DETERMINATION OF COMPRESSIBILITY AND VIRIAL COEFFICIENTS BY THE BURNETT METHOD

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An analytical method of determining the compressibility and virial coefficients from the experimental results by the Burnett method is described.

1. INTRODUCTION

One of the most well-known methods of measuring the compressibility of gases and gas mixtures is the Burnett method [1]. This method and its various modifications can be used in the range of temperatures 80-1200°K and for pressures up to 70 MPa [2-5]. In the isochoric variation of the method [6-8] one can perform accurate measurements in cases where PV is a rapidly varying function of P, i.e., close to the saturation curve, near the critical point, etc. [9-11]. The main advantage of the method is its high precision and relative simplicity experimentally, since the method does not require an exact determination of the volume and quantity of material, or the use of liquid mercury as a seal, etc. The natural experimentally measurable quantities are the pressure and temperature, which can be determined with high precision. As a result, the values of the compressibility and virial coefficients can be determined with high precision. For example, in a typical measurement with an error range of 0.01-0.03% in the pressure and 0.01-0.02°K in the temperature, one can determine the value of Z with an error less than 0.1% and B(T) to within 0.3 cm^3/mole.

The fundamental equation in the Burnett method can be written in the form

$$Z_j = P_j (Z_0/P_0) N_\infty^j \prod_{i=1}^{j} \zeta_i, \tag{1}$$

where P_j and Z_j are the pressure and compressibility of the gas after the j-th expansion; P_0, Z_0, initial values of the pressure and compressibility; N_∞, geometrical cell constant at zero pressure, $N_\infty = (V_I + V_{II})/V_I$; ζ, correction for barometric and isothermal (when the volumes V_I and V_{II} are at different temperatures) deformations.

It follows from (1) that if the values of N_∞ and Z_0/P_0 (second Burnett constant) are known, then the compressibility can be calculated at any pressure. In several recent papers [12-14] the fundamental relation (1) is expressed in virial form

$$P_j = \frac{\rho_0 RT}{N_\infty^j \prod_{i=1}^{j} \zeta_i} \left[1 + \sum_{k=2}^{n} B_k \left(\frac{\rho_0}{N_\infty^j \prod_{i=1}^{j} \zeta_i} \right)^{k-1} \right], \tag{2}$$

which opens up many possibilities for different approaches to determining the unknown quantities N_∞, $\rho_0 = 1/[(Z_0/P_0)RT]$, and B_k.

Originally, Burnett proposed to determine both constants by graphical analysis using the equation

$$N_\infty = \lim_{P \to 0} P_{j-1}/P_j, \tag{3}$$

$$P_0/Z_0 = \lim_{P \to 0} P_j N_1 N_2 \ldots N_j, \tag{4}$$

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where $N_j = N_\infty \zeta_j$ is the cell constant for the j-th expansion. However, the values obtained in this method are poor, particularly N_∞. Hence several new papers have appeared devoted to determining these quantities.

Miller [15, 16] and Silberberg [17] linearized the equations and, using a method resembling the method of least-squares, found corrections to the values of the constants. Canfield [18, 19], in order to find an accurate value of N_∞, used the linear dependence of $(Z - 1)/\rho$ on ρ for low pressures. However, it is well-known that in these coordinates, the scatter of the experimental points increases greatly with decreasing density; this makes it difficult to extrapolate the experimental isotherms to zero density in order to find N_∞. Hoover, Canfield, Kobayashi, and Leland [20] worked out a direct iterative method of getting the compressibility and lowest-order virial coefficients directly from the pressure measurements. The disadvantage of this method is that it is inapplicable for temperatures $T/T_c \leqslant 0.75$ and even for $T/T_c < 1.0$ does not give very accurate results. In addition, one is limited to pressures where the equation of state with second and third virial coefficients is applicable. Silberberg, Lin, and McKetta [21] have described an accurate graphical method of analyzing the results, but it is very laborious and its theoretical foundation is not completely rigorous. Tsiklis, Linshits, and Semenova [22] have studied the special case of (2) with the first two terms to determine ρ_0 and B_2. Obviously, this method gives good results only when the behavior of the gas can be described by an equation of state with the second virial coefficient over a wide range of low and moderate pressures; this is far from being true in all cases. A novel method of determining both constants was proposed by Altunin and Koposhilov [23], in which the values of N_∞ and Z_0/P_0 were found from the corresponding corrections to the measured pressures.

