an atomic system is implicitly described by its associated interatomic potential. Therefore, once an accurate potential has been determined, the various properties of the system can be calculated over a wide temperature range to a high degree of precision.

The first step in the construction of an accurate potential is the selection of a suitable functional form. The form should be flexible enough to reproduce all the reliable data but should not be unnecessarily complex. Furthermore, it should be realistic at short and long range and should lend itself to the inclusion of theoretical values if available. Potentials of the Lennard–Jones, exp-6, or m-6-8 [1] type are not suitable candidates because they do not satisfy all these requirements.

---

[1] Deparment of Physics, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada.

0195-928X/91/0900-0837$06.50/0

Once a suitable form has been chosen, the next stage in constructing an accurate potential is the use of a multiproperty fit to the most reliable theoretical and experimental results available. It is essential that a multiproperty fit be employed, because various properties probe the potential in different regions. If one attempts to determine the potential by fitting to only one or two properties, no unique solution can be found. However, the more properties used in the fitting procedure, the more accurately the potential is defined because each property probes and constrains the potential in a different region. What particular region of the potential is probed by each of the various properties has been described elsewhere [2, 3].

2. THE POTENTIAL

Using a multiproperty fit, Aziz et al. [4] recently presented a reliable potential for helium which appears to be a better characterization of that system than any potential previously found in the literature. Their new potential is fully consistent with both short- and long-range *ab initio* calculations, second virial coefficient data for ^{3}He and ^{4}He over an extended temperature range, and recent measurements of transport properties. It also gives a good account of both differential [5, 6] and integral [7, 8, 9] scattering cross-section measurements. Some of the sets of data reproduced by the potential are summarized in Tables I and II.

The potential form chosen by Aziz et al. [4] to represent the helium interaction was the HFD-B form, a modification [10] of the earlier HFD (Hartree–Fock dispersion) potential of Ahlrichs et al. [11]. The HFD-B potential, which has been highly successful in providing accurate potentials for other rare gas interactions [12–16], is given by

$$V(r) = \varepsilon V^*(x) \tag{1}$$

where

$$V^*(x) = A^* \exp(-\alpha^* x + \beta^* x^2) - F(x)[c_6/x^6 + c_8/x^8 + c_{10}/x^{10}] \tag{2}$$

with

$$\begin{aligned} F(x) &= \exp[-(D/x - 1)^2], \qquad && x < D \\ &= 1, && x \geqslant D \end{aligned} \tag{3}$$

and

$$x = r/r_m$$

The parameters for the new helium potential, the HFD-B(HE) [4], are listed in Table III.

Table I. Primary Second Virial Data Predicted by HFD-B(HE) Potential (Units: ml · mol^{-1})

	Temperature (K)	Error bars	Deviation from experiment
Matacotta et al [23] (^{3}He)	1.47	±0.5	+0.8
	1.50	±0.5	+0.7
	1.55	±0.5	+0.5
	1.73	±0.4	−0.0
	1.90	±0.4	−0.3
	2.20	±0.5	+0.5
	3.00	±0.5	−0.5
	4.30	±0.4	−0.4
	9.00	±0.3	−0.2
	13.80	±0.2	−0.1
	20.30	±0.2	−0.0
Matacotta et al. [23, 24] (^{3}He)	1.47	±0.5	−0.4
	1.50	±0.5	−0.4
	1.55	±0.5	−0.4
	1.73	±0.4	−0.4
	1.90	±0.4	−0.4
	2.20	±0.5	−0.2
	3.00	±0.5	−0.2
	4.30	±0.4	−0.3
	9.00	±0.3	−0.1
	13.80	±0.2	−0.0
	20.30	±0.2	−0.0
Berry [25] (^{4}He)	2.601	±1.0	−0.033
	2.748	±0.7	−0.376
	3.330	±0.7	−0.554
	4.220	±0.5	−0.281
	7.199	±0.7	−0.208
	13.804	±0.4	−0.218
	20.271	±0.2	−0.146
	27.018	±0.3	−0.113
Kemp et al. [26] (^{4}He)	27.097	±0.06	−0.08
	43.794	±0.11	−0.10
	54.358	±0.07	−0.05
	63.150	±0.09	+0.05
	82.804	±0.11	+0.04
	172.011	±0.14	+0.01
Gammon [27] (^{4}He)	98.15	±0.05	+0.06
	123.15	±0.05	−0.02
	148.15	±0.05	−0.06
	173.15	±0.05	−0.07
	198.15	±0.05	−0.08
	223.15	±0.05	−0.08
	248.15	±0.05	−0.07
	273.15	±0.05	−0.07
	298.15	±0.05	−0.06
	323.15	±0.05	−0.06
	348.15	±0.05	−0.05
	373.15	±0.05	−0.05
	398.15	±0.05	−0.04
	423.15	±0.05	−0.04
Kell et al. [28] (^{4}He)	298.15	±0.03	−0.03
	623.15	±0.01	−0.04

Table II. Primary Transport Data Predicted by HFD-B(HE) Potential

	Temperature range (K)	Error bars (%)	rms deviation[a]	Maximum ± deviation (%)
Viscosity ^{4}He (μ a · s)				
Vogel [32]	298–623	±0.3	0.034 (0.12)	−0.04 to −0.17
Vogel [33][b]	298–641	±0.3	0.015 (0.05)	−0.02 to −0.11
Clarke/Smith [34][c]	77–374	±1	0.083 (0.57)	−0.20 to −1.0
Clarke/Smith [34][c,d]	120–360	±1	0.093 (0.63)	−0.25 to −1.2
Guevara et al. [35][c]	1100–1800	±0.6	0.232 (0.38)	−0.53 to +0.59
Guevara et al. [35][c]	1100–2150	±0.6	1.14 (1.50)	−0.53 to +3.7
Kestin et al. [46]	300.65	±0.3	0.022 (0.11)	−0.11
Thermal conductivity ^{4}He ($\mathrm{mW \cdot m^{-1} \cdot K^{-1}}$)				
Assael et al. [47]	308.15	±0.2	0.21 (0.13)	+0.13
Kestin et al. [46]	300.65	±0.3	0.11 (0.07)	+0.07
Acton/Kellner [48]	4–20	±1.1	0.11 (0.73)	−1.2 to +0.34
Acton/Kelner [48][d]	4–20	±1.1	0.10 (0.68)	−1.0 to +0.37
Haarman [49]	328–468	±0.3	0.64 (0.33)	+0.21 to +0.42
Jody et al. [50]	400–2500	±4.7	10.8 (2.25)	−0.05 to +4.8

[a] Values in parentheses refer to rms percentage deviations.

