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J. Kestin and E. Paykoc, Brown University;
J. V. Sengers, University of Maryland;
and
National Bureau of Standards, Washington, D.C.

September 1971

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FOREWORD

The research reported herein was sponsored by the Arnold Engineering Development Center (AEDC), Air Force Systems Command (AFSC). The portion of research pertaining to AEDC was conducted at the Institute for Molecular Physics of the University of Maryland under delivery order F40600-69-C-0002 and at the National Bureau of Standards under delivery order (40-600)66-494. The research was carried out during the period from September 1, 1968 to April 30, 1971. Air Force project monitor for this project was Capt Michael G. Buja, AEDC (DYR). The Program Element was 61102F and the Project 8951. The experimental phase of this project was carried out at the Division of Engineering of Brown University with funds provided by the National Science Foundation under grant GK 2133X. The computer calculations were performed on a UNIVAC 1108 computer at the Computer Science Center of the University of Maryland. The manuscript was submitted for publication May 15, 1971.

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ABSTRACT

The paper presents the results of new, precise measurements of the viscosity of nitrogen, argon, and helium at 25°C. The measurements were performed over a nominal range of pressures 1-100 atm and at very closely spaced density intervals. The data were subjected to a stringent statistical analysis in order to determine whether the density expansion consists of a pure polynomial or whether a term of the form $\rho^2 \ln\rho$ must be included in it. The existence of such a term was discovered theoretically by several investigators. The analysis indicates that if such a term exists, its factor must be very small.

This result is interpreted as indicating that correlations which extend over distances of the order of a mean free path are negligible when compared with correlations which extend over distances of the order of the range of molecular interactions.

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SECTION I

INTRODUCTION

The viscosity of a gas in the low-density limit can be described by the Chapman-Enskog theory which is based on Boltzmann's equation [1,2]. This theory is akin to the perfect-gas law in that it assumes that the molecules are randomly distributed in configuration space. In addition, the theory supposes that the velocities of two molecules which are about to collide are uncorrelated (assumption of molecular chaos).

A first attempt to extend the theory to higher densities was made by D. Enskog for the case of a gas of hard spheres [3]. In this theory an estimate was made to account for the effect of correlations in configuration space; these were assumed to be the same as those for a gas in equilibrium. The assumption of molecular chaos was retained for the probability distribution in velocity space. The theory implies the existence of a power series for the viscosity of the form

$$\eta = \eta_0 + \eta_1\rho + \eta_2\rho^2 + \eta_3\rho^3 + \ldots \qquad (1)$$

Such a series is analogous to the virial expansion for the equilibrium properties of a gas. The coefficients of the power series represent the effect of successively higher-order correlations in the position variables. The range of these correlations is closely related to the range of the molecular interaction.

A proper description of the density-dependence of the transport properties also requires the inclusion of an estimate of the deviations from molecular chaos in the velocity distribution. Such deviations are brought about by sequences of correlated collisions between the molecules [4,5]. In the past it was naturally assumed that these effects could also be represented by a power series in the density. However, the range of such velocity correlations is not restricted to the range of the molecular interaction, but extends to distances of the order of the mean free path. Since the mean free path is itself a function of the density, it is not evident that a power series which is formed with density-independent coefficients does exist. In fact, a systematic generalization of the Boltzmann equation indicates that the coefficient of the quadratic term contains a contribution which is proportional to the logarithm of density, so that we should write [4,6]

$$\eta = \eta_0 + \eta_1\rho + \eta_2'\rho^2\ln\rho + \eta_2\rho^2 + \ldots \qquad (2)$$

as a replacement for equation (1). Nevertheless, in the past experimental results on transport properties were habitually represented by a power series like that in equation (1). As a result of the theoretical developments alluded to before, the adequacy of a power series cannot be taken for granted and a critical re-examination of the experimentally established density dependence of these properties is called for.

H. J. M. Hanley, R. D. McCarty and J. V. Sengers [7] undertook an investigation to determine whether existing experimental data would favor the theoretically derived representation (2) in preference to the power series (1). A detailed account of this work was presented in AEDC-TR-69-68 [4]. Data on viscosity which were available at the time did not enable them to perform such a discrimination owing to inadequate precision. J. T. F. Kao and R. Kobayashi [8] and J. W. Gracki, G. P. Flynn and J. Ross [9] reached a similar conclusion. On the other hand, the more extensive data for thermal conductivity [10,11] seemed to favor the inclusion of the logarithmic term. This conclusion was confirmed by B. Le Neindre and coworkers [11].

In spite of this agreement, we hesitated to accept the conclusion that the presence of the logarithmic term was verified experimentally. First, in one case, the precision of the measurements was only one of ±1%. Secondly, the density intervals of both sets of measurements were quite large, and this necessitated the inclusion of data pertaining to excessively high densities (800 Amagat units) in order to extract the required, statistically significant, information. The latter point is particularly important owing to the circumstance that the omission of higher-order terms beyond those quoted in equations (1) and (2) may not be justified in such an extended range.

The preceding reasons induced us to undertake new measurements of a transport property and to arrange them so that they would be conducive to a more stringent statistical analysis. We chose to measure the viscosity of nitrogen, argon and helium in an oscillating-disk instrument [12] because we could obtain in it a precision of a few parts in ten thousand. Furthermore, we were able to make measurements at very closely spaced intervals of pressure and thus to provide a large population of data in a moderate density range.

SECTION II

EXPERIMENTS

Measurements were performed on nitrogen (N_2), argon (Ar), and helium (He) at a nominal temperature of 25°C and over a range of pressures, as follows:

N_2 *up to* $P = 105$ atm *and* $\rho = 0.12$ g/cm^3

Ar *up to* $P = 100$ atm *and* $\rho = 0.17$ g/cm^3

He *up to* $P = 100$ atm *and* $\rho = 0.02$ g/cm^3

They were evaluated on a relative basis [13,14]. The temperature in the instrument was maintained by thermostating the room in which the instrument was accommodated. The characteristics of the suspension system are given in Table I.

The working equation [14]

$$\frac{\pi\rho R^4 d}{2I}\left(3 + \frac{R^2}{bd}\right)Cx_o^{\,2} + (2\theta)^{-1/2}\,\frac{\pi\rho R^4 d}{I}\left(1 - \frac{3}{2}\Delta^2\right)Cx_o - \left\{\left(1 + \frac{\pi\rho R^4 b}{3I}C\right)\frac{\Delta}{\theta} - \Delta_o\right\} + \frac{1}{90}\,\frac{\pi\rho R^2 b^3}{I}\cdot\frac{1-3\Delta^2}{\theta^2}\,\frac{C}{x_o^{\,2}} = 0 \tag{3}$$

contains the calibration factor C which is a unique function of the boundary-layer thickness [14]

$$\delta = \sqrt{\eta T_o/\pi\rho}\ . \tag{3a}$$

Here, R denotes the radius of the disk, d is its thickness, I is its moment of inertia, b_1 and b_2 are the upper and lower spacings, respectively, both equal ($b_1 = b_2 = b$), T_o is the period of oscillation *in vacuo*, $\theta = T/T_o$ is the ratio of the actual period to that in a vacuum, and Δ is the logarithmic decrement of the damped harmonic oscillation performed by the system, Δ_o denoting its value *in vacuo*. Finally

$$x_o = \delta/R = \frac{1}{R}\sqrt{\frac{\eta T_o}{2\pi\rho}} \tag{3b}$$

TABLE I

CHARACTERISTICS OF SUSPENSION SYSTEM

Suspension wire	0.002 in. diam. 92% Pt-8% W. stress relieved
Wire damping	$\Delta_o = 0.000040 \pm 0.000004$
Total separation between plates	$D = (0.28443 \pm 0.00005)$ cm
Upper and lower separations	$b = b_1 = b_2 = (0.09006 \pm 0.00005)$ cm
Disk radius	$R = (3.4906 \pm 0.001)$ cm
Disk thickness	$d = (0.10431 \pm 0.00012)$ cm
Moment of inertia of suspension system	$I = (53.6032 \pm 0.0006)$ g·cm^2
Natural periods of oscillation	$T_o = (29.244 \pm 0.002)$ sec at 25°C (first calibration) $T_o = (29.216 \pm 0.002)$ sec at 25°C (second calibration)

computed numerically from equation (3), determines the viscosity, η.

In order to perform the three series of measurements, it turned out to be necessary to calibrate the instrument twice; owing to a mishap, the suspension wire was damaged before the measurements on argon were completed, and the oscillating system had to be re-assembled. The points obtained with the aid of the second calibration are marked with asterisks in the tables of results.

The calibration curve $C(\delta)$ was determined with respect to nitrogen in the pressure range 1-25 atm and at 25°C. The calibration values were taken directly from Table XV of reference [12]. Since the temperature in the instrument was not exactly equal to 25°C, differing from it, however, by at most 0.3°C, a small correction was applied. We based this correction on the circumstance that the excess viscosity is a function of density,

$$\eta(\rho,T) - \eta_0(0,T) = f(\rho) \tag{4a}$$

in both representations [15], equations (1) and (2). Hence, along an isochore ρ = const, we find that

$$\eta(\rho,T^*) = \eta(\rho,T) - \{\eta_0(0,T) - \eta_0(0,T^*)\}, \tag{4b}$$

where T^*=25°C. In this manner the only correction that needs to be applied is deduced from the zero-density values, η_0, a linear estimate of 0.47 μPoise/°C being adequate for the purpose. The corrected value now refers to the actual density maintained in the instrument but to a slightly different temperature. To account for this fact, the pressure recorded during the measurement must be corrected accordingly.