A general approach to the problem was proposed by Barieau and Dalton [24] based on the nonlinear least-squares method and a search for the unknown parameters according to the procedure of Newton. This method has been developed further by Hall and Canfield [12], and Waxman et al. [14, 25], in which the quality of the experimental values was looked at more closely, and more rigorous and better computational methods were used. Wielopolski and Warnowny [13] proposed an analytical method of finding N_∞, ρ_0, and B_k which is simpler than in [12, 25]. However, the virial coefficients are simply identified with the corresponding coefficients of the polynomial in (2); this is not rigorous in general and does not give the true values of the second and third virial coefficients [26, 27].

Hence at present the most rigorous and accurate methods of finding the compressibility and lowest order virial coefficients from the Burnett data are the methods of Hall—Canfield and Waxman et al. However, the practical application of these methods, particularly the computational programs, is very complex; this apparently has prevented their being widely used. We have worked out a method which is considerably simpler without loss of rigor or accuracy in the results.

2. NEW METHOD OF DETERMINING THE CONSTANTS

The principal difficulty in the use of the analytical methods described above is that one must solve a complicated set of nonlinear equations. Therefore, the fundamental problem is to find a way to avoid having to do this.

For finding the value of N_∞, in principle two approaches can be used: 1) either determine it for each series of expansions for all materials; or 2) determine the value for a standard gas and assume that it remains constant for a given temperature. In most papers, particularly recent ones, the first approach is favored. However, the detailed highly accurate study in [25], as well as our analysis of the most carefully conducted experiments using the Burnett method, lead to the conclusion that the second approach is also valid and the results are no less accurate than in the first approach. In this approach the true value of N_∞ is found by first obtaining an approximate value with the help of the classical method, and then refining this value by various methods [18, 19, 25]. If one takes helium as the standard gas (as is often done), then this approach is more logical and significantly simpler than the other method.

Using (1) we obtain the following expression for the ratio of compressibilities for the j-th and (j − 1)-st expansions:

$$Z_j/Z_{j-1} = P_j N_{\infty j} \zeta_j / P_{j-1} \tag{5}$$

from which we have

$$N_{\infty j} = (Z_j/Z_{j-1})/(P_j\zeta_j/P_{j-1}). \tag{6}$$

The thermodynamical properties of helium are now known very accurately over wide ranges of temperature and pressure [28]. It follows from the analysis of this data that in the usually studied interval of temperature 140-1000°K and for pressures up to 9 MPa, an equation of state with the first two virial coefficients is valid (the maximum error due to ignoring higher order virial coefficients is less than 0.0005%):

$$Z = 1 + \beta P + \gamma P^2, \tag{7}$$

where β and γ are related to the bulk virial coefficients by the relations

$$\beta = B/(RT),\ \gamma = (C - B^2)/(RT)^2. \tag{8}$$

Knowing the values of β and γ along the isotherm under study, one can calculate Z_j for the corresponding experimental pressure P_j and from (6) find $N_{\infty j}$ for the j-th expansion. The final value of N_∞ is taken to be

$$N_\infty = \Bigl(\sum_{j=1}^{n} N_{\infty j}\Bigr)\Big/ n. \tag{9}$$

We use the following temperature dependence for B and C:

$$B(T) = 7.682 + 24.182\tau - 45.932\tau^2 + 36.759\tau^3 - 11.121\tau^4,\ 80 \leqslant T \leqslant 1200\ \mathrm{K}, \tag{10}$$

$$C(T) = 48.20 + 268.07\tau - 403.61\tau^2 + 415.97\tau^3 - 151.85\tau^4,\ \ 80 \leqslant T \leqslant 600\ \mathrm{K}, \tag{11}$$

where $\tau = 100/T$.

These equations were obtained from the results of [2, 3, 29-32] and reproduce all the experimental data with an error less than the experimental error. Starting with 140°K, the range of pressures for which (7) is correct decreases with decreasing temperature. In this case it is necessary to use in addition the data of [2, 28] for calculating Z_j.

With the value of N_∞ known, the problem of finding the other parameters is considerably simplified. We write (2) in the form

$$P_jQ_j = \sum_{k=1}^{n} A_k/Q_j^{k-1}, \tag{12}$$

where

$$Q_j = N_\infty^j \prod_{i=1}^{j} \zeta_i;\ A_k = B_k\rho_0^k RT.$$

It can easily be seen that Eq. (12) has the same form as the usual virial equation. We note also that an increase in 1/Q rigorously corresponds to a widening of the density interval. Hence (12) can be treated as a virial equation, and the problem of finding the coefficients A_k reduces to the known problem of determining the virial coefficients from the compressibility data. With the help of the methods of [33, 34], the latter problem does not represent any serious difficulty. Hence the method of [33] allows one to find not only the A_k, but also establishes the optimum number of coefficients. However, it should be pointed out that in certain cases the method fails to cover all of the experimental data (points corresponding to the highest pressures) and some of the higher-order virial coefficients cannot be determined. Nevertheless, the method is useful because it gives a more rigorous determination of the values of all of the lower-order coefficients.