[b] Data obtained using edge-correction *C*.

[c] These capillary flow viscosity results have been renormalized using the accurate value (error ±0.1%) near room temperature of Vogel [32].

[d] Smoothed.

Table III. Parameters for HFD-B(HE) Potential[a]

A^*	1.8443101×10^5
α^*	10.43329537
c_6	1.36745214
c_8	0.42123807
c_{10}	0.17473318
C_6 (au)	1.461
C_8 (au)	14.11
C_{10} (au)	183.5
β^*	−2.27965105
β ($\mathrm{\AA^{-2}}$)	−0.259660
D	1.4826
ε/k (K)	10.948
r_m (Å)	2.963
σ (Å)	2.636_9

[a] The values for C_6, C_8, C_{10}, β, ε/k, r_m, and D are used to define the potential and are not truncated. Not all figures displayed are significant. Some are displayed only to avoid round-off errors.

3. THE MULTIPROPERTY FIT

While a detailed explanation of the fitting procedure used and a critical evaluation of the data applied in determining the new potential for helium have already been described elsewhere [4], a brief summary of those considerations is in order here.

The repulsive wall of the potential was determined by fixing the interatomic potential energy at a separation of one Bohr to the value obtained by Ceperley and Partridge [17], who used quantum Monte Carlo techniques for determining the Born–Oppenheimer interaction energy, and to the room temperature viscosity value of Vogel at 300 K. This viscosity measurement probes the wall of the potential chiefly at a separation of about 4 Bohr. The potential is in excellent agreement with the values of Ceperley and Partridge over the whole range of their calculations, which extends from 1.0 to 3.0 Bohr (see Table IV). Also, it predicts the recent *ab initio* calculations of Liu and McLean [18] obtained by their interacting correlated fragment (ICF) model to within 0.6% at the separations 3.5 and 4.0 Bohr. These results were not available at the time the HFD-B(HE) was presented. While the potential does not fit within their very tight error bars at all separations, it still gives very good agreement with their values (see Table V and Ref. 18). It should be noted that the new *ab initio* potential of Liu and McLean (called the LM-2) does not agree with the results of Ceperley and Partridge to within their mutual error bars at 3.0 Bohr. Nevertheless, this is due primarily to very small error bars on both sets of data, rather than any significant difference between the results at this separation. (The Ceperley and Partridge value is less than 1% higher than

Table IV. Percentage Deviations of HFD-E(HE) Potential from the Theoretical Values of Ceperley and Partridge [17]

r (au)	$V(r)$ [17] (K)	± error (%)	Deviation of HFD-B(HE) from $V(r)$ of Ref. 17 (%)
1.00	2.919×10^5	0.41	−0.20
1.25	1.746×10^5	0.69	+0.52
1.50	1.050×10^5	0.76	−0.21
1.75	6.140×10^4	0.98	+0.96
2.00	3.580×10^4	0.84	+1.50
2.25	2.095×10^4	0.96	+0.68
2.50	1.192×10^4	1.85	+1.54
2.8346	5.540×10^3	2.89	+1.68
3.00	3.800×10^3	0.53	+0.49

Table V. Percentage Deviations of HFD-B(HE) Potential from the *ab Initio* Values of Liu and McLean [18]

r (au)	$V(r)$ (LM-2 of Ref. 18) (K)	± error (%)	Deviation of HFD-B(HE) from LM-2 (%)
3.0	3767.	0.29	+1.37
3.5	1112.	0.27	+0.60
4.0	293.1	0.20	−0.59
4.6	39.13	0.49	−3.24
5.0	−0.33	n/a	n/a
5.6	−10.94	0.27	−0.07
6.0	−9.64	0.21	+0.45
6.5	−6.868	0.19	+0.34
7.0	−4.601	0.13	−0.18
7.5	−3.057	0.16	−0.61
8.0	−2.062	0.24	−0.39
8.5	−1.412	0.21	−0.20
9.0	−0.985	0.30	−0.30
10.0	−0.512	0.59	+0.06
12.0	−0.164	1.83	−1.10

the LM-2 of Liu and McLean.) Needless to say, some of these error estimates must be somewhat too optimistic. When one looks at the *ab initio* results of van Duijneveldt and co-workers [19, 20], the potential agrees well with their somewhat larger percentage errors (Table VI). In view of all this, the upper repulsive wall of the HFD-B(HE) appears to be well defined.

Table VI. Percentage Deviations of HFD-B(HE) Potential from the *ab Initio* Values of van Duijneveldt and Co-Workers [19, 20]

r (au)	$V(r)$ [19, 20] (K)	± error (%)[a]	Deviation of HFD-B(HE) from $V(r)$ of Refs. 19 and 20 (%)
3.00	3795.	1	+0.62
4.00	295.12	1	−1.27
5.00	−0.001	1	n/a
5.60	−10.919	1	−0.27
6.00	−9.610	1	+0.14
7.00	−4.610	1	+0.02
8.00	−2.06	1	−0.49

[a] The error bars of ±1% apply to the dispersion energy component of the theoretical $V(r)$ only. The total uncertainty would be somewhat higher.