The density ρ, was calculated from the equation

$$\frac{PM}{\rho RT} = 1 + B_1P + B_2P^2 + B_3P^3 \tag{5}$$

with

$$R = 82.082 \text{ atm cm}^3/\text{g K}$$

$$M = 28.014 \text{ g/mole (on carbon 12 scale).}$$

The virial factors B_1, B_2, B_3 listed in Table II were interpolated from NBS Circular 564 [16], and a further linear interpolation routine between 20°C and 30°C was programmed in conjunction with the working equation (3). In the latter, the values of C were evaluated from known values of x_o, the reverse procedure being used for the determination of the viscosity, η, in measurements proper.

The two calibration curves are seen plotted in Figure 1; they can be correlated as follows:

$$C_1 = 1.089\ 0645 - \frac{0.002\ 5248}{\delta + 0.07} - \frac{0.000\ 7367}{(\delta + 0.07)^2} - \frac{0.000\ 0334}{(\delta + 0.07)^3} \quad (6a)$$

(standard deviation 1.5×10^{-4}*)*

$$C_2 = 1.089\ 5202 - \frac{0.003\ 1036}{\delta + 0.07} - \frac{0.000\ 5315}{(\delta + 0.07)^2} - \frac{0.000\ 0217}{(\delta + 0.07)^3} \quad (6b)$$

(standard deviation 1.8×10^{-4}*)*

The experimental results are given in Tables III-V. The density of nitrogen was evaluated in the same way as during a calibration, that of argon and helium was evaluated in like manner, except for the virial coefficients which are also listed in Table II.

The data for argon were taken from the measurements performed by E. Whalley, Y. Lupien and W. G. Schneider [17], those for helium were taken from the work of W. G. Schneider and J. A. H. Duffie [18]; it proved to be sufficient to retain terms up to $O(P^2)$ only.

In all cases a small correction for temperature was applied to reduce the data to 25°C as mentioned earlier in connection with the calibration curves. The actual temperature corrections were deduced from runs at slightly different temperatures, and turned out to have the following values:

Ar 0.71 μPoise/°C

He 0.46 μPoise/°C

In this manner, the standard viscosity, listed in the last column of Tables III-V, represents the viscosity of the gas at 25°C and at the actual density that prevailed during the measurement. For this reason, we evaluated the corresponding, corrected pressure and listed it in the second column in the tables, as already mentioned.

TABLE II - VIRIAL COEFFICIENTS FOR NITROGEN, ARGON, AND HELIUM

Gas	Temperature, °C	B_1, $(atm)^{-1}$	B_2, $(atm)^{-2}$	B_3, $(atm)^{-3}$
N *	20	-2.42×10^{-4}	2.13×10^{-6}	3.84×10^{-9}
	30	-1.47×10^{-4}	2.04×10^{-6}	2.64×10^{-9}
Ar**	20	-7.5194×10^{-4}	2.389×10^{-6}	- - -
	30	-6.2381×10^{-4}	2.1046×10^{-6}	- - -
He***	20	4.633×10^{-4}	- - -	- - -
	30	4.471×10^{-4}	- - -	- - -

*From reference [16]

**From reference [17]

***From reference [18]

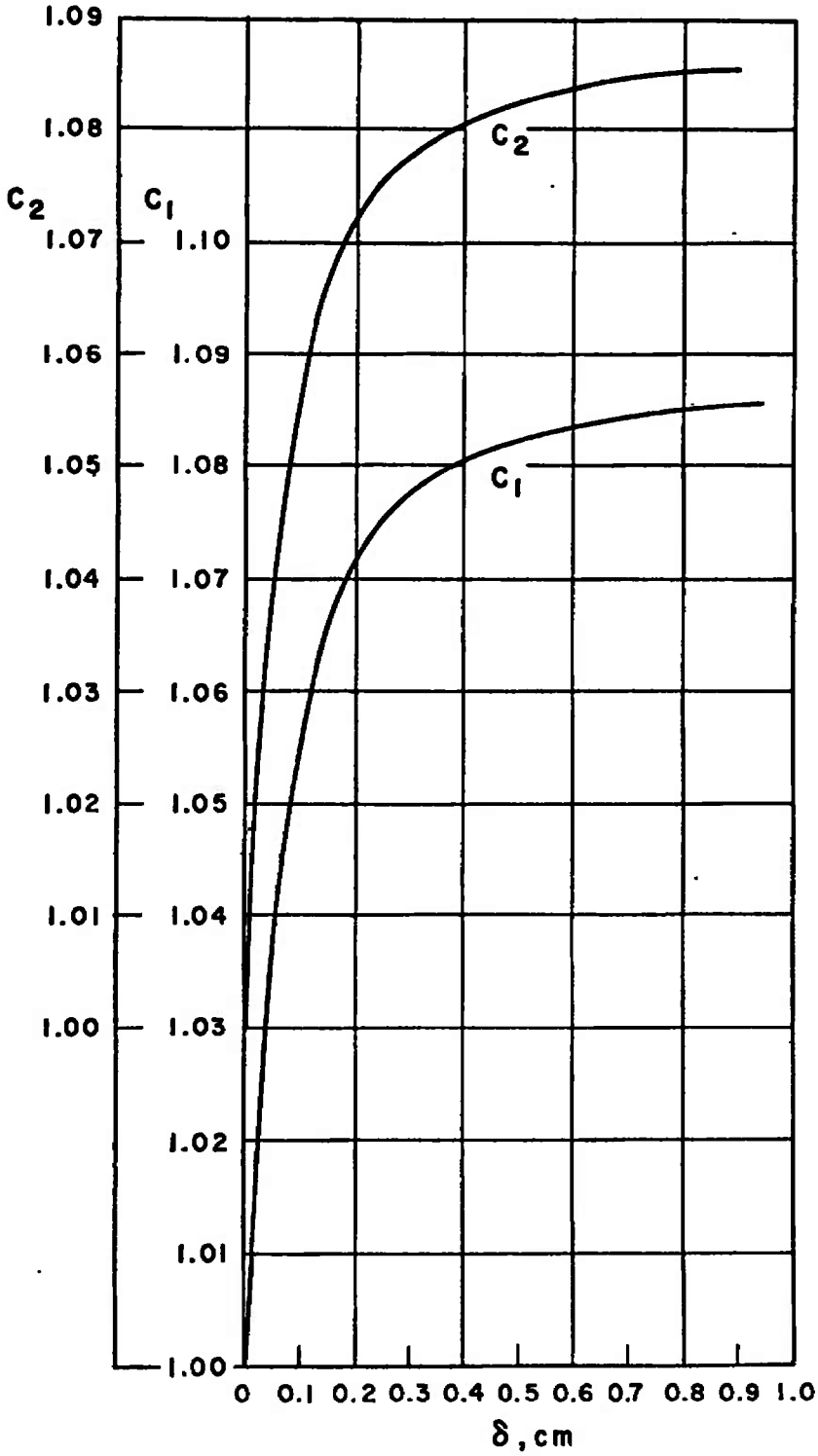

Figure 1. Calibration curves with respect to nitrogen at 25°C nominal and in the pressure range 1-25 atm.

Graphs of standard viscosity *versus* density are presented in Figures 2, 3 and 4. We estimate that the accuracy of the measurements is of the order of ±0.2%. The precision and internal consistency is, of course, much better, as will emerge from the analysis.

In assessing the reliability of the preceding experimental values, it is necessary to realize, as pointed out in our earlier experimental work, that equation (3) is quite insensitive to the assumed value of density. This is due to the fact that the damping in an oscillating-disk viscometer which is equipped with closely spaced fixed plates is very nearly independent of density. For example, if the density in equations (3) and (3a) were changed by as much as 1%, the viscosity evaluated from equation (3b) would change by less than 0.1%. Thus, an accurate knowledge of the density is inessential for the evaluation of viscosity. By contrast, the values of density assigned to the corrected pressures at 25°C must be computed with great care when an analysis of the density expansion of viscosity is undertaken. Nevertheless, the analysis is not sensitive to uncertainties in the actual values used for the virial coefficients B_k in equation (5). A choice of other values for these virial coeficients would represent a coordinate transformation $\rho' = f(\rho)$ where $f(\rho)$ is a power series in ρ. Such a coordinate transformation does not affect the presence or absence of a logarithmic term in equation (2).

