

Fig. 3. Trace of autocorrelograph shown in Fig. 2c.

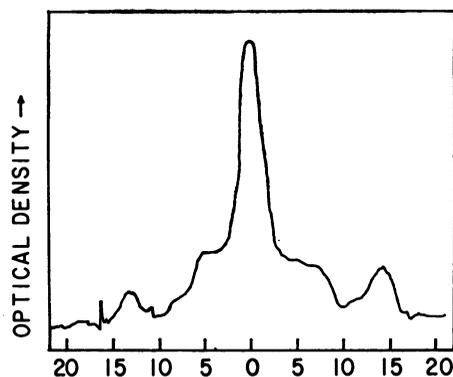


Fig. 4. Trace of autocorrelograph obtained with polyuridylic acid stained with  $Tl_2CO_3$ .

cause of the noise. To do this we examined the optical autocorrelation functions of images of the strands.

Formally an autocorrelation function of a two-dimensional transmission function  $T(x,y)$  is

$$\Phi(\xi,\eta) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} T(x,y)T(x+\xi,y+\eta) dx dy$$

Physically it can be considered a measure of the similarity of two patterns, one obtained from the other by a displacement

$$\begin{matrix} \rightarrow & \rightarrow \\ \xi i & + \eta j \end{matrix}$$

without rotation. The measure is the average value of the product of the transmissions of the displaced and undisplaced pattern. Clearly the autocorrelation of a periodic pattern will peak at displacements corresponding to periodicities in the original pattern. Even if the transmission function is the sum of a periodic and a nonperiodic contribution, the autocorrelation function will have these characteristic peaks, but with lower contrast. This is the basis of the present application, in which autocorrelation functions of noisy micrographs were examined for evidence of underlying periodicity.

The autocorrelation of photographic plates is readily obtained by measurement of the transmission of light through two identical copies, as a func-

tion of the displacement of one relative to the other, since the transmitted intensity is proportional to the product of their respective transmissions. Here it is obtained by an elegant reported procedure (6) in which the plate to be analyzed (Fig. 1, A) and an identical one (B), at  $1/M$  times the magnification, are placed parallel to each other a distance  $a$  apart. An unexposed photographic emulsion (C) is placed parallel to the others and a distance  $b$  from B on the side away from A, where  $(a + b) = bM$ . The intensity at the unexposed plate of diffuse light, transmitted first through A and then through B, is directly proportional to the two-dimensional autocorrelation function of the photograph.

Figure 2a is a portion of an electron micrograph of polyuridylic acid deposited from a solution of the  $Tl_2HgHDA(OH)$ . The contrast of the granularity of the background is high, and the stained polynucleotide chain (Fig. 2a, arrows) is not easily detected. However, the autocorrelograph (Fig. 2d) clearly shows peaks at displacements of about 8 Å in a direction parallel to the chosen strand. It is easily demonstrated that the spacing is associated with the chosen strand, since the peak is lost when the strand is masked on both transparencies (Fig. 2e). When all of the plate is blocked out except for a narrow strip containing the polynucleotide chain, the autocorrelograph shows remarkably clear peaks, regularly spaced (Fig. 2c); a densitometer trace indicates that the spacing is 8 Å (Fig. 3).

The autocorrelograph of an equivalent area adjacent and parallel to the strand showed no frequent spacing; nor did micrographs of grids streaked on  $Tl_2HgHDA(OH)$  without polynucleotide. Strands of polyuridylic acid, deposited in the presence of  $10^{-2}M Tl_2CO_3$  buffer, gave autocorrelation functions that though less distinct had nearly the same periodicity of 7 to 8 Å (Fig. 4).

Thus it appears that polyuridylic acid stained with  $Tl_2HgHDA$ , or simply as the Tl salt, when deposited by our procedures exhibits a structural periodicity of about 8 Å. This spacing probably corresponds to the internucleotide distance in the extended polynucleotide chain. In agreement with these results, recent results with yeast alanine transfer-RNA also point to an 8-Å internucleotide spacing in extended polynucleotide chains (7).

Finally, molecular models indicate that 8 Å is about the maximum possible

internucleotide spacing; the fact that this is also the observed average spacing implies that the strands are fully extended.

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#### Gallstone of Unusual Composition: Calcite, Aragonite, and Vaterite

Abstract. A gallstone of almost perfect octahedral symmetry was composed of a mixture of crystallites of the three polymorphous forms of calcium carbonate: calcite, aragonite, and vaterite.

The crystalline material present in an unusual gallstone (1) was determined by sampling different regions of the stone and identifying the substances by the

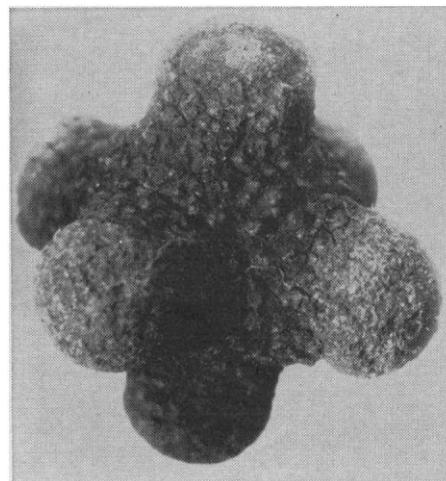


Fig. 1. The gallstone; the greatest dimension is 2 cm.

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