The long-range tail of the potential is fixed by the use of new *ab initio* calculations [21, 22] of the dispersion coefficients, C_6, C_8, and C_{10}. Also, this region is sensitive to low-temperature second virial coefficients. The HFD-B(HE) potential reproduces the ^{3}He virials of Matacotta *et al.* [23, 24] over their range of measurement from 1.47 to 20.3 K and the ^{4}He virials of various researchers [25–30] (some of which have been smoothed [31]) over an extensive temperature range from 2.6 to 423 K. Additionally, the HFD-B(HE) is within the error bars of the ICF results [18] for the long-range part of the calculation (8.5 to 12.0 Bohr). All in all, it appears that the long-range part of the potential is well characterized.

When the HFD-B(HE) was constructed, no reliable *ab initio* results were available for the portion of the potential intermediate between the short- and the long- range sections. The potential reproduces the entire range (298 to 641 K) of Vogel's data [32, 33] as well as the intermediate-temperature (77 to 300 K) data of Clarke and Smith [34] and the high-temperature results of Guevara et al. [35] for the range from 1100 to 1800 K.

Integral cross-section data have been measured for crossed beams of ^{3}He–^{3}He and ^{4}He–^{4}He by both aufm Kampe et al. [8] and Feltgen et al. [7] and for ^{3}He–^{4}He crossed beams by the latter group. These data sense the potential in the region from 3.46 to 4.00 Bohr. The HFD-B(HE) gives an excellent account of these data [4]. Depending on the relative energy of the crossed beams, the potential from the low repulsive wall to the inflection point in the outer attractive wall can be probed by differential collision cross-section data (dccs). Measurements of dccs have been reported for crossed beams of ^{3}He–^{4}He [5] and ^{4}He–^{4}He [6]. Results were obtained at both low (58 K) and high (732 K) relative energies, and the agreement with the HFD-B(HE) potential was seen to be very good [4].

As mentioned above, no reliable *ab initio* potentials were available for the intermediate part of the interaction when the HFD-B(HE) experimental helium potential was constructed. However, it would be interesting to compare this potential with the most recent *ab initio* results [18]. When we do so we find very good agreement for the well depth (ε/k) as well as for the separations at which the zero crossing point (σ) and potential minimum (r_m) occur. These parameters are listed for both the LM-2 and the HFD-B(H(E) potentials in Table VII.

In summary then, the interaction is well characterized by the HFD-B(HE) potential for both the short and the long range, as well as at intermediate separations.

Table VII. Parameters of Recent *ab Initio* and Experimental Potentials

Parameter	LM-2 [18]	HFD-B(HE) [4]
ε/k (K)	10.94 ± 0.03	10.948
σ (Å)	2.642	2.637
r_m (Å)	2.969	2.963

4. RECOMMENDED VALUES

Since the HFD-B(HE) potential was constructed by a fit to the most reliable experimental and theoretical results (many of which were not available when earlier potentials were presented [4]) and since it agrees well with other reliable primary data (see Tables I and II and Ref. 4) and with subsequent *ab initio* calculations [18–20], we can be confident that the new potential is a highly accurate characterization of the helium interaction. In view of this, we present recommended second virial coefficients (for ^{3}He and ^{4}He) and transport properties (for ^{3}He, ^{3}He–^{4}He, and ^{4}He) based on this potential in Tables VIII, IX, and X. In these tables, the virials for both ^{3}He and ^{4}He have been calculated using the first two quantum corrections [36] for temperatures above 700 K and by using a full quantum procedure below 700 K. Chapman-Cowling expressions [36] have been used to evaluate viscosity coefficients, thermal conductivity coefficients, and thermal diffusion factors in third order as well as diffusion coefficients in second order.

For purposes of interpolation between the tabulated values, we have generated empirical functions for these five properties. These functions are given by the following expressions, in which T represents the temperature in degrees Kelvin.

Second virials:

$$B(T) = T^{-0.25}(A + BT^{-0.5} + CT^{-1} + DT^{-1.5} + ET^{-2}), \qquad T < 700\text{ K} \tag{4}$$

$$B(T) = G(\ln T), \qquad T \geqslant 700\text{ K} \tag{5}$$

Viscosity: $$\eta(T) = \exp[G(\ln T)] \tag{6}$$

Thermal conductivity: $$\lambda(T) = \exp[G(\ln T)] \tag{7}$$

Diffusion: $$D(T) = \exp[G(\ln T)] \tag{8}$$

Thermal diffusion: $$\alpha_T(T) = \exp[G(\ln T)] \tag{9}$$

Table VIII. Properties of ^{3}He as a Function of Temperature[a]

