

x-ray powder method, using a Nonius Guinier-type camera. The gallstone was removed from a 75-year-old female diabetic; she was operated on for what was thought to be appendicitis, but in fact she had an inflamed gallbladder containing a cream of calcium carbonate and cholesterol, and the gallstone.

The stone has almost perfect octahedral symmetry (Fig. 1); the maximum dimension is 2 cm and the weight is 3 g. There is no distinguishable nucleus. From x-ray photographs the dark-brown granular interior was determined to be calcite, aragonite, and vaterite; the dark-brown rough surface, calcite and vaterite. The small nodules on the surface are not single crystals. A weak diffraction line at 15 Å in the photograph of the interior has not been identified. No cholesterol was found, but the dark brown color indicates the presence of amorphous bile pigments.

Gallstones composed of calcium carbonate, with or without cholesterol, frequently occur (2), but we have found no record of any stone having all the three forms. Calcite and aragonite are

well-known naturally occurring minerals, but vaterite has been found only occasionally; as well as in gallstones, it has been identified in the shells of certain young gastropods and geologically in a calc-silicate rock at Ballycraigy, Northern Ireland (3). Conditions governing the deposition and coprecipitation of calcite, aragonite, and vaterite have been discussed by Stolkowski (4) and Kamhi (5). Which, if any, of these conditions exist in the gallbladder during the formation of carbonate-containing stones is not yet known.

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## Discrepancies between Viscosity Data for Simple Gases

**Abstract.** *It has been known for some time that Kestin and his co-workers have reported dilute gas viscosity coefficients which differ from the usually accepted values. Recent work from the Los Alamos Scientific Laboratory supplements Kestin's results. We show that there is no evidence for not accepting this different data. We feel that the whole subject of dilute gas viscosity measurements above room temperature should be reexamined both from the experimenter's and correlater's viewpoints. There is evidence that published tables may be incorrect by as much as 10 percent above 600°K.*

Numerous experimental workers have studied the viscosity of dilute gases. One might indeed assume that the experimental aspects of the viscosity have been sufficiently covered for the common gases and that the viscosity coefficient tables available are satisfactory. This assumption is not valid. We show here that, because of a serious and largely ignored disagreement over experimental values, published tables of the viscosity coefficient above room temperature are likely to be incorrect.

Kinetic theory provides rigorous expressions for dilute gas transport coefficients. For the viscosity coefficient,  $\eta$ , we have (1):

$$10^7 \eta = \frac{266.93 (MT)^{3/2}}{\sigma^2 \Omega^{(2,2)*} (T^*)}, \text{ g cm}^{-1} \text{ sec}^{-1} \quad (1)$$

Here  $M$  is the molecular weight,  $T$  (°K) is the temperature, and  $\sigma$  (Å) is a parameter representing the distance between two molecules. The term  $\Omega^{(2,2)*} (T^*)$  is called the collision integral: the entity involving the collision dynamics and the intermolecular potential between two molecules. The collision integral is determined as a function of reduced temperature,  $T^*$ . The reduced temperature is defined by the relation  $T^* = T/(\epsilon/k)$ , where  $\epsilon$  is the maximum energy of attraction between the molecules, and  $k$  is Boltzmann's constant.

Equation 1 can, however, be applied to experiment only if we know the intermolecular potential function. Quantum mechanics has yet to derive a function from first principles, so we are required to use a function based on a

model. Hence the ability to use Eq. 1 to correlate and predict experimental data rests on a proper choice of the model function.