After the values A_1, A_2, ..., A_k are calculated, using the condition $B_1 = 1$, we can find ρ_0, then B_2, B_3, etc. In cases where all of the coefficients necessary to describe the data cannot be determined, the compressibility is calculated from (1).

TABLE 1. Comparison of the Values of N_∞ Calculated According to the Method of the Present Paper and the Method of Hall and Canfield*

Series	323,15 K		273,15 K		223,15 K	
	our paper	[3]	our paper	[3]	our paper	[3]
1	1,56490(5)	1,56490(2)	1,56489(2)	1,56490(2)	1,56486(4)	1,56484(2)
2	1,56486(5)	1,56486(2)	1,56492(3)	1,56491(2)	1,56483(4)	1,56483(2)

Series	113,15 K		103,15 K		83,15 K	
	our paper	[2]	our paper	[2]	our paper	[2]
1	1,56316(3)	1,56318(3)	1,56322(5)	1,56324(4)	1,56326(2)	1,56330(3)
2	1,56318(5)	1,56320(3)	1,56323(6)	1,56327(4)	1,56325(5)	1,56327(3)

*The values of $\delta(N_\infty)\cdot 10^5$ are given in parentheses.

TABLE 2. Comparison of the Values of N_∞ Calculated According to the Method of the Present Paper and the Method of Waxman*

Series	223,15 K		273,15 K		323,15 K	
	our paper	[29]	our paper	[29]	our paper	[29]
1	1,58807(3)	1,58819(3)	1,58816(1)	1,58823(4)	1,58833(3)	1,58845(3)
2	1,58799(3)	1,58808(2)	1,58819(7)	1,58825(4)	1,58849(2)	1,58859(2)
3	1,58805(5)	1,58814(3)	1,58816(3)	1,58819(4)	1,58838(4)	1,58850(3)
4	1,58801(4)	1,58810(3)	1,58819(5)	1,58822(4)	1,58834(4)	1,58846(3)

*The values of $\delta(N_\infty)\cdot 10^5$ are given in parentheses.

3. RESULTS AND DISCUSSION

Cell Constant. Tables 1 and 2 give values of N_∞ calculated with our method, the Hall—Canfield method [12], and the method of Waxman et al. [25]. It can be seen that the value of N_∞ obtained with our method agrees with the Hall—Canfield value within the error of its determination. The value of N_∞ found using the graphical-analytic method of Waxman et al. is too large by $1\cdot 10^{-4}$ (0.006%) in comparison to our value.

The results show that our method gives N_∞ within 0.002-0.004% for a given series of expansions. Discrepancies among the different series, as shown in Table 2, can be somewhat larger (0.004-0.007%).

The effect of temperature on the value of N_∞ is of particular interest. Physically, the value of N_∞ should not depend on temperature, and this is supported by the data of [2, 3, 30]. But the results of [29] (see Table 2) and [35] show that N_∞ changes rather significantly with temperature. All of these experiments were carried out with great care and are highly accurate; thus, it is difficult to explain the observed discrepancies in the dependence of N_∞ on temperature. Therefore, in order to increase the accuracy of the final results, we determine the cell constant N_∞ for each isotherm.

Compressibility. Our method of calculating the compressibility was checked on data for argon in the temperature range 143-323°K [3, 30], for nitrogen in the range 83-273°K [2, 35], and for krypton in the range 223-323°K [29]. The values of Z obtained were compared with those calculated in the methods of Canfield [18, 19], Hall and Canfield [12], and Waxman et al. [25]. The results are shown in Table 3.

It is clear from Table 3 that the values of Z calculated with our method agree closely with those calculated form the analytical methods of Hall—Canfield and Waxman et al., while the deviations in the values found by the Canfield method exceed the error in determining Z. We believe that the explanation is that the second constant in the Canfield method is found graphically from (4) with insufficient accuracy.