T (K or °C)	B ($\mathrm{ml \cdot mol^{-1}}$)	η ($\mu\mathrm{Pa \cdot s}$)	λ ($\mathrm{mW \cdot m^{-1} \cdot K^{-1}}$)	D (1.013 bar) ($10^{-4}\,\mathrm{m^2 \cdot s^{-1}}$)	α_T
1.2 K	−206.021	0.660	6.825	2.283×10^{-4}	0.08560
2	−131.163	0.985	10.252	5.150×10^{-4}	0.12078
5	−47.491	1.356	14.228	2.284×10^{-3}	0.10144
50	9.576	5.322	55.271	9.831×10^{-2}	0.08338
100	11.985	8.326	86.456	3.103×10^{-1}	0.07898
150	12.335	10.866	112.802	6.112×10^{-1}	0.07596
200	12.273	13.158	136.568	9.916×10^{-1}	0.07366
250	12.093	15.287	158.650	1.446	0.07181
300	11.877	17.300	179.511	1.969	0.07026
0°C	11.995	16.243	168.555	1.681	0.07106
20	11.907	17.030	176.721	1.894	0.07046
40	11.819	17.813	184.832	2.118	0.06990
60	11.730	18.582	192.806	2.353	0.06936
80	11.641	19.339	200.653	2.599	0.06883
100	11.554	20.084	208.376	2.855	0.06835
150	11.341	21.903	227.223	3.539	0.06721
200	11.139	23.665	245.490	4.284	0.06620
250	10.948	25.381	263.266	5.089	0.06526
300	10.767	27.055	280.611	5.954	0.06440
350	10.597	28.692	297.578	6.876	0.06360
400	10.436	30.297	314.207	7.854	0.06285
450	10.286	31.874	330.535	8.889	0.06214
500	10.141	33.424	346.596	9.979	0.06149
600	9.873	36.455	377.993	12.321	0.06023
700	9.630	39.405	408.557	14.876	0.05913
800	9.409	42.287	438.406	17.641	0.05808
900	9.205	45.100	467.536	20.587	0.05693
1000	9.017	47.871	496.235	23.758	0.05603
1200	8.681	53.279	552.230	30.695	0.05442
1400	8.386	58.534	606.639	38.411	0.05297
1600	8.126	63.666	659.765	46.894	0.05164
1800	7.892	68.694	711.817	56.130	0.05042
2000	7.681	73.628	762.890	66.109	0.04926
2200	7.489	78.485	813.157	76.826	0.04820
2400	7.312	83.275	862.730	88.269	0.04721
2600	7.150	88.001	911.629	100.433	0.04626
2800	6.999	92.675	959.996	113.313	0.04534
3000	6.859	97.297	1007.827	126.905	0.04449
3500	6.546	108.670	1125.493	163.977	0.04252
4000	6.277	119.821	1240.852	205.411	0.04073
4500	6.041	130.795	1354.365	251.179	0.03908
5000	5.832	141.619	1466.323	301.240	0.03755
5500	5.645	152.317	1576.962	355.580	0.03613

[a] In this table α_T has been calculated in third-order Chapman-Cowling approximation, assuming a mixture of ^{3}He–^{4}He containing 99.999% ^{3}He.

Table IX. Properties of ^{4}He as a Function of Temperature[a]

T (K or °C)	B (ml·mol^{-1})	η (μPa·s)	λ (mW·m^{-1}·K^{-1})	D (1.013 bar) (10^{-4} m^2·s^{-1})	α_T
1.2 K	−370.968	0.343	2.730	1.093×10^{-4}	0.05124
2	−194.723	0.458	3.581	3.089×10^{-4}	0.06831
5	−64.430	1.224	9.541	1.717×10^{-3}	0.07710
50	8.690	6.099	47.721	8.448×10^{-2}	0.07936
100	11.624	9.576	74.921	2.681×10^{-1}	0.07535
150	12.122	12.505	97.813	5.292×10^{-1}	0.07264
200	12.127	15.145	118.450	8.592×10^{-1}	0.07056
250	11.983	17.599	137.621	1.253	0.06887
300	11.790	19.919	155.741	1.708	0.06742
0°C	11.897	18.701	146.224	1.457	0.06817
20	11.818	19.609	153.317	1.642	0.06761
40	11.737	20.510	160.359	1.837	0.06708
60	11.654	21.396	167.280	2.041	0.06658
80	11.571	22.268	174.092	2.254	0.06609
100	11.489	23.127	180.803	2.476	0.06565
150	11.286	25.223	197.166	3.069	0.06458
200	11.091	27.254	213.031	3.716	0.06363
250	10.905	29.231	228.468	4.415	0.06276
300	10.729	31.161	243.539	5.165	0.06194
350	10.563	33.049	258.278	5.966	0.06119
400	10.404	34.901	272.739	6.815	0.06046
450	10.256	36.717	286.919	7.713	0.05981
500	10.114	38.503	300.856	8.659	0.05919
600	9.850	41.994	328.108	10.692	0.05801
700	9.611	45.393	354.636	12.910	0.05697
800	9.391	48.714	380.557	15.310	0.05599
900	9.190	51.955	405.846	17.871	0.05490
1000	9.003	55.148	430.758	20.623	0.05406
1200	8.669	61.378	479.365	26.645	0.05255
1400	8.376	67.431	526.595	33.343	0.05118
1600	8.117	73.343	572.711	40.707	0.04992
1800	7.885	79.136	617.896	48.724	0.04877
2000	7.674	84.820	662.229	57.386	0.04767
2200	7.483	90.415	705.864	66.689	0.04666
2400	7.307	95.933	748.896	76.623	0.04573
2600	7.145	101.377	791.343	87.182	0.04483
2800	6.995	106.761	833.328	98.362	0.04395
3000	6.855	112.087	874.848	110.161	0.04314
3500	6.543	125.188	976.988	142.341	0.04127
4000	6.274	138.034	1077.126	178.308	0.03956
4500	6.039	150.676	1175.661	218.037	0.03798
5000	5.830	163.145	1272.847	261.492	0.03652
5500	5.643	175.469	1368.888	308.663	0.03516

[a] In this table α_T has been calculated in third-order Chapman-Cowling approximation, assuming a mixture of ^{3}He–^{4}He containing 99.999% ^{4}He.

