

The laminar flow of suspensions in tubes

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substantially uniform density over an area of several square millimetres can be obtained. In this connexion it is important to note a difference in the "geometry" of radiation measurement by a counter and autoradiography. The volume of the ionizable gas in a Geiger-Müller tube and the distance between the tube and the sample ensure that the counting rate is proportional to the total radioactivity integrated over the whole area of the sample. On the other hand, the thin sensitive layer and close contact normally used for autoradiography mean that the film records the point-to-point distribution of radioactivity over the area of the specimen. Straightforward autoradiography will, therefore, only give the desired uniformity of density when the specimen itself is uniform. More uniform density from uneven specimens could, however, be obtained by increasing the distance between the film and the source. If a number of sources were measured simultaneously in this way it might be neces-

sary to place a metal plate with suitably positioned apertures between the specimens and the film, to reduce the extent to which the density opposite one source is influenced by "cross-fire" from adjacent sources.

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The laminar flow of suspensions in tubes

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The conception of viscosity as action per unit volume is used in the analysis of the flow of a suspension or macromolecular solution in a tube. The flow mechanism adduced involves the postulation of molecular vortices of which the suspended particles or molecules form the nuclei. This mechanism is used as a basis for the derivation of a relationship between the specific viscosity of the suspension and the linear dimensions of the vortices. The laws of Einstein, Staudinger, Huggins and their co-workers are derived simply and as special cases of this relationship. The law relating viscosity with concentration is also derived with reference to the same mechanism. This law assumes the form $\eta_{sp} = an + bn^2$ where a and b are constants and η_{sp} and n are respectively the specific viscosity and molecular concentration of the solution. It is shown to be in good agreement with typical experimental results. The effect of the aspect ratio of the suspended particles or molecules is also considered and it is shown that the longest particle dimension rotates in a plane normal to the vortex axis. Although no quantitative relationship between viscosity and velocity gradient has been derived the mechanism is observed to lead to results in qualitative agreement with the experimental data.

1. INTRODUCTION

The very extensive use of capillary viscometers in investigations into the physical characteristics of suspensions, and particularly in the indirect determination of macromolecular weights, calls for a closer study of the mechanics of flow for fluids of this kind than has apparently been attempted hitherto. Although an enormous number of so-called viscosity measurements have been made on such fluids there seem to have been but few attempts to interpret these in terms of any clearly-conceived flow model. Many empirical relationships have been devised but these have in the main yielded no significant ideas relevant to the processes involved.

The well-known equations of Einstein, Staudinger and Huggins^(1,2,3) represent perhaps the most successful attempts so far to investigate theoretically the relation between the apparent viscosity of a solution and the physical constants of the solvent and of the particles of solute dispersed in it. Although these laws show good agreement with experiment in certain cases, they have limited applicability and provide no satisfactory basis for correlating viscosity and concentration where the relation between these is known to be non-linear.

In the present paper an attempt is made to analyse the mechanical processes of flow of suspensions and macro-

molecular solutions of non-electrolytes through tubes. The evidence derived from this analysis is employed in the construction of a flow mechanism and this is in turn used in the derivation of fundamental viscosity relationships. These are compared with the corresponding experimental relationships.

2. THE NATURE OF VISCOSITY IN SUSPENSIONS

Consideration of the dimensions of viscosity, as defined by Newton's equation, shows it to be a measure of the action per unit volume within a fluid. In the case of a homogeneous fluid moving through a tube under laminar flow conditions the action results from the relative motion in translation of adjacent parts of the fluid. In these circumstances a fluid can be regarded as having a continuous structure, and, for the purposes of analysis, can be imagined as being made up of elementary cylinders of liquid each subject to viscous forces resulting from the translational motion relative to it of neighbouring elements. The integration of the equation representing the dynamic equilibrium of such an elementary cylinder under the opposing actions of the force due to the applied pressure and the viscous resisting force leads to the familiar expression for the velocity along a streamline and thence to Poiseuille's formula. This approach, however,

breaks down in the case of an inhomogeneous fluid, for here the action arises not only through the relative motion in translation of adjacent parts of the continuous phase, but also to the motion of the solute molecules, or the particles, within it.