We have recently clarified the overall relation of model functions, theoretical expressions (such as Eq. 1), and experiment (2-4). Two points from our study are relevant here, so we list them:

1) Potential functions have been derived and classified in terms of families. For example we have the  $m$ -6 family where  $m$  is a family parameter. If  $U(r)$  is the interaction potential of two molecules separated by distance  $r$ , the functions of the  $m$ -6 family are written:

$$U(r) = \epsilon \left[ \left( \frac{\sigma}{r} \right)^m - \left( \frac{\sigma}{r} \right)^6 \right] / \left[ \left( \frac{6}{m} \right)^{6/m-6} - \left( \frac{6}{m} \right)^{m/m-6} \right] \quad (2)$$

where the distance parameter  $\sigma$  is equal to the distance separating the molecules when  $U(r) = 0$ . The family parameter,  $m$ , depicts the repulsive part of the interaction. When  $m = 12$ , we have the famous 12-6 or Lennard-Jones potential.

The exponential:6 (Exp:6) family is frequently used. The characteristic family parameter here represents the steepness of the repulsive part of the interaction potential and is given the symbol  $\alpha$ . Other families of interest are the Kihara and the Morse.

Now we have shown that all families are essentially part of the same large family with respect to their relation to experiment. This means that if we can fit a member of a given family to experimental data via a theoretical expression, we can get the same fit from a member of another family. Alternatively, members of different families can be interchanged without materially altering the fit. There is nothing special about any of the families known at this time. They are equivalent.

2) We can *only* make a significant selection of a function and its parameters from experiment if we have data *outside* the reduced range of about

$$2 < T_{12-6}^* < 5 \quad (3)$$

Here  $T_{12-6}^*$  is given by  $T_{12-6}^* = T/(\epsilon/k)_{12-6}$ , where  $(\epsilon/k)_{12-6}$  is the well-depth parameter for the gas from the 12-6 function (5). *Any* function will fit data in the range of Eq. 3. We must have data outside this range if we are to find a function suitable for extrapolation purposes. We give an example for the case of argon. Since  $(\epsilon/k)_{12-6}$  for

argon is about 120°K, the range of Eq. 3 is approximately  $240 < T^\circ\text{K} < 600$ . We have found that all the functions of all the families tested can, via Eq. 1, fit experimental viscosity data in this range. An example has been published (6, figure 2; see also 3, 4). The point we wish to make is that only one member of a family can fit this data *and* data above 600°K. Furthermore, if we fit data above 600°K with a given function, we will automatically fit data in the range  $240 < T^\circ\text{K} < 600$ .

We mention the above two points because our discussion makes use of fits of kinetic theory expressions to experimental data. We feel that the fits given below are satisfactory and that any conclusions are not influenced by our choice of potential functions.

There is a disagreement between two sets of experimental viscosity coefficient data for several gases. The disagreement is depicted for the case of argon in Fig. 1a. One set, open circles, represents data points which are generally accepted as correct and which are *used to compute the tabulations available*. Here we have fitted this set to Eq. 1 with the 40-6 potential function. The references for these points are given in another publication (6), but data above 500°K are mainly from the work of Trautz and Vasilesco (7). The other set, filled circles, are points from the work of Kestin and co-workers (8-10) (300° to 570°K) and from recent work by a group at the Los Alamos Scientific Laboratory, which we designate by G.-M.-W. (11) (1100° to 2100°K). These filled-circle points were also fitted to the 40-6 function. It is very obvious that the discrepancy is striking (12).

The discrepancy between the work of Kestin and the work of others has been known, of course, for some time. Mason also reported several years ago (13) that the high temperature viscosity data of Trautz and Vasilesco could be in error. For tabulation purposes, however, the discrepancy has usually been overlooked, possibly due to the simple fact that Kestin's results are different. In fairness to this negative point of view, it is only recently that high temperature data have become available to supplement Kestin's work. Also, much of Kestin's results lies in the temperature range of Eq. 3. This being the case it is difficult to pin down a correlating function from these data alone (point 2 above) and the discrepancy could have been caused by systematic error. The situation is different when we take Kes-

tin and G.-M.-W. together. One can show by plotting their data on the same graph that their data are compatible and, furthermore, here we have data which are definitely outside the range of Eq. 3 and so we can make positive statements on the correlating potential function (14).