Table X. Properties of an Equimolar Binary Mixture of ^{3}He and ^{4}He as a Function of Temperature

T (K or °C)	η (μPa · s)	λ (mW · m^{-1} · K^{-1})	D (1.013 bar) (10^{-4} m^2 · s^{-1})	α_T
1.2 K	0.468	4.247	1.617×10^{-4}	0.06429
2	0.656	5.989	4.088×10^{-4}	0.08748
5	1.276	11.476	2.010×10^{-3}	0.08727
50	5.751	51.119	9.166×10^{-2}	0.08118
100	9.011	80.103	2.900×10^{-1}	0.07700
150	11.762	104.578	5.718×10^{-1}	0.07415
200	14.244	126.655	9.280×10^{-1}	0.07197
250	16.549	147.165	1.353	0.07021
300	18.726	166.540	1.844	0.06871
0°C	17.570	156.260	1.572	0.06948
20	18.434	163.941	1.773	0.06891
40	19.281	171.479	1.983	0.06836
60	20.113	178.884	2.203	0.06784
80	20.932	186.173	2.433	0.06734
100	21.738	193.351	2.673	0.06688
150	23.705	210.855	3.313	0.06577
200	25.611	227.826	4.011	0.06480
250	27.466	244.339	4.766	0.06390
300	29.277	260.456	5.575	0.06306
350	31.048	276.219	6.439	0.06228
400	32.784	291.679	7.355	0.06155
450	34.489	306.854	8.325	0.06087
500	36.164	321.771	9.345	0.06024
600	39.441	350.944	11.539	0.05902
700	42.630	379.341	13.932	0.05795
800	45.746	407.090	16.521	0.05694
900	48.786	434.160	19.282	0.05583
1000	51.781	460.836	22.251	0.05496
1200	57.626	512.890	28.749	0.05340
1400	63.305	563.474	35.974	0.05200
1600	68.850	612.870	43.919	0.05070
1800	74.283	661.273	52.568	0.04952
2000	79.614	708.766	61.914	0.04840
2200	84.862	755.516	71.950	0.04736
2400	90.036	801.623	82.666	0.04641
2600	95.141	847.107	94.058	0.04548
2800	100.190	892.096	106.119	0.04458
3000	105.184	936.592	118.848	0.04376
3500	117.468	1046.064	153.563	0.04184
4000	129.512	1153.403	192.364	0.04009
4500	141.364	1259.032	235.222	0.03849
5000	153.053	1363.226	282.101	0.03699
5500	164.606	1466.205	332.986	0.03560

where

$$G(\ln T) = A + B\ln T + C(\ln T)^2 + D(\ln T)^3 + E(\ln T)^4 \qquad (10)$$

Coefficients for these functions are given in Table XI to XV. Also presented in these tables are the standard deviations (or standard perceptage deviations) of the empirical values from the values calculated directly from the potential as described above.

The form of the interpolating function for low-temperature second virials was taken from Kemp et al. [26] and was found by Aziz and Janzen [37] to provide a good fit to the helium virials. However, the previous work [37], which dealt only with virial coefficients, covered a shorter temperature range than that which is presented here. Also, the present function

Table XI. Coefficients of the Interpolating Functions for $B(T)$[a]

	1.2–60 K	60–700 K	700–6000 K
		^{3}He	
A	0.68805656×10^2	0.52337031×10^2	-0.52417146×10^2
B	-0.31043767×10^3	0.24706852×10^3	0.35838558×10^2
C	0.49158072×10^2	-0.74918481×10^4	-0.69152085×10^1
D	-0.18553409×10^3	0.47144623×10^5	0.54855342
E	0.14299747×10^3	-0.11381361×10^6	$-0.15993132 \times 10^{-1}$
N_p	97	42	121
SD	0.039	0.0035	0.0014
Max. dev.	0.166	0.0065	0.0038
		^{4}He	
A	0.64688468×10^2	0.52305677×10^2	-0.56274964×10^2
B	-0.24926729×10^3	0.24862057×10^3	0.37537482×10^2
C	-0.38225353×10^3	-0.76162986×10^4	-0.71967894×10^1
D	0.47113857×10^3	0.47118007×10^5	0.56932205
E	-0.38200454×10^3	-0.11376339×10^6	$-0.16567007 \times 10^{-1}$
N_p	97	42	131
SD	0.0344	0.0036	0.0014
Max. dev.	0.0777	0.0070	0.0036

[a] For each temperature range we have listed the number of points, N_p, used to generate the interpolating function from a least-squares fit to the values calculated directed from the potential. The standard deviation and maximum deviation of the N_p empirical values from the values calculated directly from the potential have also been listed. $B(T)$ in units of ml · mol^{-1}. Note that above 700 K the interpolating function for this property differs from the one used below 700 K [see Eqs. (4) and (5) in text].

is based on a slightly improved quantum-mechanical evaluation of virial coefficients which resulted from an extensive analysis of the procedure used to calculate them [38]. The modified procedure (which gives excellent agreement with the earlier work of Boyd et al. [39] for the Lennard–Jones potential) removes a slight discrepancy which existed at very low temperatures (below 7 K) between different computational procedures [4].

Table XII. Coefficients of the Interpolating Functions for $\eta(T)$[a]

	1.2–5 K	5–100 K	100–6000 K
		^{3}He	
A	0.16989916	0.25186458	0.18814962
B	0.10577423	-0.50977537×10^{-1}	0.40347603×10^{-1}
C	-0.13011182×10^{-1}	0.47516510×10^{-1}	0.47932545×10^{-2}
D	-0.35119561×10^{-1}	-0.87415490×10^{-2}	-0.44217252×10^{-3}
E	0.15023438×10^{-1}	0.60291939×10^{-3}	0.21854344×10^{-4}
N_p	24	20	60
SD (%)	0.1156	0.1594	0.0100
Max. dev. (%)	0.2696	0.5009	0.0549
		^{4}He	
A	0.12726030	0.56505992×10^{-1}	0.19899303
B	-0.44423427×10^{-1}	0.17019719	0.41787638×10^{-1}
C	0.15133155	-0.40933417×10^{-1}	0.45643745×10^{-2}
D	-0.52138162×10^{-1}	0.70066275×10^{-2}	-0.42659253×10^{-3}
E	0.29716142×10^{-2}	-0.44542143×10^{-3}	0.21477877×10^{-4}
N_p	24	20	59
SD (%)	0.2911	0.0922	0.0076
Max. dev. (%)	0.6422	0.2836	0.0426
		^{3}He–^{4}He[b]	
A	0.14548525	0.13449321	0.18983041
B	0.42558761×10^{-1}	0.82264220×10^{-1}	0.43753322×10^{-1}
C	0.39920636×10^{-1}	-0.58838126×10^{-2}	0.41191173×10^{-2}
D	-0.20620653×10^{-1}	0.76392873×10^{-3}	-0.38449324×10^{-3}
E	0.34592386×10^{-2}	-0.29031267×10^{-4}	0.20019806×10^{-4}
N_p	24	20	60
SD (%)	0.0450	0.0120	0.0056
Max. dev. (%)	0.1206	0.0234	0.0281