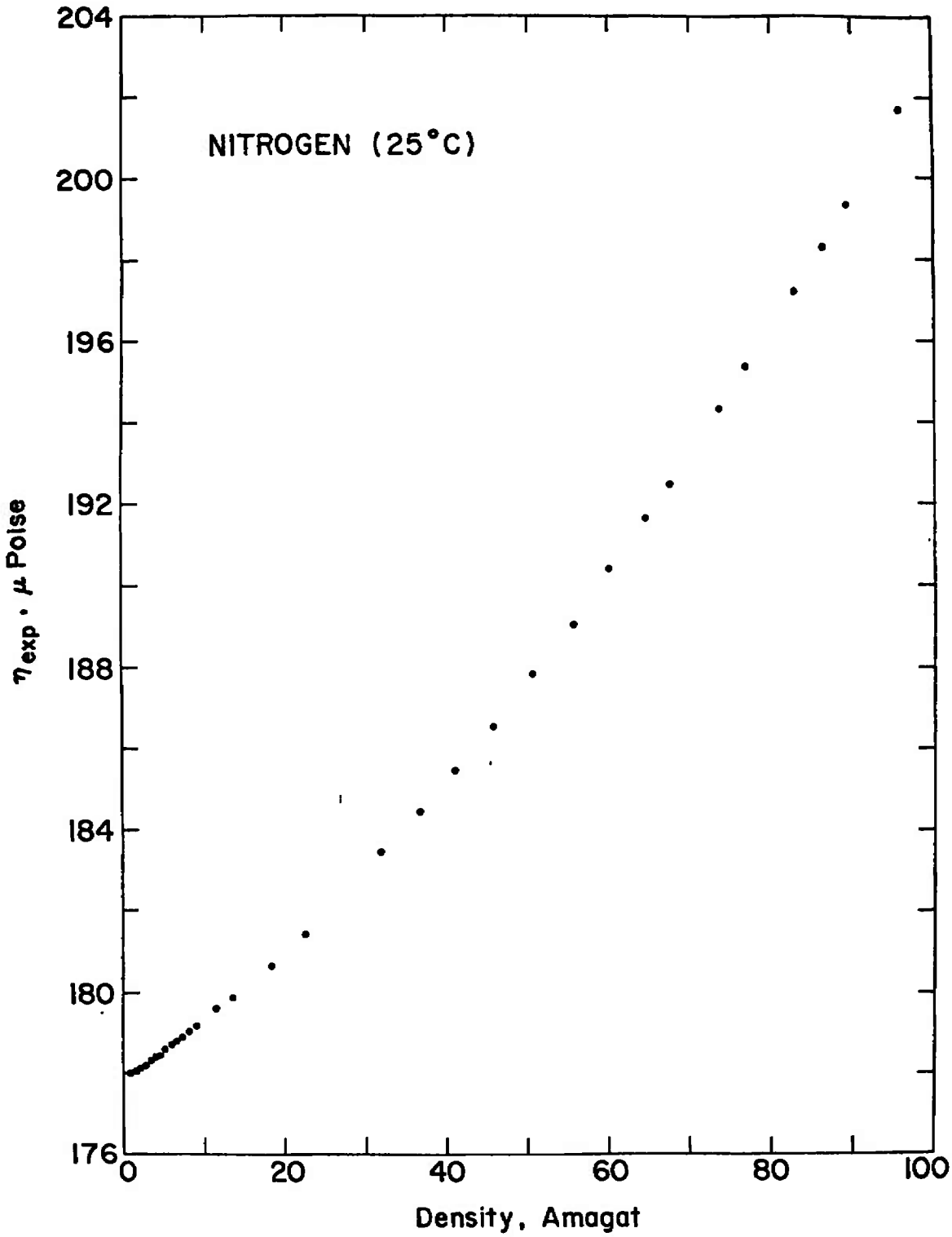

Figure 2. Viscosity of nitrogen as a function of density.

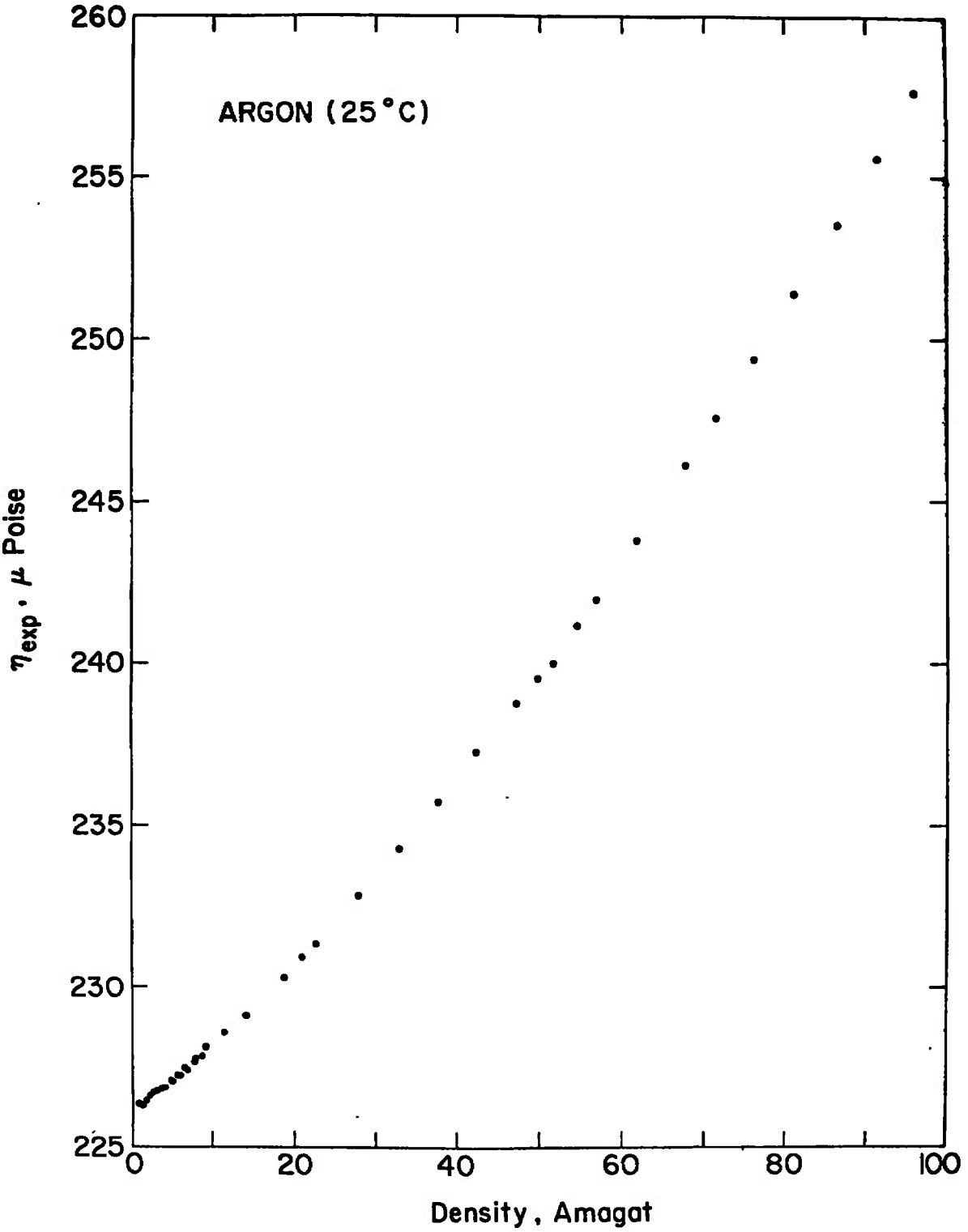

Figure 3. Viscosity of argon as a function of density.

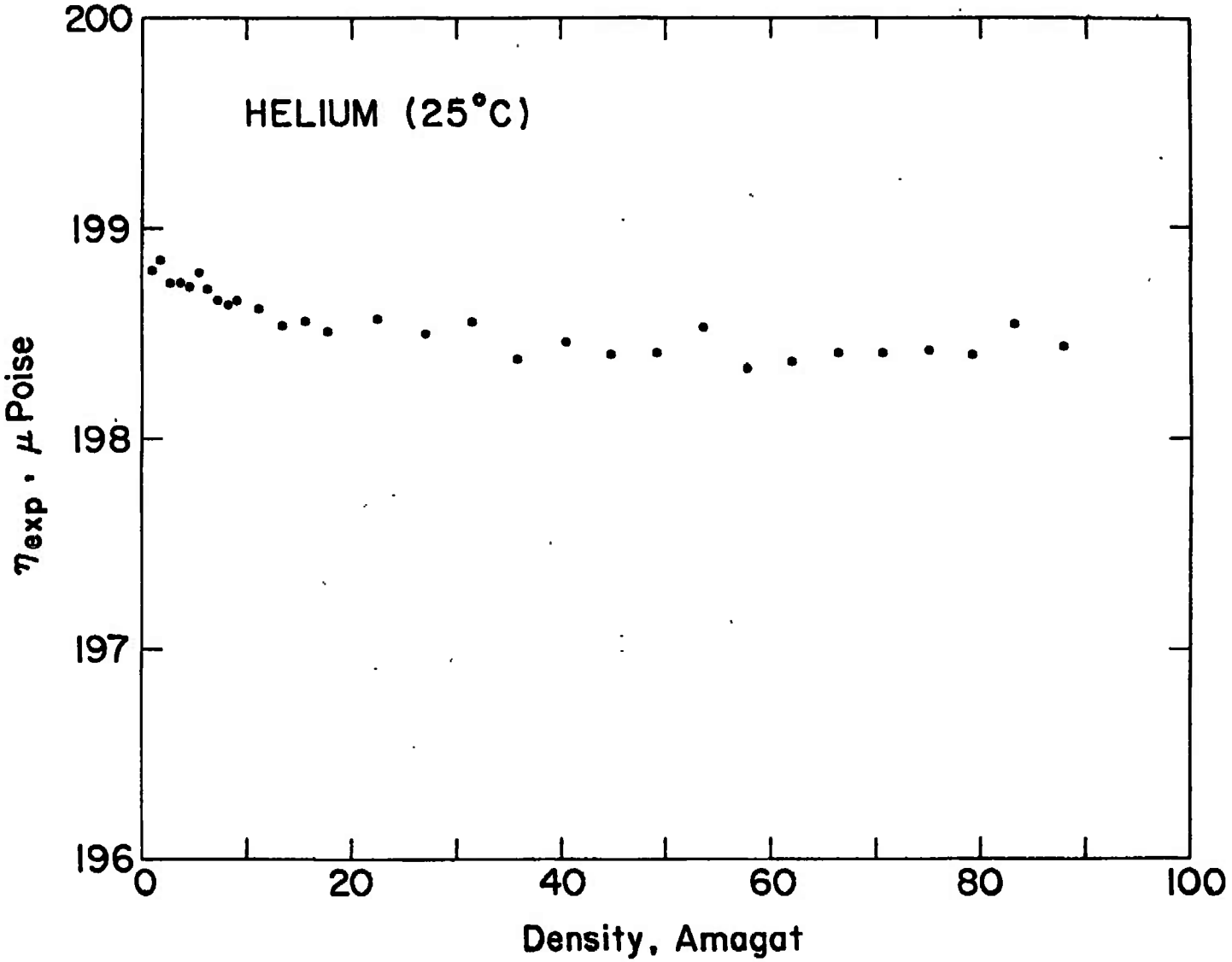

Figure 4. Viscosity of helium as a function of density.