Figure 1b shows the fit of Kestin and the G.-M.-W. data alone. We found that an  $m$  of about 12 or 15 was the best. Eventually we used the Exp:6 function with  $\alpha = 15$ . This is equivalent to an  $m$  of about 13. (Remember, point 1 above, there is absolutely no significance in changing from one family to another. We changed to the Exp:6 family only because we did not have available collision integrals for  $m = 13$  or 14).

We now show that there is no justification for omitting the Kestin (and now G.-M.-W.) work. Indeed, we give substantial evidence that this work is correct.

Part of our analysis is based on the fact that kinetic theory predicts that the thermal conductivity of a dilute monatomic gas,  $\lambda$ , is linearly related to the viscosity by the expression

$$\lambda = f c_v \eta \quad (4)$$

where  $c_v$  is the specific heat per gram at

constant volume and  $f$  is a constant. Theory predicts that this constant is very close to 2.50. Explicitly, the kinetic theory expression for  $\lambda$  is (1):

$$10^7 \lambda = \frac{1989.1 (T/M)^{1/2}}{\sigma^2 \Omega^{(2,2)*} (T^*)}, \quad \text{cal cm}^{-1} \text{ sec}^{-1} \text{ }^\circ\text{K} \quad (5)$$

If one can properly fit experimental viscosity data to Eq. 1, then one must be able to represent experimental thermal conductivity data by Eq. 5 without altering the correlation parameters. We thus determined theoretical thermal conductivity coefficients for argon from Eq. 5 using the function and parameters found suitable for the open circle viscosity data. These values were compared with published experimental values (15-20). Figure 2a shows the deviation curve. This figure clearly indicates that the correlation of  $\lambda$  is not satisfactory, even allowing for the wide scatter of the thermal conductivity data. There is a systematic trend of the data away from a proper fit. This deviation has been observed before but usually has been attributed to  $f$  rather than to errors in  $\eta$  or  $\lambda$ . There is no theoretical evidence that  $f$  (that is, Eqs. 1 and 5) can change by an amount necessary