[a] For each temperature range we have listed the number of points, N_p, used to generate the interpolating function from a least-squares fit to the values calculated directly from the potential. The standard percentage deviation and maximum percentage deviation of the N_p empirical values from the values calculated directly from the potential have also been listed. $\eta(T)$ in units of μPa·s.

[b] Equimolar mixture.

Not only does the present compilation cover a more extended temperature region (up to 5500°C) than the Kestin et al. [40] correlation for helium, but also it avoids some of the shortcomings in that correlation. First, Kestin and co-workers [41] modified their two-parameter corresponding-states correlation [42] by introducing three additional parameters

Table XIII. Coefficients of the Interpolating Functions for $\lambda(T)$[a]

	1.2–6 K	6–100 K	100–6000 K
		^{3}He	
A	0.17250113×10^{1}	0.24510532×10^{1}	0.18950909×10^{1}
B	0.11066948×10^{1}	-0.39357003	0.41959206
C	-0.23791468	0.42790020	$0.44148461 \times 10^{-1}$
D	-0.22689270	$-0.78968312 \times 10^{-1}$	$-0.40495942 \times 10^{-2}$
E	0.10568403	$0.54676436 \times 10^{-2}$	$0.20496787 \times 10^{-3}$
N_p	23	22	96
SD (%)	0.1654	0.1942	0.0111
Max. dev. (%)	0.3328	0.5440	0.0716
		^{4}He	
A	0.10867908×10^{1}	0.45115868	0.17422523×10^{1}
B	-0.71523303	0.15503343×10^{1}	0.42099563
C	0.19645000×10^{1}	-0.34530174	$0.44716199 \times 10^{-1}$
D	-0.84676307	$0.58290023 \times 10^{-1}$	$-0.41680638 \times 10^{-2}$
E	0.11378514	$-0.36560317 \times 10^{-2}$	$0.21093366 \times 10^{-3}$
N_p	25	22	96
SD (%)	0.4953	0.1003	0.0090
Max. dev. (%)	1.2307	0.3021	0.0699
		^{3}He–^{4}He[b]	
A	0.13533989×10^{1}	0.13232822×10^{1}	0.17576245×10^{1}
B	0.46083494	0.72052654	0.45114778
C	0.37439598	$-0.18129049 \times 10^{-1}$	$0.38178334 \times 10^{-1}$
D	-0.21305900	$0.42591571 \times 10^{-3}$	$-0.35474270 \times 10^{-2}$
E	$0.39334308 \times 10^{-1}$	$0.18853572 \times 10^{-3}$	$0.18949471 \times 10^{-3}$
N_p	25	22	122
SD (%)	0.0866	0.0238	0.0062
Max. dev. (%)	0.2338	0.0623	0.0368

[a] For each temperature range we have listed the number of points, N_p, used to generate the interpolating function from a least-squares fit to the values calculated directly from the potential. The standard percentage deviation and maximum percentage deviation of the N_p empirical values from the values calculated directly from the potential have also been listed. $\lambda(T)$ in units of $\mathrm{mW \cdot m^{-1} \cdot K^{-1}}$.

[b] Equimolar mixture.

which would account for differences among the various gas partners at short and long range. For the short-range of helium, they relied on the Hartree–Fock calculations of McLaughlin and Schaefer [43], which were also incorporated in the earlier HFDHE2 potential of Aziz et al. [44]. This wall is much too repulsive as evidenced by the very accurate calculations

Table XIV. Coefficients of the Interpolating Functions for $D(T)^{a}$

	1.2–30 K	30–400 K	400–6000 K
		^{3}He	
A	$-0.86826950 \times 10^{1}$	$-0.86387347 \times 10^{1}$	$-0.70713776 \times 10^{1}$
B	0.15881680×10^{1}	0.15899322×10^{1}	0.73681475
C	$0.29604797 \times 10^{-1}$	$0.43788147 \times 10^{-2}$	0.17957873
D	$-0.96169609 \times 10^{-2}$	$0.47278754 \times 10^{-3}$	$-0.15682613 \times 10^{-1}$
E	$0.11748521 \times 10^{-2}$	$0.16636955 \times 10^{-4}$	$0.58321249 \times 10^{-3}$
N_p	32	30	81
SD (%)	0.1969	0.0039	0.0347
Max. dev. (%)	0.7388	0.0152	0.1055
		^{4}He	
A	$-0.95245487 \times 10^{1}$	$-0.91210832 \times 10^{1}$	$-0.75087938 \times 10^{1}$
B	0.22001340×10^{1}	0.18083026×10^{1}	0.88614331
C	-0.19037336	$-0.49512322 \times 10^{-1}$	0.15138520
D	$0.28448071 \times 10^{-1}$	$0.65129352 \times 10^{-2}$	$-0.13321992 \times 10^{-1}$
E	$-0.14611847 \times 10^{-2}$	$-0.24169036 \times 10^{-3}$	$0.50924578 \times 10^{-3}$
N_p	32	30	81
SD (%)	0.2143	0.0100	0.0268
Max. dev. (%)	0.7271	0.0424	0.0824
		^{3}He–^{4}He^{b}	
A	$-0.90773181 \times 10^{1}$	$-0.88562918 \times 10^{1}$	$-0.72748092 \times 10^{1}$
B	0.18991163×10^{1}	0.16874189×10^{1}	0.80644517
C	$-0.94370570 \times 10^{-1}$	$-0.19715870 \times 10^{-1}$	0.16645114
D	$0.14469563 \times 10^{-1}$	$0.31807290 \times 10^{-2}$	$-0.14586292 \times 10^{-1}$
E	$-0.69568366 \times 10^{-3}$	$-0.99644608 \times 10^{-4}$	$0.54894588 \times 10^{-3}$
N_p	32	30	81
SD (%)	0.1317	0.0056	0.0308
Max. dev. (%)	0.4484	0.0222	0.0940