It has been shown that, in general, the motion of a viscous liquid cannot be irrotational,⁽⁴⁾ and consideration of the variation of velocity gradient within a tube, and hence of the forces acting on a suspended particle, suggests that under these circumstances rotation of the particle certainly occurs.

The total action involved in the flow of an inhomogeneous fluid is thus seen to consist in general of three components. These are respectively, the action arising through the relative motion in translation of adjacent parts of the continuous phase, that due to the rotation within it of the macromolecules or other inhomogeneities and the action resulting from the translation of these suspended particles relative to the continuous phase. This last component, as will be shown later, is small compared with the other two except at fairly high concentrations.

3. FLOW MECHANISM

The observations made in the last section lead directly to the conception of the flow mechanism in the cases under consideration as involving the translation of the solvent and the simultaneous translation and rotation of the particles of solute within it.

The suggested rotation of solute particles will obviously involve the movement with them of some entrained solvent and for this reason the proposed flow mechanism must be modified slightly so as to involve the translation through the tube of what may be called "macromolecular vortices" rather than of isolated particles.

Let us now consider some of the implications of this proposed flow mechanism, and, in the first instance, let us consider the flow of a suspension involving only a single solute particle.

If η_s denotes the coefficient of apparent viscosity of the solution (that is, the volume-average action per unit volume) and η the coefficient of viscosity of the solvent, the total action, A_1 , within the tube due to the single vortex is given by:

$$A_1 = \pi R^2 l (\eta_s - \eta) \quad (1)$$

where R and l are respectively the radius and length of the tube. It should be noted that, since action per unit volume varies from point to point, it is necessary to deal with the total action within the tube and the total internal volume of the tube as a means of arriving at any relation between measured values of viscosity and the constants of the solute and solvent.

Regarding the vortex as an "ideal" vortex of radius a and length h , two extreme cases can readily be distinguished, e.g. one in which a is negligible compared with h ,—a rod-like vortex,—and the other in which a is large compared with h ,—a disk-like vortex.

(i) *h much greater than a.* In this case the total action due to the rotation of the vortex will arise almost wholly as a result of the viscous forces on its curved surface and the resisting couple G acting on it will be given by:

$$G = 4\pi\eta ha^2\omega \text{ (see Ref. 5)} \quad (2)$$

where ω is its angular velocity.

As an isolated vortex no forces tending to produce translation relative to the solvent will act on it.

$$\text{Hence } A_1 = 4\pi\eta ha^2\omega^2 l^2 / v^2 \quad (3)$$

where v is the linear velocity of the solvent on the streamline along which the vortex moves.

The factors appearing on the right-hand side of equation (3) can be grouped and equation (3) rewritten as

$$A_1 = k_1 \eta F \quad (4)$$

where k_1 is a "flow" factor and F a "form" factor of the vortex, i.e. $F = 4\pi ha^2$. (F is the viscous resisting couple per unit angular velocity in a fluid of unit viscosity.)

Assuming that Poiseuille's law holds in this case $v = P(R^2 - r^2)/4\eta l$ and hence, in accordance with the requirement of least action, $\omega = dv/dr$, i.e. $\omega = Pr/2\eta l$, where r is the radius of the streamline, P being the applied pressure difference.

$$\text{Therefore, } k_1 = 4l^2 r^2 / (R^2 - r^2)^2$$

Again, for least action A_1 must have its least value and therefore the vortex must flow along a streamline adjacent to the tube axis. Assuming a fixed value of r , k_1 is seen to be constant.

For very dilute solutions in which it can be assumed that the motion of any one vortex is unaffected by neighbouring vortices, the total action within a tube A , when the number of particles per unit volume in the solution is n , is $n\pi R^2 l$ times as great, assuming all vortices to be identical.

$$\text{i.e. } A = nA_1 \pi R^2 l \quad (5)$$

(In view of the "normal" particle size distribution usually assumed, the present analysis can only be regarded as applicable to "average" particles.) Now $\eta_s - \eta = A/\pi R^2 l$ in the present case.

$$\text{Therefore } \eta_s - \eta = k_1 n \eta F \quad (6)$$

from equation (4).