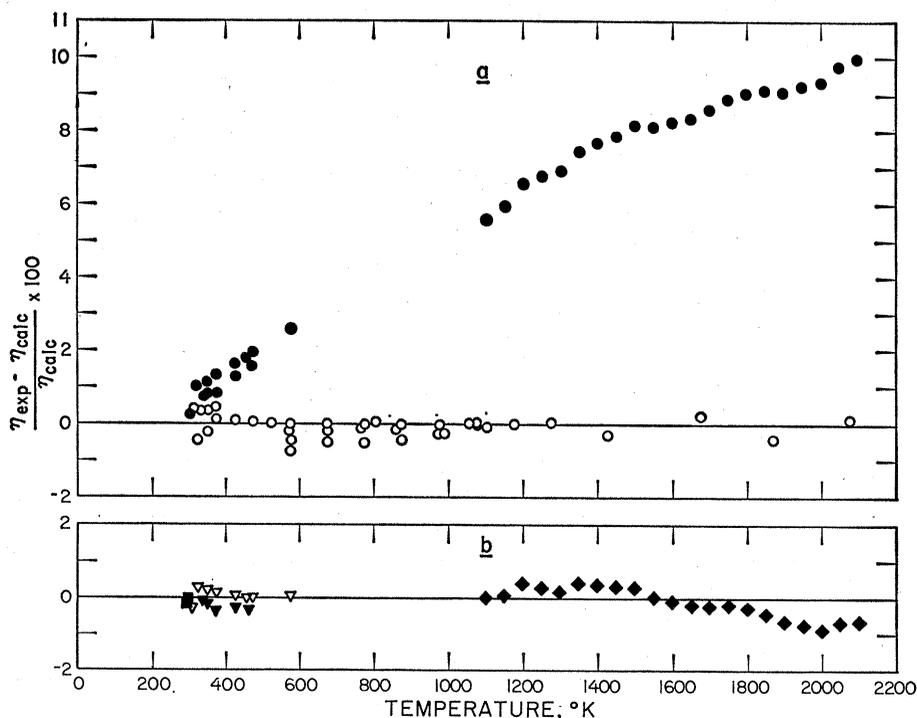


Fig. 1. (a) Fit of experimental viscosity coefficients of dilute argon with Eq. 1, and the 40-6 function with  $\sigma = 3.15 \text{ \AA}$ ,  $e/k = 224.1^\circ\text{K}$ . Open circles represent data of Trautz and Vasilesco (7) and other workers (6). Filled circles represent data of Kestin and co-workers (8-10) and Wageman and co-workers (11). (b) Fit of Kestin and G.-M.-W. argon viscosity data with Eq. 1 and the Exp:6 function with  $\alpha = 15$ ,  $\sigma = 3.68 \text{ \AA}$ ,  $e/k = 156.5^\circ\text{K}$ . (This function is very similar to the 13-6 function.) References:  $\blacksquare$  (8);  $\blacktriangledown$  (9);  $\nabla$  (10);  $\blacklozenge$  (11).

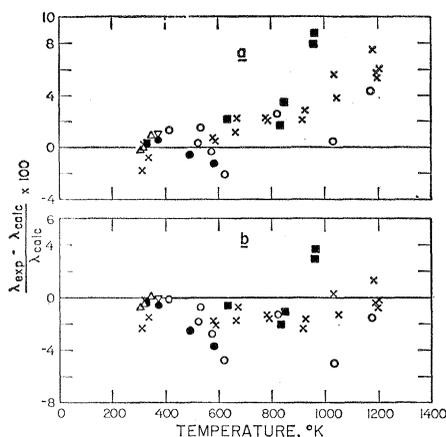


Fig. 2. (a) Correlation of dilute argon thermal conductivity data with parameters suitable for the viscosity correlation of Fig. 1a. References:  $\Delta$  (15);  $\nabla$  (16);  $\circ$  (17);  $\bullet$  (18);  $\blacksquare$  (19);  $\times$  (20). (b) Correlation of dilute argon thermal conductivity with parameters suitable for the viscosity correlation of Fig. 1b. Symbols as in Fig. 2a.

to cause the systematic deviation of Fig. 2a.

Let us see what happens when we fit the thermal conductivity to the Exp:6 function with  $\alpha = 15$ , that is, "fit" on the basis of the Kestin and G.-M.-W. data. The result, given as Fig. 2b, shows a substantial improvement over Fig. 2a (21).

We have another simple, but less exact, procedure which does not involve a potential function. Assuming that the gases obey the principle of corresponding states, a reduced viscosity,  $\eta_R$ , should be a universal function of reduced pressure  $p/p_c$  and reduced temperature  $T/T_c$ . Here  $p_c$  and  $T_c$  are the

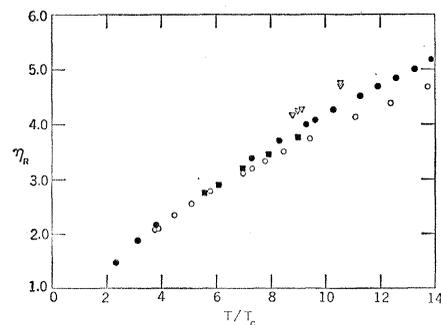


Fig. 3. Plot of the viscosity reduced by critical values, Eq. 6, versus  $T/T_c$ , where  $T_c$  is the critical temperature. The filled circles are reduced argon viscosities from Kestin and G.-M.-W.; open circles are reduced argon viscosities from Trautz and the workers who agree with him. The squares are reduced viscosities of neon, from Hanley and Childs (3). The triangles are viscosities for hydrogen (23). Both the neon and hydrogen data are measurements taken below 400°K.