[a] For each temperature range we have listed the number of points, N_p, used to generate the interpolating function from a least-squares fit to the values calculated directly from the potential. The standard percentage deviation and maximum percentage deviation of the N_p empirical values from the values calculated directly from the potential have also been listed. $D(T)$ in units of $10^{-4}\ \mathrm{m^2 \cdot s^{-1}}$ and at a pressure of 1.013 bar.

[b] Equimolar mixture.

of Ceperley and Partridge [17] and the high-energy beam results of Rol and co-workers [45]. At 3000°C, the difference between the Kestin et al. correlation and the present compilation is just under 2%. Second, for temperatures below 100 K, the older and outdated HFDHE2 potential was used [40] to calculate the low-temperature properties. The shallower well

Table XV. Coefficients of the Interpolating Functions for α_T[a]

	1.2–10 K	10–200 K	200–6000 K
		^{3}He[b]	
A	$-0.27028024 \times 10^{1}$	$-0.23115056 \times 10^{1}$	$-0.60513626 \times 10^{1}$
B	0.14625261×10^{1}	-0.16861962	0.23573005×10^{1}
C	-0.91928121	$0.89980951 \times 10^{-1}$	-0.57336413
D	$0.16551025 \times 10^{-1}$	$-0.20207417 \times 10^{-1}$	$0.60621419 \times 10^{-1}$
E	$0.56685104 \times 10^{-1}$	$0.13663435 \times 10^{-2}$	$-0.24986989 \times 10^{-2}$
N_p	29	27	129
SD (%)	0.7492	0.0872	0.0827
Max. dev. (%)	1.5856	0.1587	0.2013
		^{4}He[c]	
A	$-0.31523183 \times 10^{1}$	$-0.31303643 \times 10^{1}$	$-0.66563128 \times 10^{1}$
B	0.11555672×10^{1}	0.54493333	0.26757295×10^{1}
C	-0.88175287	-0.15870566	-0.64078007
D	0.30495979	$0.18218589 \times 10^{-1}$	$0.66930479 \times 10^{-1}$
E	$-0.38180781 \times 10^{-1}$	$-0.84072789 \times 10^{-3}$	$-0.27158746 \times 10^{-2}$
N_p	29	27	131
SD (%)	0.1754	0.0186	0.0892
Max. dev. (%)	0.5767	0.0313	0.1764
		^{3}He–^{4}He[d]	
A	$-0.29513403 \times 10^{1}$	$-0.27449807 \times 10^{1}$	$-0.62039288 \times 10^{1}$
B	0.12796252×10^{1}	0.21121600	0.24283340×10^{1}
C	-0.90518692	$-0.43150449 \times 10^{-1}$	-0.58809755
D	0.19492623	$0.45688958 \times 10^{-3}$	$0.61984391 \times 10^{-1}$
E	$-0.16205778 \times 10^{-2}$	$0.17519007 \times 10^{-3}$	$-0.25444801 \times 10^{-2}$
N_p	29	27	116
SD (%)	0.3536	0.0405	0.0852
Max. dev. (%)	0.6274	0.0810	0.2022

[a] For each temperature range we have listed the number of points, N_p, used to generate the interpolating function from a least-squares fit to the third-order values calculated directly from the potential. The standard percentage deviation and maximum percentage deviation of the N_p empirical values from the values calculated directly from the potential have also been listed. α_T is dimensionless.

[b] Calculated using a ^{3}He–^{4}He mixture containing 99.999% ^{3}He.

[c] Calculated using a ^{3}He–^{4}He mixture containing 99.999% ^{4}He.

[d] Equimolar mixture.

of that potential is principally responsible for predicting second virials that are 1.5 ml · mol^{-1} outside the experiment error limits for ^{4}He at 2.6 K and 2.7 ml · mol^{-1} for ^{3}He at 1.47 K. The transport properties at very low temperatures (~1.2 K) also differ by as much as 3.5%.

5. CONCLUSIONS

The potential reproduces a host of bulk and microscopic properties as well as *ab initio* calculations. These include second virials of ^{3}He and ^{4}He, viscosity and thermal conductivity of ^{4}He, dccs for ^{3}He–^{4}He and ^{4}He–^{4}He, and tccs for ^{3}He–^{3}He, ^{3}He–^{4}He, and ^{4}He–^{4}He, as well as the repulsive wall of Ceperley and Partridge. It also closely agrees with the new theoretical LM-2 potential at all separations including the potential well region. Furthermore, the long range has been properly fixed because it contains the accurate new dispersion coefficients. In essence, the potential acts as an all-encompassing correlation function, containing a large amount of information in compact form. Based on this potential, recommended second virials and transport properties have been presented both in tabular form and in terms of empirical functions for the purpose of interpolating between the tabulated values.

In conclusion, this correlation should supersede previous correlations for the properties of helium presented here.

ACKNOWLEDGMENTS

This research is supported in part by the Natural Sciences and Engineering Research Council of Canada. Thanks to Professor P. J. Dunlop for the use of his computer programs to calculate mixture viscosity and mixture thermal conductivity.