In words, the specific viscosity of a very dilute suspension (that is, the fractional change in the viscosity of the solvent due to the suspension in it of the solute) is directly proportional to the number of suspended particles per unit volume provided that the "form" factor of the molecular vortices remains constant.

(ii) *a much greater than h.* In this case the total action due to the rotation of the vortex arises almost wholly as a result of the viscous forces acting on the plane vortex surfaces. Here $G = (32/3)\eta a^3\omega$,⁽⁶⁾ and the "form" factor is in this case given by $F = (32/3)a^3$.

Subject only to the different value of F in this case equation (6) can be shown, as previously, to hold.

(iii) *h and a of the same order of magnitude.* Subject again only to a further different value of the form factor, equation (6) can be shown similarly to hold in this case also.

4. THE LAWS OF EINSTEIN, STAUDINGER AND HUGGINS

Empirical equations based on the analyses of Einstein, Staudinger and others are commonly used in the determination of molecular weights from viscosity measurements. It is of interest, therefore, to consider the significance of such equations in the light of the flow mechanism postulated in the last section.

Previous workers have made no hypothesis in regard to the existence of molecular vortices but have dealt with the suspended particles themselves and have variously assumed that they are spherical (Einstein),⁽¹⁾ cylindrical (Staudinger and Hever)⁽²⁾ or rod-like (Huggins).⁽³⁾

By replacing the molecular vortices assumed in the present analysis by particles having the shapes referred to, these laws can be derived very simply.

Thus, assuming the particles to be spherical

$$G = 8\pi\eta a^3\omega \text{ (see Ref. 7)}$$

and therefore, $F = 8\pi a^3$.

Now the total volume of the spheres in unit of volume of the suspension, ϕ , is given by $\phi = n(4/3)\pi a^3$, and therefore, from equation (6),

$$\eta_s - \eta = k_2\phi\eta \quad (7)$$

where k_2 is a constant.

The specific viscosity is therefore directly proportional to the volume-concentration as shown by Einstein.

Next, let us assume that the particles are cylindrical with lengths great compared with their radii, and rotate about their cylindrical axes. Here, as in Section 3(i), F has the value $4\pi a^2h$ where a is now the radius of the cylindrical particle and h its length.

For particles of constant section the mass M of the particle is proportional to h .

The mass-concentration, c , of the solution is nM and, from equation (6), therefore

$$\eta_s - \eta = k_3\phi c \quad (8)$$

where k_3 is a constant.

That is, the specific viscosity is directly proportional to the mass-concentration and independent of the particle size, as shown by Staudinger and Hever for solutions of mono-saccharides and disaccharides.

Finally, let us assume that the particles are rod-like and rotate about axes normal to their lengths. Here, the molecules, together with the entrained solvent must be regarded as comprising disk-like particles. The molecular weight is therefore proportional to the diameter of the disk.

As in Section 3(ii), $F = (32/3)a^3$.

But $a^3 = k_4M^3$, where M is the molecular weight and k_4 a constant. Therefore $F = k_5M^3$, k_5 being constant.

If c is the mass-concentration of the solution, from equation (6), we have

$$\eta_s - \eta = k_6\eta cM^2 \quad (9)$$

k_6 being constant.

This agrees with the results of Huggins and other workers, who showed that the intrinsic viscosity (that is, the limiting value as the concentration approaches zero of the specific viscosity per unit concentration) is proportional to the square of the molecular weight.

The applicability of equations (8) and (9) in certain limited cases has led to the general adoption of an empirical molecular weight equation of the form $\eta_{int} = kM^\alpha$ where η_{int} is the intrinsic viscosity and k and α are constants. In the light of equation (6) it would appear that α is a shape factor for the suspended particles. Its value, as seen from the two extreme cases cited in Sections 3(i) and 3(ii), lies somewhere between 0 and 2.