critical pressure and temperature respectively. The reduced viscosity is then given by

$$\eta_R = \eta_{\text{expt}} M^{-1/2} p_c^{-2/3} (RT_c)^{(1/6)} \quad (6)$$

We know that the principle of corresponding states is not precisely obeyed for the noble gases and is obeyed only approximately for polyatomic gases of simple structure. Nevertheless Eq. 6 and others like it provide a simple method to correlate data. Figure 3 shows a plot of the reduced viscosity of argon from Trautz and the workers who agree with him (open circles), and from Kestin and G.-M.-W. (filled circles). Also included are reduced viscosities of neon (squares) and hydrogen (triangles). These latter viscosities are reduced from measurements taken below 400°K and are assumed not to have the systematic deviation represented by Fig. 1a. We can only expect to see a trend from Fig. 3. This is what we get: no points fall on a single line, but it is apparent that the filled circles are closer to such a line than are the open circles.

From Fig. 2b, and, to a much lesser extent, Fig. 3, the evidence is in favor of the Kestin and G.-M.-W. data. However, we have found that both the 40-6 and 13 (approx.)-6 functions fit the two sets of viscosity data. Can we decide between the two functions from an independent experiment? We think we can. The repulsive part of the intermolecular potential can be estimated from precise and powerful experiments on the elastic scattering of high-energy beams of neutral atoms. Results from these experiments for argon indicate that the repulsive exponent is between 7 and 14 (22). Hence, if we assume that the 40 and 13 exponents of the 40-6 and 13-6 functions respectively are indicative of the true repulsion, then the conclusion from scattering experiments is that one must favor the 13-6 potential and rule out the 40-6 potential.

We have demonstrated that the work of Kestin and G.-M.-W. cannot be excluded from future estimates of the viscosity coefficient of argon above room temperature. We have based our arguments on argon because (i) Eqs. 1 and 4 are valid only for monatomic gases, (ii) thermal conductivity data for the other monatomic gases are not available in the required temperature range to allow us to repeat Figs. 2a and 2b, and (iii) the elastic scattering of the argon-argon system has been particularly well studied.

Our conclusion is not restricted to

argon, however. Trautz and his co-workers have had a tremendous influence on high-temperature viscosity estimates for many gases. If he (and other authors who agree with him) is incorrect for argon, he is possibly incorrect for the other gases. We mention, in this respect, that Kestin and co-workers report a disagreement between their results and Trautz's and other workers' for several gases (10) (for example,  $\text{CO}_2$ ,  $\text{N}_2$ , Ne,  $\text{H}_2$ , He, Kr, air). The Los Alamos group (11), as well, report results for hydrogen, helium, and nitrogen which differ from the previously accepted values, but which seem to agree with Kestin's results.

Hence we believe that the dilute gas viscosity coefficients above room temperature for all simple gases (Ar, Ne, Kr, He,  $\text{H}_2$ ,  $\text{O}_2$ ,  $\text{N}_2$  and so forth) should be reexamined both from the experimenter's and correlator's points of view. Not only are new experiments clearly required, but an analysis of the old experiments, with an eye to discover possible overlooked corrections, is necessary.

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precision. We have not done this; our only purpose was to check the thermal conductivity on the basis of a fit to viscosity which was not prejudiced by other considerations. We stress that any adjustment would involve a small change in  $\epsilon/k$  or  $\sigma$ , not in  $\alpha$  or  $m$ .

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## Lateral Hypothalamus: Food Current Intensity in Maintaining Self-Stimulation of Hunger

*Abstract. Rats displaying stimulus-bound eating will press bars for currents slightly above eating threshold only when food is near the bar. At higher currents self-stimulation is maintained without food. Such currents may spread to activate consummatory feedback appropriate to the drive elicited; or, for more intensely stimulated drive mechanisms, wider ranges of sensory feedback may be reinforcing.*

In this study we examined the possibility that reinforcement in self-stimulation is dependent upon sensory feedback appropriate to a drive that is electrically elicited. The examination bears upon the proposition (1) that electrical intracranial stimulation is self-administered by an animal only if both a drive system and a reinforcement system are activated jointly. In the usual situation, activation is presumably accomplished by the current spreading to both systems in the brain. However, even if the current is somehow regulated to activate only one system, self-stimulation should still occur provided the other system is concurrently activated in some other way. For example, an animal may not self-administer current that stimulates a drive only, but, on the other hand, it may self-administer current if the reinforcement system is operated at the same time in some normal fashion, such as by making available sensory stimuli appropriate to the consummation of the drive.