TABLE III - THE VISCOSITY OF NITROGEN

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density* ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
1	1.000	1.000	0.85037	0.0011453	0.91590	24.920	177.9_4	177.9_8
2	1.714	1.716	0.64939	0.0019645	1.5710	24.864	178.0_0	178.0_6
3	2.300	2.302	0.56057	0.0026365	2.1084	24.727	178.0_0	178.1_3
4	3.009	3.009	0.49049	0.0034470	2.7566	24.959	178.1_8	178.1_9
5	3.715	3.718	0.44124	0.0042597	3.4065	24.722	178.1_9	178.3_2
6	4.341	4.344	0.40438	0.0049765	3.9797	24.819	178.3_2	178.4_1
7	4.979	4.981	0.38142	0.0057072	4.5641	24.890	178.3_9	178.4_4
8	5.579	5.583	0.36034	0.0063979	5.1164	24.784	178.4_9	178.5_9
9	6.613	6.616	0.33116	0.0075827	6.0639	24.874	178.6_7	178.7_3
10	7.307	7.312	0.31503	0.0083814	6.7027	24.804	178.7_2	178.8_1
11	8.029	8.036	0.30053	0.0092126	7.3674	24.744	178.7_8	178.9_0
12	8.954	8.964	0.28460	0.010279	8.2202	24.651	178.8_8	179.0_4
13	9.931	9.922	0.27082	0.011378	9.0991	25.280	179.3_0	179.1_7
14	12.567	12.579	0.24056	0.014433	11.542	24.702	179.4_5	179.5_9
15	14.880	14.864	0.22165	0.017057	13.641	25.334	180.0_4	179.8_8

TABLE III - THE VISCOSITY OF NITROGEN (continued)

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density* ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
16	20.051	20.068	0.19095	0.023045	18.429	24.749	180.5_4	180.6_6
17	24.644	24.638	0.17274	0.028307	22.637	25.068	181.4_8	181.4_5
18	29.838	29.806	0.15753	0.034258	27.396	25.318	182.6_4	182.4_9
19	34.816	34.807	0.14609	0.040016	32.001	25.084	183.4_9	183.4_5
20	39.919	39.958	0.13663	0.045946	36.743	24.712	184.2_9	184.4_3
21	44.682	44.663	0.12966	0.051357	41.070	25.131	185.5_2	185.4_6
22	49.853	49.840	0.12310	0.057304	45.826	25.077	186.5_8	186.5_4
23	55.126	55.106	0.11750	0.063344	50.657	25.113	187.8_9	187.8_4
24	60.603	60.602	0.11241	0.069635	55.688	25.003	189.0_5	189.0_5
25	65.434	65.419	0.10862	0.075132	60.083	25.070	190.4_4	190.4_1
26	70.401	70.421	0.10504	0.080825	64.636	24.916	191.6_1	191.6_5
27	73.599	73.560	0.10305	0.084386	67.484	25.162	192.5_5	192.4_7
28	80.403	80.375	0.09911	0.092094	73.648	25.102	194.3_7	194.3_2
29	84.009	83.970	0.09727	0.096140	76.884	25.139	195.4_4	195.3_7
30	90.643	90.624	0.09414	0.10360	82.850	25.062	197.2_3	197.2_0

TABLE III - THE VISCOSITY OF NITROGEN (continued)

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm^3	Density* ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
31	94.555	94.530	0.09248	0.10795	86.328	25.079	198.3_4	198.3_0
32	97.821	97.783	0.09120	0.11156	89.215	25.115	199.3_6	199.3_1
33	105.237	105.251	0.08852	0.11979	95.797	24.957	201.6_5	201.6_7

*1 Amagat = 0.00125046 g/cm^3

TABLE IV - THE VISCOSITY OF ARGON

Point number n	Experimental pressure P_e, atm	Corrected pressure P, atm	Boundary layer thickness δ, cm	Density ρ, g/cm³	Density** ρ, Amagat	Experimental temp. T_e, °C	Viscosity η_e, μPoise	Standard viscosity (at 25°C) η, μPoise
1	1.000	1.000	0.80283	0.0016340	0.91597	24.911	226.2_8	226.3_4
2	* 1.497	1.498	0.65559	0.0024472	1.3718	24.872	226.2_0	226.2_9
3	2.041	2.041	0.56212	0.0033360	1.8701	25.040	226.4_8	226.4_5
4	* 2.480	2.483	0.50924	0.0040593	2.2755	24.687	226.3_9	226.6_1
5	3.034	3.034	0.46106	0.0049633	2.7823	24.982	226.6_9	226.7_0
6	* 3.500	3.496	0.42949	0.0057211	3.2071	25.302	226.9_5	226.7_4
7	4.048	4.050	0.39893	0.0066301	3.7166	24.837	226.7_0	226.8_2
8	* 4.514	4.509	0.37817	0.0073832	4.1388	25.315	227.0_8	226.8_6
9	5.110	5.113	0.35518	0.0083756	4.6951	24.826	227.0_1	227.1_3
10	* 5.487	5.484	0.34287	0.0089853	5.0369	25.155	227.1_7	227.0_6
11	6.035	6.032	0.32714	0.0098873	5.5425	25.141	227.3_5	227.2_5
12	* 6.501	6.496	0.31508	0.010651	5.9706	25.212	227.3_9	227.2_4
13	7.158	7.155	0.30041	0.011737	6.5794	25.124	227.5_7	227.4_9
14	* 7.488	7.484	0.29354	0.012279	6.8832	25.150	227.5_3	227.4_2
15	* 8.498	8.581	0.27564	0.013943	7.8160	25.177	227.8_2	227.6_9

TABLE IV - THE VISCOSITY OF ARGON (continued)

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density** ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
16	8.586	8.585	0.27425	0.014095	7.9012	25.036	227.7_8	227.7_5
17	* 9.491	9.489	0.26073	0.015588	8.7382	25.067	227.9_0	227.8_5
18	10.029	10.032	0.25375	0.016486	9.2415	24.924	228.0_7	228.1_2
19	12.403	12.405	0.22826	0.020418	11.446	24.956	228.5_6	228.5_9
20	15.288	15.279	0.20580	0.025193	14.122	25.183	229.2_6	229.1_3
21	20.266	20.258	0.17890	0.033506	18.782	25.111	230.4_1	230.3_3
22	22.637	22.626	0.16939	0.037476	21.008	25.145	231.0_5	230.9_4
23	24.542	24.562	0.16255	0.040727	22.830	24.763	231.1_9	231.3_6
24	29.985	29.996	0.14735	0.049894	27.969	24.896	232.7_5	232.8_3
25	35.156	35.198	0.13619	0.058715	32.914	24.644	234.0_1	234.2_6
26	40.191	40.193	0.12774	0.067227	37.685	24.986	235.7_0	235.7_1
27	45.090	45.120	0.12077	0.075656	42.410	24.801	237.1_0	237.2_4
28	50.261	50.256	0.11467	0.084481	47.357	25.028	238.7_8	238.7_6
29	52.869	52.832	0.11200	0.088920	49.846	25.211	239.6_6	239.5_1
30	54.650	54.655	0.11014	0.092064	51.608	24.971	239.9_6	239.9_8

TABLE IV - THE VISCOSITY OF ARGON (continued)

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density** ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
31	57.779	57.725	0.10741	0.097371	54.583	25.280	241.3_6	241.1_6
32	60.154	60.122	0.10535	0.10152	56.909	25.161	242.0_6	241.9_5
33	65.162	65.144	0.10146	0.11023	61.792	25.083	243.8_4	243.7_8
34	71.421	71.380	0.09729	0.12108	67.874	25.172	246.2_3	246.1_1
35	75.300	75.297	0.09491	0.12791	71.702	25.012	247.5_6	247.5_5
36	79.926	79.968	0.09233	0.13607	76.277	24.842	249.2_5	249.3_6
37	85.029	85.020	0.08986	0.14491	81.232	25.031	251.4_2	251.4_0
38	90.303	90.274	0.08751	0.15412	86.395	25.095	253.5_7	253.5_0
39	95.303	95.253	0.08548	0.16285	91.289	25.156	255.6_7	255.5_6
40	100.032	100.084	0.08365	0.17132	96.037	24.845	257.5_5	257.6_6