REFERENCES

1. M. Klein and H. J. M. Hanley, *J. Chem. Phys.* **53**:4722 (1970).
2. R. A. Aziz and M. J. Slaman, *Chem. Eng. Comm.* **78**:153 (1989).
3. J. H. Dymond and B. J. Alder, *J. Chem. Phys.* **51**:309 (1969).
4. R. A. Aziz, F. R. W. McCourt, and C. C. K. Wong, *Mol. Phys.* **61**:1487 (1987).
5. A. L. J. Burgmans, J. M. Farrar, and Y. T. Lee, *J. Chem. Phys.* **64**:1345 (1976).
6. J. M. Farrar and Y. T. Lee, *J. Chem. Phys.* **56**:5801 (1972).
7. R. Feltgen, H. Krist, K. A. Kohler, H. Pauly, and F. Torello, *J. Chem. Phys.* **76**:2360 (1982).
8. W. aufm Kampe, D. E. Oates, W. Schrader, and W. G. Bennewitz, *Chem. Phys. Lett.* **18**:323 (1973).
9. R. Feltgen, H. Pauly, F. Torello, and H. Vehmeyer, *Phys. Rev. Lett.* **30**:820 (1973).
10. R. A. Aziz and H. H. Chen, *J. Chem. Phys.* **67**:5719 (1977).

11. R. Ahlrichs, P. Penco, and G. Scoles, *Chem. Phys.* **19**:119 (1977).
12. R. A. Aziz and M. J. Slaman, *Mol. Phys.* **58**:679 (1986).
13. R. A. Aziz and M. J. Slaman, *Mol. Phys.* **57**:825 (1986).
14. R. A. Aziz and M. J. Slaman, *Chem. Phys.* **130**:187 (1989).
15. D. A. Barrow and R. A. Aziz, *J. Chem. Phys.* **89**:6189 (1988).
16. D. A. Barrow, M. J. Slaman, and R. A. Aziz, *J. Chem. Phys.* **91**:6348 (1989).
17. D. M. Ceperley and H. J. Partridge, *J. Chem. Phys.* **84**:820 (1986).
18. B. Liu and A. D. McLean, *J. Chem. Phys.* **91**:2348 (1989).
19. J. H. van Lenthe, R. J. Vos, J. G. C. M. van Duijneveldt-van de Rijdt, and F. B. van Duijneveldt, *Chem. Phys. Lett.* **143**:435 (1988).
20. F. B. van Duijneveldt, Private communication (1990).
21. A. Thakkar, *J. Chem. Phys.* **75**:4496 (1981).
22. A. Koide, W. J. Meath, and A. R. Allnatt, *J. Phys. Chem.* **86**:1222 (1982).
23. F. C. Matacotta, G. T. McConville, P. P. M. Steur, and M. Durieux, *Metrologia* **24**:61 (1987).
24. G. T. McConville, Private communication (1987).
25. K. H. Berry, *Metrologia* **15**:89 (1979).
26. R. C. Kemp, W. R. G. Kemp, and L. M. Besley, *Metrologia* **23**:61 (1986/1987).
27. B. H. Gammon, *J. Chem. Phys.* **64**:2556 (1976).
28. G. S. Kell, G. E. McLaurin, and E. Whalley, *J. Chem. Phys.* **68**:2199 (1978).
29. J. C. Holste, M. Q. Watson, M. T. Bellomy, P. T. Eubank, and R. K. Hall, *A.I.Ch.E. J.* **26**:954 (1980).
30. D. Gugan and W. D. Michel, *Metrologia* **16**:149 (1980).
31. P. P. M. Steur, M. Durieux, and G. T. McConville, *Metrologia* **24**:69 (1987).
32. E. Vogel, *Ber. Bunsenges. Phys. Chem.* **88**:997 (1984).
33. E. Vogel, *Z. Wilhelm-Pieck-Universitaet Rostock* **33**:34 (1984).
34. A. G. Clarke and E. B. Smith, *J. Chem. Phys.* **51**:4156 (1969).
35. F. A. Guevara, B. B. McInteer, and W. E. Wageman, *J. Chem. Phys.* **12**:2493 (1969).
36. J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1954).
37. R. A. Aziz and A. R. Janzen, *Metrologia* **25**:57 (1988).
38. R. A. Aziz and M. J. Slaman, *Metrologia* **27**:211 (1990).
39. M. E. Boyd, S. Y. Larsen, and J. E. Kilpatrick, *J. Chem. Phys.* **50**:4034 (1969).
40. J. Kestin, K. Knierim, E. A. Mason, B. Najafi, S. T. Ro, and M. Waldman, *J. Chem. Phys. Ref. Data* **13**:229 (1984).
41. B. Najafi, E. A. Mason, and J. Kestin, *Physica* **119A**:387 (1983).
42. J. Kestin, S. T. Ro, and W. A. Wakeham, *Physica* **58**:165 (1972).
43. D. R. McLaughlin and H. F. Schaefer III, *Chem. Phys. Lett* **12**:244 (1971).
44. R. A. Aziz, V. P. S. Nain, J. S. Carley, W. L. Taylor, and G. T. McConville, *J. Chem. Phys.* **70**:4330 (1979).
45. P. K. Rol, as cited in R. A. Aziz, *Inert. Gases, Chem. Phys. Ser. Vol. 34*, M. L. Klein, ed. (Springer, Berlin, 1984).
46. J. Kestin, R. Paul, A. A. Clifford, and W. A. Wakeham, *Physica* **100A**:349 (1980).
47. M. J. Assael, M. Dix, A. Lucas, and W. A. Wakeham, *J. Chem. Soc. Faraday. Trans. I* **77**:439 (1981).
48. A. Acton and K. Kellner, *Physica B* **90**:192 (1977).
49. J. W. Haarman, *A.I.P. Conf. Proc.* **11**:193 (1973).
50. B. J. Jody, S. C. Saxena, V. P. S. Nain, and R. A. Aziz, *Chem. Phys.* **22**:53 (1977).