To test this possibility, male albino rats, with permanently implanted monopolar electrodes (2, 3), were allowed the opportunity of self-stimulating a known hunger-drive mechanism in the lateral hypothalamus (4, 5) at current intensities just above the thresholds for eliciting stimulus-bound eating and yet, presumably, too weak to affect a reinforcement system also. The following questions were then asked: (i) Would

a thoroughly satiated rat repeatedly press a bar to deliver such a current if food were absent? (ii) If the animal would not press, would it do so if food were continuously available immediately by the bar?

Three prerequisites were necessary in the selection of subjects. First, they should display clear stimulus-bound eating of wet mash to electrical stimulation of the lateral hypothalamus (ESLH). [Screening procedures (4, 5) and threshold measurements (6) for stimulus-bound eating have been described.] Second, at currents well above eating threshold, they should press a bar at moderate rates to self-administer 2-second trains of ESLH delivered through the same electrode that elicits eating. Third, if a dish of wet mash were by the bar, they should readily eat in response to each self-delivery of the stimulation. In our study, seven rats met the first criterion and these also met the latter two criteria. This high coincidence of stimulus-bound eating and self-stimulation by way of the same electrode has been noted before (7).

Two experiments comprised the study. The second, a modification of the first, was to control for some possible procedural artifacts. Accordingly, experiment 1 and only those features of experiment 2 that were different will be described.

The slightly suprathreshold current employed for each of four animals used

in experiment 1 is indicated in the upper right of Fig. 1a by an open circle below the bar showing the eating threshold. At that current each animal's bar-pressing was measured as a joint function of whether bar-pressing delivered stimulation and whether food was by the bar. At least 20 minutes before all tests, animals were satiated by putting a dish of wet mash in their home cage. Four conditions were presented to each rat in the following order: (i) stimulation plus no food (S-NF); (ii) stimulation plus food (S-F); (iii) no stimulation plus no food (NS-NF); and (iv) no stimulation plus food (NS-F). Seven blocks of these four conditions comprised each animal's test, and each condition lasted 2 minutes. The order of conditions was chosen to keep the initially high extinction rates that might follow the S-F condition from falsely elevating the S-NF or NS-F rates.

Tests were administered in two Plexiglas boxes that were identical except that one always contained a dish of wet mash by the bar and the other did not. The animal and the manipulandum were rotated from box to box every 2 minutes, and the animal was placed in the middle of the box facing the bar. On every other trial within a given box, pressing the bar did not deliver ESLH.

In experiment 1, if an animal failed to press within 10 seconds after being placed in a box, it was reminded of what a lever press on that trial would deliver by being primed with a free sample (2 seconds) of such stimulation. If the animal still did not press, this priming was repeated up to twice more on the 15th and 20th second of the trial. Thus all priming occurred within the first quarter of each 2-minute period and without regard to the animal's orientation in the box. Priming in the no-stimulation periods (NS-NF and NS-F) consisted of the noise of the stimulator, but no current was passed.

New rats were employed for experiment 2. The slightly suprathreshold current used for each is shown in the upper right of Fig. 1b by an open circle below the bar indicating the eating threshold. The design was the same as for experiment 1 with the following exceptions: Each press delivered 3 seconds of stimulation instead of 2, and the response measure was the number of reinforcements obtained rather than the absolute number of presses. Eight instead of seven blocks of four conditions were used. The order of the four