*Calculations performed using calibration 2. **1 Amagat = 0.0017839 g/cm³

TABLE V - THE VISCOSITY OF HELIUM

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density* ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
1	1.190	1.191	2.1798	0.00019465	1.0905	24.807	198.7_1	198.8_0
2	2.014	2.016	1.6757	0.00032937	1.8452	24.751	198.7_3	198.8_5
3	3.000	3.003	1.3729	0.00049045	2.7476	24.718	198.6_1	198.7_4
4	4.021	4.026	1.1859	0.00065719	3.6817	24.662	198.5_9	198.7_4
5	5.001	5.007	1.0639	0.00081704	4.5773	24.647	198.5_5	198.7_2
6	6.008	6.015	0.97070	0.00098111	5.4964	24.644	198.6_3	198.7_9
7	6.987	6.993	0.90039	0.0011401	6.3871	24.734	198.5_9	198.7_1
8	8.028	8.036	0.83996	0.0013096	7.3367	24.693	198.5_1	198.6_6
9	9.049	9.057	0.79142	0.0014752	8.2644	24.744	198.5_2	198.6_4
10	9.954	9.965	0.75460	0.0016225	9.0896	24.658	198.5_0	198.6_6
11	12.397	12.404	0.67682	0.0020173	11.301	24.839	198.5_4	198.6_2
12	14.866	14.867	0.61852	0.0024153	13.531	24.973	198.5_3	198.5_4
13	17.227	17.230	0.57487	0.0027962	15.665	24.943	198.5_4	198.5_6
14	19.622	19.627	0.53884	0.0031816	17.824	24.930	198.4_8	198.5_1
15	24.882	24.885	0.47920	0.0040244	22.546	24.966	198.5_5	198.5_7

TABLE V - THE VISCOSITY OF HELIUM (continued)

Point number n	Experimental pressure P_e,atm	Corrected pressure P,atm	Boundary layer thickness δ,cm	Density ρ,g/cm³	Density* ρ,Amagat	Experimental temp. T_e,°C	Viscosity η_e,μPoise	Standard viscosity (at 25°C) η,μPoise
16	29.965	29.968	0.43710	0.0048353	27.089	24.975	198.4_9	198.5_0
17	34.918	34.921	0.40543	0.0056220	31.496	24.977	198.5_5	198.5_6
18	39.919	39.918	0.37948	0.0064122	35.923	25.007	198.3_8	198.3_8
19	45.090	45.097	0.35747	0.0072274	40.490	24.955	198.4_4	198.4_6
20	50.002	49.990	0.33990	0.0079940	44.784	25.073	198.4_4	198.4_0
21	55.160	55.186	0.32381	0.0088047	49.326	24.862	198.3_5	198.4_1
22	60.134	60.148	0.31062	0.0095753	53.643	24.932	198.5_0	198.5_3
23	64.855	64.889	0.29920	0.010308	57.748	24.845	198.2_7	198.3_4
24	69.857	69.853	0.28877	0.011073	62.034	25.017	198.3_7	198.3_7
25	75.061	75.109	0.27876	0.011878	66.543	24.811	198.3_2	198.4_1
26	79.960	79.963	0.27052	0.012619	70.695	24.987	198.4_1	198.4_1
27	85.165	85.207	0.26233	0.013416	75.160	24.854	198.3_5	198.4_2
28	90.064	90.080	0.25542	0.014153	79.289	24.947	198.3_8	198.4_0
29	94.589	94.712	0.24934	0.014851	83.199	24.613	198.3_7	198.5_5
30	100.049	100.152	0.24271	0.015667	87.770	24.692	198.2_9	198.4_4

*1 Amagat = 0.0001785 g/cm³

SECTION III

METHOD OF ANALYSIS

The verification of a proposed mathematical form for the density-dependence of the viscosity is hampered by the fact that a series expansion contains an *infinite* number of terms. Moreover, no rigorous expressions are available for the numerical evaluation of the coefficients in such an expansion beyond the Chapman-Enskog zero-density value of η_0. Thus there exists no *a priori* information regarding the number of terms of the expansion that would be adequate in representing the viscosity in a specified density interval. Even the dilute-gas value, η_0, cannot be established independently with sufficient precision, and its most probable value must also be extracted from the experimental data.

Nevertheless, it is possible to formulate heuristic criteria that ought to be satisfied, if a postulated equation is to be consistent with a population of experimental data. In Chapter IV of AEDC-TR-69-68, prepared in collaboration with H. J. M. Hanley and R. D. McCarty [4,7], such a set of criteria was formulated in conjunction with a systematic algorithm for the determination of successively higher-order terms in an equation such as (1) or (2), but truncated at some point.

In order to understand the algorithm, we consider the succession of polynomials

$$\eta = \eta_0 + \eta_1\rho \tag{7a}$$

$$\eta = \eta_0 + \eta_1\rho + \eta_2\rho^2 \tag{7b}$$

$$\eta = \eta_0 + \eta_1\rho + \eta_2\rho^2 + \eta_3\rho^3 \tag{7c}$$

each containing one more term than its predecessor. We now fit the first equation to the first n=5 points in the table of results for a particular gas, and record the least-square values of η_0 and η_1 together with their standard deviations and the variance, σ_η, for the set. This procedure is continued with n increased by unity in each successive fit. It is clear that at first the variance, σ_η, decreases with n, but after a certain value n=n' (or $\rho=\rho'$) has been reached, the variance, having passed through a stationary value, begins to increase, because the quadratic term comes into play. The same occurs with respect to the standard deviations of the coefficients. The calculation is then stopped, and the same procedure is followed with equations (7b) and

(7c) except that progressively larger values of n and n' are used. In this manner the algorithm allows us to determine an interval $0 < \rho < \rho'$ which provides the best fit of the data to a particular equation. As a last step, we repeat the same calculation in relation to the alternative form

$$\eta = \eta_0 + \eta_1\rho + \eta_2'\rho^2 \ln\rho + \eta_2\rho^2 \tag{7d}$$

In addition, detailed deviation plots are prepared to ensure that the distribution of the former is random, as expected. We say that an equation is *consistent* with the experimental data if the following, obviously necessary, conditions are satisfied:

(i) The deviations of the experimental data from those calculated with a fitted equation should be random, so that the standard deviation can be interpreted as true measure of the experimental precision.

(ii) When the values returned for the coefficients of the equation are studied as a function of n (or ρ), they should be independent of n within the limits prescribed by their respective standard deviations.

(iii) Within their standard deviation the coefficients should not change when the next term is added to the equation and the density interval is increased correspondingly.

It should be noted that the algorithm ensures that criteria *(i)* and *(ii)* are satisfied. The final decision as to whether a power series or the series with the logarithmic term included yields a superior empirical representation is made with reference to criterion *(iii)*.

SECTION IV

NUMERICAL EXPERIMENTS

Before applying the procedure described in the preceding section to a set of experimental results, it was thought advisable to perform a preliminary test on a set of artificially generated numbers and so to ascertain whether the algorithm is capable of producing a clear discriminant for the presence of the logarithmic term. For this purpose we generated data from the following three equations

$$\eta = 226.09 + 0.199\rho + 1.5 \times 10^{-3}\rho^2 - 1.8 \times 10^{-6}\rho^3 \quad (8a)$$

$$\eta = 226.09 + 0.199\rho - 0.2 \times 10^{-3}\rho^2 \ln\rho + 2 \times 10^{-3}\rho^2 \quad (8b)$$

$$\eta = 226.09 + 0.199\rho - 2.0 \times 10^{-3}\rho^2 \ln\rho + 2 \times 10^{-3}\rho^2 \quad (8c)$$

The coefficients in these equations have been chosen to correspond closely to the case of argon. Numerical data were generated at the densities (in Amagat units) listed in Table IV for argon with a random error added on to them; the latter was taken from a normal distribution with a variance of $\sigma=0.07$*.

Equations (7) were fitted to the data thus generated by the method of least squares. We used the Omnitab computer program for statistical analysis which uses a Gram-Schmidt orthogonalization procedure [19]. In a comparative study by R. H. Wampler [20] this method was shown to be reliable for the purpose at hand.

The results obtained for the fitted equations were studied as a function of ρ' to determine the range $0 < \rho < \rho'$, if any, in which criteria (*i*) and (*ii*) were satisfied. The most probable values for the coefficients of equations (7) are presented in Table VI. In this and the subsequent table n refers to the number of data points.

It is seen that the data generated by the cubic equation (8a) produce polynomials (7a), (7b) and (7c) which do satisfy criterion (*iii*). More precisely, the quadratic equation reproduces the values of η_0 and η_1 obtained with the linear equation and the cubic equation reproduces the values of η_0, η_1 and η_2 derived from the quadratic equation. Thus we

*The authors are indebted to Dr. D. T. Gillespie for providing them with suitable sets of random numbers.