TABLE 3. Deviation of the Compressibility Calculated According to the Present Method from the Values Obtained by Other Methods

Method	Literature source	Mean-square deviation, %	Maximum deviation, %
Hall and Canfield [12]	[2, 3, 30]	0,015	0,035
Waxman et al. [25]	[29]	0,022	0,048
Canfield [18, 19]	[35]	0,075	0,21

TABLE 4. Virial Coefficients for Helium at 0°C and for Nitrogen at —140°C Obtained by Various Methods

Method	k	Helium	Nitrogen
		B_k, $(cm^3/mole)^{k-1}$	B_k, $(cm^3/mole)^{k-1}$
Classical [1]	2	12,012	—91,88
	3	105,96	3087,1
Canfield [18, 19]	2	12,112	—90,62
	3	107,92	1951,9
Hoover et al. [20]	2	11,96	—91,99
	3	117,51	3119,0
Hall and Canfield [12]	2	12,237	—91,28
	3	94,89	2869,2
Barieau and Dalton [24]	2	12,243	—
	3	97,17	—
Wielopol'ski and Warowny [13]	2	12,082	—91,48
	3	105,84	3148,1
Present paper	2	11,985	—91,61
	3	111,85	3213,6

The error in the value of Z is found with the help of a method described in [12]; we do not consider this here. We consider only the effect of N_∞ on the accuracy of the final value of the compressibility, since this gives a crucial contribution in the error in Z. Numerous calculations show that an error in N_∞ of 0.01% results in an error in Z of 0.015% at low pressures (2 to 5 bar) and as high as 0.15% for high pressures (500 to 700 bar). The corresponding errors in Z for an error in N_∞ of 0.006% are 0.007 and 0.08%, respectively, and an error in N_∞ of 0.002% gives corresponding errors in Z of 0.003 and 0.04%, respectively.

The magnitude of the error resulting from inaccurate thermostatic control is easily obtained from (1) and for a given isotherm T is equal to $2\Delta T/T$, where ΔT is the uncertainty in the temperature.

Virial Coefficient. In all of the control examples discussed above, the values of the lowest virial coefficients calculated in the present method agreed with the values found in the methods of Hall—Canfield and Waxman et al. within the limits of the total error in their determination. As an example, Table 4 gives the values of the second and third virial coefficients for helium at 0°C and for nitrogen at —140°C obtained by various methods from the data of [35].

The standard deviation of one of the coefficients B_k is given by the well-known formula $\sigma B_k = \sigma\sqrt{C_{kk}}$, where σ is the mean-square deviation of the pressure calculated from (2) and C_{kk} is the diagonal element of the inverse matrix made up of values of the argument. However, in our method each coefficient except for the first is determined at fixed values of all of the previous coefficients, and therefore the error found from the above formula is not the true error. Detailed analysis shows that an error in N_∞ of 0.007% leads to errors not larger than 0.4 cm^3/mole for B_2 and 10% for B_3; an error of 0.003% in N_∞ results in errors of 0.25 cm^3/mole and 6%, respectively.

4. CONCLUSIONS

We have shown that our method of determining the compressibility and lowest-order virial coefficients from the Burnett data is not inferior in rigor and accuracy to the methods of Hall—Canfield and Waxman et al. [12, 25], yet it is much simpler since it is not necessary to solve a complicated set of nonlinear equations. The computational program for our method was developed in ALGOL for the BÉSM-6 computer, but can also be used on the series ES computer with some simple modifications.

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CONTRIBUTION OF CRYSTALLINE LATTICE DEFECTS TO THERMAL CONDUCTIVITY OF POROUS MATERIALS

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The relationship of pore thermal resistance and effective thermal conductivity of porous media to the processes of crystalline lattice defect formation and motion is demonstrated.

The effect of lattice defects on the thermal conductivity of crystalline solids is widely known [1, 2]. However, the dependence of thermal resistance of pores and intergrain boundaries in porous media on this factor has been studied insufficiently. At the same time, the thermophysical properties of porous oxide systems depend decisively on the intensity of heat transfer between grains.

A number of studies [3-6] have analyzed the temperature dependence of thermal conductivity of such systems in rarefied gaseous media. In particular, the possibility of reversible change in the interparticle contact spot under the action of thermal stresses has been noted [3]. The effects of change in gas pressure within pores [4], and heat–mass transfer with participation of both the gaseous [5] and solid [6] phases have been considered. However, these mechanisms cannot satisfactorily explain the available experimental data, according to which, e.g., [7], the thermal conductivity of high-purity (99.99%) yttrium oxides in a vacuum obeys the same laws as do technological-grade refractories, remaining stable for long time periods. Results of experiments with especially pure oxides compel assumption of the existence of a heat–mass transfer mechanism within polycrystalline porous materials related to development and motion of crystalline lattice defects within the temperature gradient field near the surface of pores and microcracks.

The effect of such processes can be analyzed conveniently using the "quasichemical approach" of Wagner, Crager, and Wink [8], with the aid of which the crystal-structure defect concentration can be determined for a given temperature and gaseous medium composition. In the presence of a temperature gradient within the pores and microcracks, the change in the equilibrium constant K of the defect formation process follows a law

$$K = A \exp(\Delta S^0/k) \exp(-\Delta H^0/kT). \tag{1}$$

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