TABLE VI - TEST OF THE ALGORITHM

Data generated with	n	ρ'	$\eta_0 \pm \sigma_{\eta_0}$	$\eta_1 \pm \sigma_{\eta_1}$	$(\eta_2 \pm \sigma_{\eta_2}).10^3$	$(\eta_3 \pm \sigma_{\eta_3}).10^6$	$(\eta_2' \pm \sigma_{\eta_2}').10^3$	σ_η
	12	6	226.08±0.05	0.202±0.014	- -	- -	- -	0.08
	30	52	226.07±0.03	0.202±0.003	1.37±0.07	- -	- -	0.07
Eqn. (8a)	40	96	226.09±0.03	0.198±0.004	1.5±0.1	-2.1±0.7	- -	0.07
	40	96	226.10±0.03	0.192±0.006	2.5±0.4	- -	-0.23±0.09	0.08
	(exact)		(226.09)	(0.199)	(1.5)	(-1.8)	(0)	(0.07)
	13	7	226.08±0.05	0.204±0.012	- -	- -	- -	0.07
	35	72	226.06±0.02	0.207±0.002	1.06±0.03	- -	- -	0.07
Eqn. (8b)	40	96	226.07±0.03	0.204±0.004	1.2±0.1	-2.0±0.7	- -	0.07
	40	96	226.09±0.03	0.198±0.006	2.1±0.4	- -	-0.23±0.08	0.07
	(exact)		(226.09)	(0.199)	(2)	(0)	(-0.2)	(0.07)
	15	8	226.10±0.04	0.187±0.009	- -	- -	- -	0.07
	24	28	226.00±0.04	0.242±0.008	-6.1±0.3	- -	- -	0.08
Eqn. (8c)	31	55	226.03±0.04	0.230±0.008	-4.8±0.4	-33±5	- -	0.07
	40	96	226.09±0.03	0.198±0.006	2.1±0.4	- -	-2.03±0.08	0.07
	(exact)		(226.09)	(0.199)	(2)	(0)	(-2)	(0.07)

conclude that the cubic polynomial is consistent with the data set. The value $\eta_1 = 0.192 \pm 0.006$, obtained from the logarithmic equation (7c), is somewhat lower than the corresponding term $\eta_1 = 0.202 \pm 0.003$ in the linear equation. Nevertheless, the two values agree within their combined standard deviations so that our criteria do not permit us to rule out a logarithmic term when it is multiplied with a coefficient of 2×10^{-4}. This conclusion is confirmed by the results obtained from the data generated according to equation (8b). This equation contains a logarithmic term with a coefficient of this magnitude. According to our criteria, both the cubic equation (7c) and the logarithmic equation (7d) are consistent with this data set.

Finally, we considered the data generated by equation (8c) where the coefficient of $\rho^2 \ln \rho$ is ten times larger than that in the previous equation. Our procedure now shows very clearly that the polynomials are not consistent with the data set. In other words, the quadratic and cubic equations do not reproduce the values of η_0 and η_1 of the linear equation, nor does the cubic equation reproduce the value of η_2 of the quadratic equation. On the other hand, the logarithmic equation (7d) does reproduce the factors η_0 and η_1 within their combined standard deviations and is thus consistent with the data.

We conclude that our algorithm can, indeed, distinguish between a polynomial such as equation (7c) or a logarithmic equation (7d). Of course, for data with a finite precision, the distinction can only be made with a finite resolution. With our current relative precision of 3×10^{-4} we can no longer detect a logarithmic contribution if its coefficient is as small as 2×10^{-4}.

SECTION V

ANALYSIS OF THE EXPERIMENTAL DATA

The experimental viscosity data were subjected to the analysis described in the previous two sections. The most probable values thus deduced for the coefficients of the equation (7a), (7b), (7c) and (7d) are listed in Table VII. In fitting the equations equal weights were assigned to the individual data points. However, essentially the same results are obtained when a weight proportional to the value of viscosity is assigned to the data, as shown in Table VIII. We note that the analysis indicates that the relative precision of the experimental data is better than 3×10^{-4}. Deviation plots showing $\eta_{exp} - \eta_{calc}$ for nitrogen and argon are presented in Figures 5, 6, 7, 8. In particular from Figures 6 and 8 we note that the deviation plots obtained with the cubic equation (7c) and the logarithmic equation (7d) are virtually indistinguishable for nitrogen as well as for argon. Evidently the two equations satisfy criterion (*i*) equally well.

If we apply in addition criterion (*iii*), we see from Table VII and VIII that the power series is consistent with the experimental data for nitrogen and argon to a high degree of satisfaction. This means that the linear, quadratic and cubic equations lead to the same coefficients. The logarithmic equation (7d) does not reproduce the value of η_1 of the linear equation nearly as well. Thus we conclude that from viscosity data with a relative precision of 3×10^{-4} a $\rho^2 \ln\rho$ term cannot be detected in the case of nitrogen or argon. It turns out that in the case of helium we cannot obtain significant information for all coefficients, but the results for the linear and quadratic equation are again in satisfactory agreement.

As demonstrated in the previous section our procedure would have revealed the existence of a logarithmic term if its coefficient were significantly larger than 2×10^{-4} μPoise/Am^2. Therefore, we conclude that such a coefficient is not larger than the above value, if it exists at all. In order to make this statement more precise, we rewrite equation (7d) as

$$f(\eta) = \eta - \eta_2' \rho^2 \ln\rho = \eta_0 + \eta_1 \rho + \eta_2 \rho^2 \qquad (9)$$

In this new equation η_2' is not considered as a coefficient to be determined, but as a parameter. Each value for the parameter η_2' defines a new set of data points $f(\eta)$. For the "correct" value of η_2' the data $f(\eta)$ should satisfy our criteria when fitted to a polynomial. Even when we relax

our criteria to allow variations within twice the standard deviations, we conclude that values for η_2' such that $|\eta_2'| > 6 \times 10^{-4}$ μPoise/Am^2 cannot possibly be reconciled with our experimental data for nitrogen or argon.

TABLE VII

Statistical analysis of the experimental data (equal weights).

	n	ρ' Am	$\eta_0 \pm \sigma_{\eta_0}$ μPoise	$\eta_1 \pm \sigma_{\eta_1}$ μPoise/Am	$(\eta_2 \pm \sigma_{\eta_2}).10^3$ μPoise/Am^2	$(\eta_3 \pm \sigma_{\eta_3}).10^6$ μPoise/Am^3	$(\eta_2' \pm \sigma_{\eta_2'}).10^3$ μPoise/Am^2	σ_η μPoise
N_2	7	5	177.85±0.02	0.133±0.007	- -	- -	- -	0.02
	21	41	177.85±0.02	0.132±0.002	1.31±0.06	- -	- -	0.03
	33	96	177.85±0.02	0.131±0.002	1.36±0.06	-1.5±0.4	- -	0.04
	33	96	177.87±0.02	0.126±0.004	2.1±0.3	- -	-0.18±0.04	0.04
Ar	14	7	226.10±0.04	0.199±0.009	- -	- -	- -	0.06
	27	42	226.09±0.02	0.196±0.003	1.59±0.09	- -	- -	0.05
	40	96	226.08±0.02	0.198±0.003	1.54±0.09	-1.9±0.6	- -	0.06
	40	96	226.09±0.03	0.192±0.005	2.4±0.4	- -	-0.22±0.07	0.06
He	14	18	198.83±0.02	-0.019±0.002	- -	- -	- -	0.03
	22	54	198.82±0.02	-0.019±0.003	0.24±0.05	- -	- -	0.05

TABLE VIII

STATISTICAL ANALYSIS OF THE EXPERIMENTAL DATA (proportional weights).

	η	ρ' Am	$\eta_0 \pm \sigma_{\eta_0}$ μPoise	$\eta_1 \pm \sigma_{\eta_1}$ μPoise/Am	$(\eta_2 \pm \sigma_{\eta_2}).10^3$ μPoise/Am^2	$(\eta_3 \pm \sigma_{\eta_3}).10^6$ μPoise/Am^3	$(\eta_2' \pm \sigma_{\eta_2'}).10^3$ μPoise/Am^2	σ_η %
N_2	7	5	177.85±0.02	0.133±0.007	- -	- -	- -	0.01
	21	41	177.85±0.02	0.132±0.002	1.31±0.06	- -	- -	0.02
	33	96	177.85±0.02	0.131±0.002	1.37±0.06	-1.6±0.4	- -	0.02
	33	96	177.87±0.02	0.126±0.004	2.1±0.3	- -	-0.18±0.05	0.02
Ar	14	7	226.10±0.04	0.199±0.009		- -	- -	0.03
	27	42	226.09±0.02	0.196±0.003	1.59±0.09	- -	- -	0.02
	40	96	226.08±0.02	0.199±0.003	1.54±0.09	-1.9±0.6	- -	0.03
	40	96	226.09±0.03	0.192±0.005	2.4±0.4	- -	-0.22±0.07	0.03
He	14	18	198.83±0.02	-0.019±0.002	- -	- -	- -	0.02
	22	54	198.82±0.02	-0.019±0.003	0.24±0.05	- -	- -	0.03

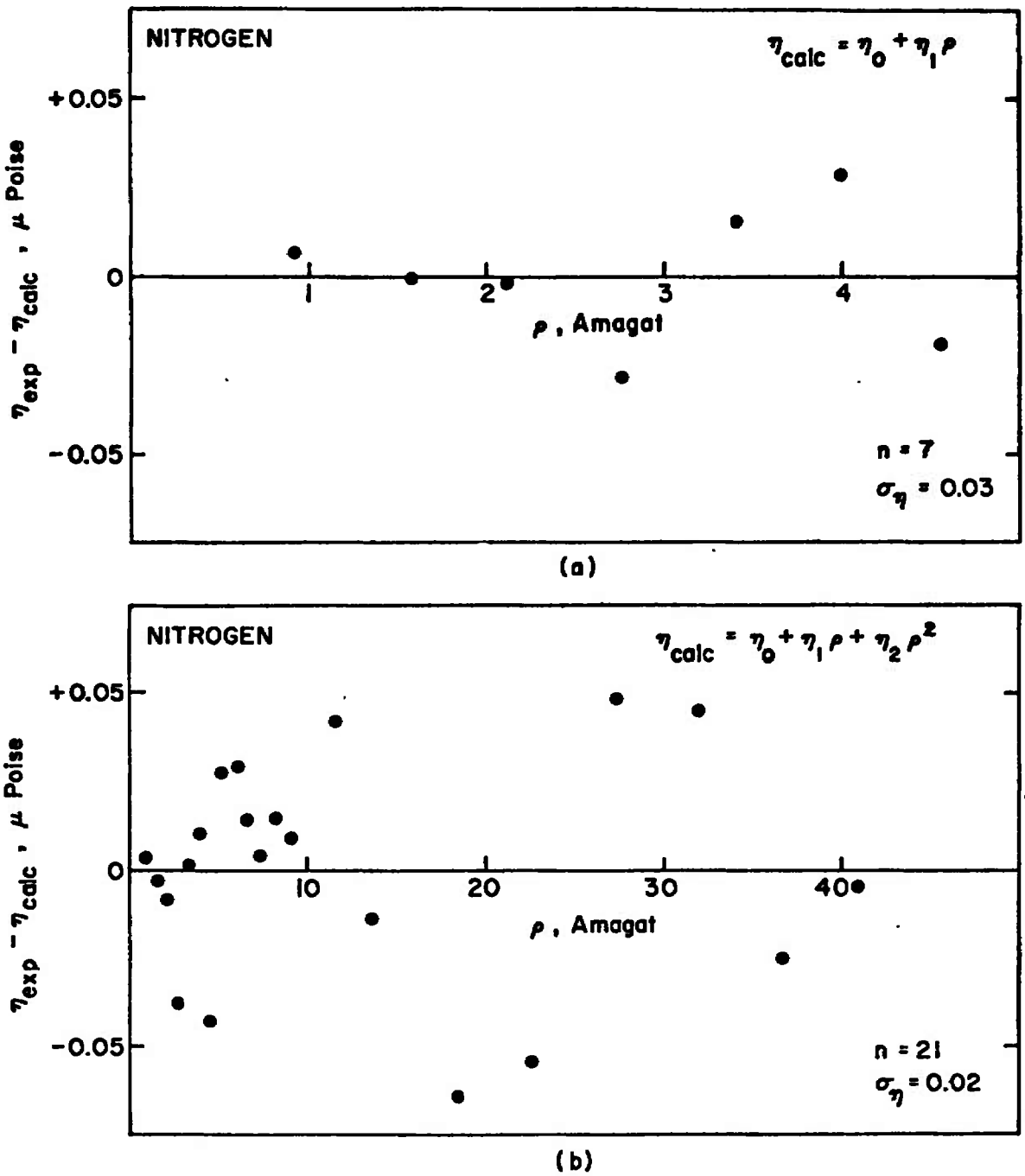

Figure 5. Deviation plots for the viscosity of nitrogen (a) Equation (7a); (b) Equation (7b).

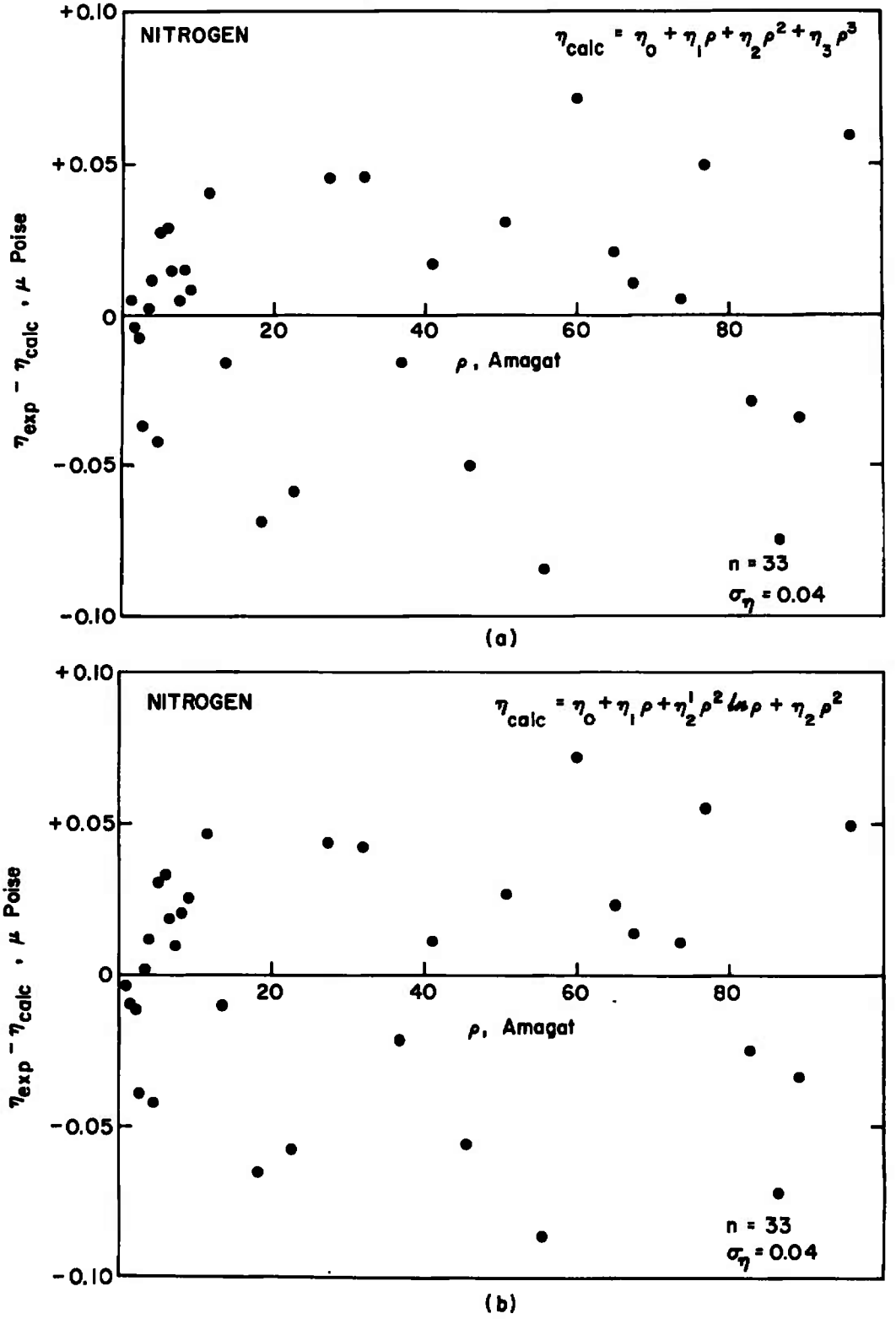

Figure 6. Deviation plots for the viscosity of nitrogen (a) Equation (7c); (b) Equation (7d).

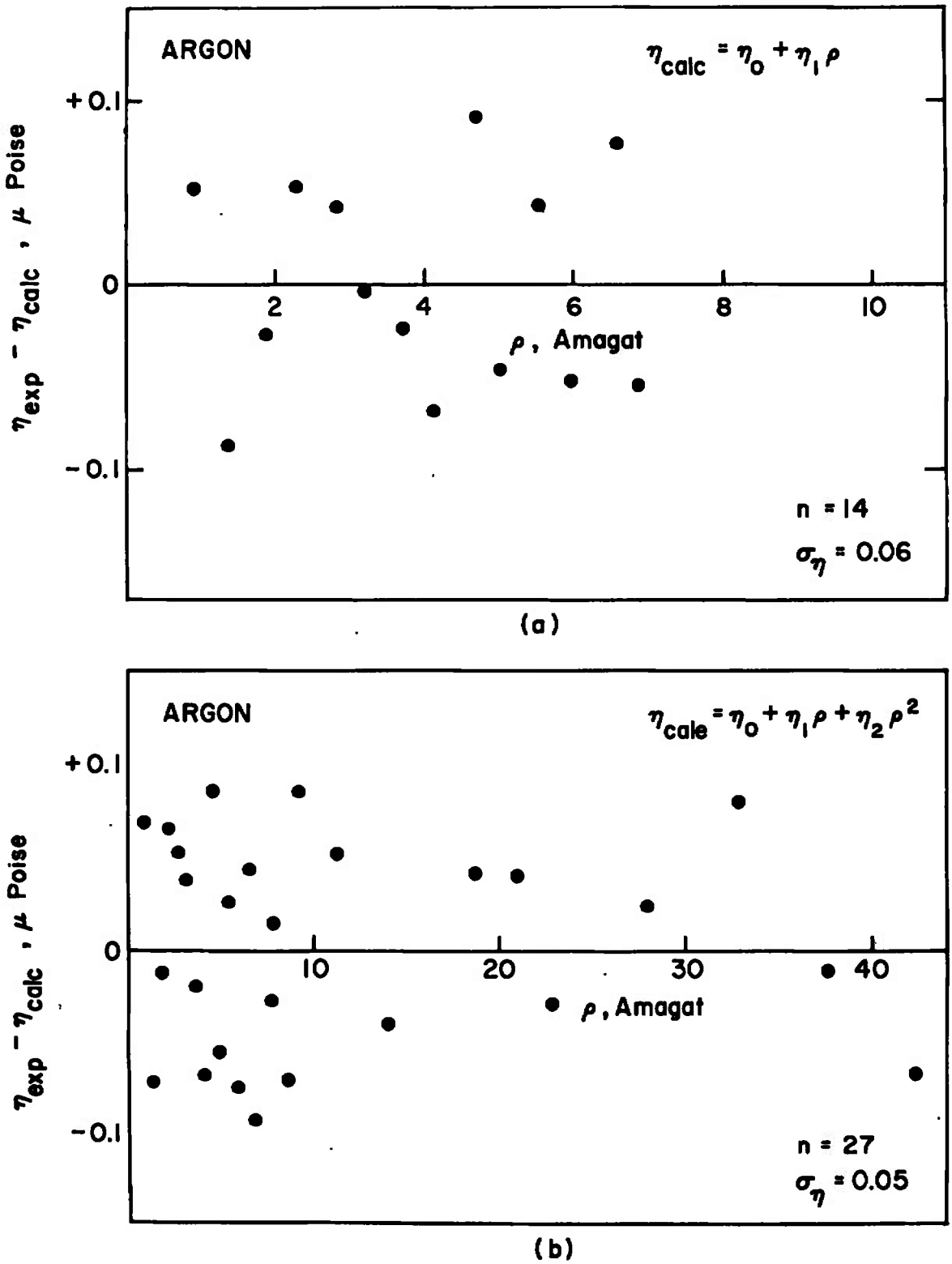

Figure 7. Deviation plots for the viscosity of argon (a) Equation (7a); (b) Equation (7b).

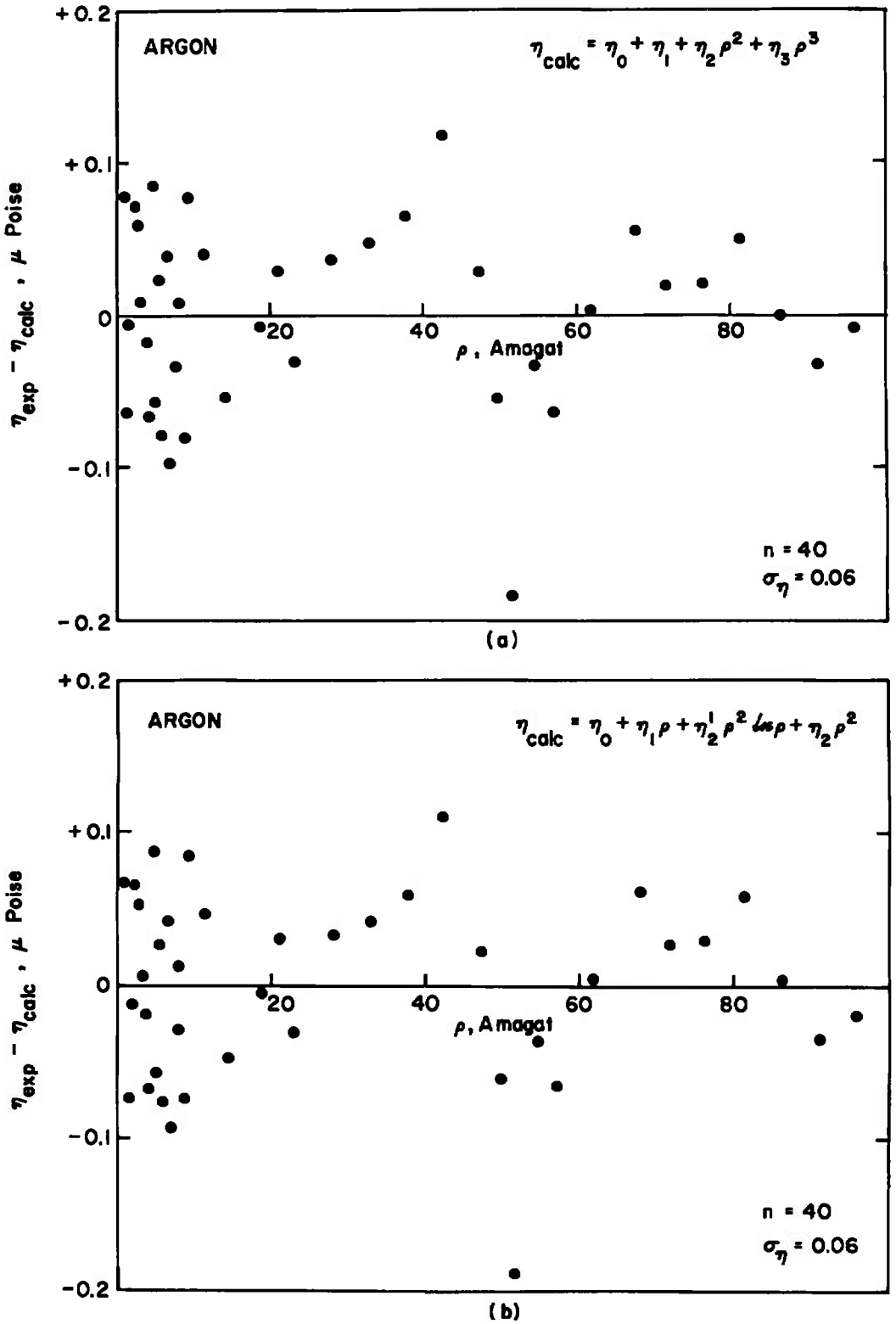

Figure 8. Deviation plots for the viscosity of argon (a) Equation (7c); (b) Equation (7d).

SECTION VI

DISCUSSION

We have demonstrated that the experimentally determined density dependence of the viscosity of argon and nitrogen at 25°C can be consistently represented by a power series in the density. Implicit in this conclusion is the assumption that the first four terms of the density expansion (1) and (2) are adequate to describe the data in the density range under consideration. The existence of a logarithmic term of the form $\rho^2 \ln\rho$ cannot be detected on the basis of viscosity data whose relative precision is 3×10^{-4}. If such a term exists, it is concluded that the absolute value of its factor, η_2', must be appreciably smaller than 6×10^{-4} μPoise/Am^2 for argon as well as nitrogen at 25°C. From a practical point of view, there is no need to include such a term in describing the density-dependence of the viscosity within the best precision currently available.

We would like to point out that the same conclusion is suggested by the *theoretical* results recently reported in AEDC-TR-71-51[5]. Although the theory does predict the existence of a logarithmic term $\rho^2 \ln\rho$, for a comparison with experiment it is important to have an estimate of its factor, η_2', relative to the magnitude of the coefficient η_2 of the quadratic term. A theoretically calculated value for η_2' is not available at the present. Nevertheless, we venture to make an estimate based on some considerations developed in AEDC-TR-71-51. In that report a distinction was made between correlations that are of a *statistical* nature versus those that are of a purely *dynamical* nature. Statistical correlations are related to those molecular configurations where the particles are close together in their mutual interaction range. The dynamical correlations are brought about by sequences of successive binary collisions that are spatially separated by distances larger than the molecular interaction range. In AEDC-TR-71-51 we indicated that the contributions from purely dynamical correlations are considerably smaller than those from statistical correlations. It was shown that for the first density coefficient η_1 the dynamical correlations constitute at most a few percent relative to the statistical correlations, referred to as double-overlap term or Enskog term.

It should be remembered that the origin of the $\rho^2 \ln\rho$ term in (2) is entirely of a dynamical nature, whereas the coefficient η_2 of the quadratic term will be determined primarily by statistical correlations. Thus we expect that the coefficient η_2' would be considerably smaller than the corresponding coefficient η_2. This explanation is supported by some calculations for a two-dimensional gas reported in AEDC-TR-69-68 [4,21]. The corresponding ratio for a

two-dimensional gas η_1'/η_1 turned out to be as small as 3×10^{-2}. We think that this result is indicative, notwithstanding the fact that recent developments have indicated that transport coefficients of a two-dimensional gas may not exist in the long wave length limit [22].

Based on the foregoing considerations we estimate that η_2'/η_2 should be not larger than a few percent. Since $\eta_2 \approx 10^{-3}$ μPoise/Am^2 for argon and nitrogen, we estimate that η_2' would be at most of the order of 10^{-5} μPoise/Am^2. This estimate is not only consistent with our experimental results, but we conclude, in retrospect, that a logarithmic term would not reveal itself even if the precision of the experiment would be increased by one or two orders of magnitude.

Both experiment and theory indicate that a power series representative for the density dependence of the transport properties will be adequate for practical purposes.

APPENDIX

In addition to the principal conclusions of the present paper regarding the form of the density expansion, the experimental data can be utilized in a practical way to provide us with very precise data on the zero-density value of the viscosity of the three gases and on the linear correction term. Referring to Table VII we can summarize them as follows:

NITROGEN: $\eta_0 = (177.85 \pm 0.02)$ μPoise at 25°C

$\eta_1 = (0.132 \pm 0.007)$ μPoise/Amagat at 25°C

ARGON: $\eta_0 = (226.09 \pm 0.04)$ μPoise at 25°C

$\eta_1 = (0.198 \pm 0.009)$ μPoise/Amagat at 25°C

HELIUM: $\eta_0 = (198.82_5 \pm 0.02)$ μPoise at 25°C

$\eta_1 = -(0.019 \pm 0.003)$ μPoise/Amagat at 25°C

Evidently, the preceding optimum values disregard the possibility of the existence of unaccounted for systematic errors.

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13. ABSTRACT

The paper presents the results of new, precise measurements of the viscosity of nitrogen, argon, and helium at 25°C. The measurements were performed over a nominal range of pressures 1-100 atm and at very closely spaced density intervals. The data were subjected to a stringent statistical analysis in order to determine whether the density expansion consists of a pure polynomial or whether a term of the form $^{2}\ell n$ must be included in it. The existence of such a term was discovered theoretically by several investigators. The analysis indicates that if such a term exists, its factor must be very small. This result is interpreted as indicating that correlations which extend over distances of the order of a mean free path are negligible when compared with correlations which extend over distances of the order of the range of molecular interactions.

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