5. THE VISCOSITY-CONCENTRATION LAW

The mechanisms considered to date all lead to a linear relationship between viscosity and concentration. This relationship rests on the assumption that individual molecular vortices or the individual particles themselves are independent of their neighbours; it must therefore be regarded as only an approximation and an approximation moreover which becomes progressively less valid as concentration is increased.

Departure from such a linear relationship is, furthermore, clearly indicated by the relevant experimental data; this confirms the validity of the straight-line law only as an approximation in the case of very dilute solutions.

In view of this limitation of current theory it becomes important to consider whether the proposed molecular vortex theory can be extended so as to take account of the conditions existing in more concentrated solutions.

The flow mechanism outlined in Section 3 leads, in accordance with the principle of least action, to the conclusion that an individual particle flowing through a tube would follow a path adjacent to the tube axis. The stability of the vortices associated with a succession of such particles, however, requires the formation about the tube axis of a "Kármán vortex street."⁽⁸⁾ Let us then consider some of the main implications of this requirement in the present case.

According to the theory of vortex streets each vortex has a translational velocity, U , impressed on it as a result of the combined action of all other vortices, where

$$U = K/2\sqrt{(2)}L \quad (10)$$

K being the vortex strength and L the distance measured in the direction of flow between the vortices in either "lane."⁽⁹⁾ By definition the vortex strength, K , is given by the equation

$$K = 2\pi a^2\omega \quad (11)$$

where a is the radius of the "ideal" vortex and ω its angular velocity.

Since, in the present case, a and ω are both small the molecular vortices postulated must in general be "weak" vortices. Even at moderate concentrations of the solutions, then, U will be small, but will increase as concentration increases under given external conditions.

An approximate equation relating specific viscosity and concentration, applicable at moderate and even fairly high concentrations, can now be derived.

Let n be the number of solute molecules per unit volume of solution. The number emerging from both lanes in the vortex street per second is therefore $2(v + U)/L$.

$$\text{Therefore, } n = 2(v + U)/LV \quad (12)$$

where V is the volume of solution discharged per second from the tube. Neglecting U compared with v , and making use of the approximate constancy of v/V , we have,

$$n = k_7/L \quad (13)$$

$$\text{or, from equation (10), } n = k_8U \quad (14)$$

In words, the relative velocity of the solute with respect to the solvent increases approximately linearly with concentration. The action, A_T , involved as a result of this relative translation, since the resisting force is proportional to ηU , is given by

$$A_T = k_9\eta n^2 \quad (15)$$

Since the action A_R , due to rotation, as previously shown, is given by

$$A_R = k_{10}\eta n \quad (16)$$

the total action, A , is

$$A = \eta(k_{10}n + k_9n^2) \quad (17)$$

k_9 and k_{10} being constants.

That is, the specific viscosity of a solution is related parabolically with its concentration.

As a means of testing equation (17) by experiment the typical results of Kendall and Monroe⁽¹⁰⁾ have been used. In the first and third columns of the table the corresponding values obtained by them for the molecular concentration

and the viscosity of a solution of naphthalene in benzene at 25° C are quoted. The values of (concentration)² and specific viscosity given respectively in the second and fourth columns were calculated directly from these. Using the co-ordinates of two points on the specific viscosity-concentration graph the constants k_9 and k_{10} of equation (17) were evaluated. The specific viscosity-concentration law in the present case was hence found to be

$$\eta_{sp} = 1.60.10^{-2}n + 1.08.10^{-4}n^2 \quad (18)$$

The values of the specific viscosity obtained by substituting the values of n and n^2 , given in the first two columns, in equation (18), are recorded in the fifth column, the percentage discrepancies between these calculated values and the experimental values of column four being shown in the last column.

Corresponding experimental and theoretical values of specific viscosity at various concentrations

n	n^2	η_{sa}	Experimental η_{sp}	Calculated η_{sp}	Discrepancy %
0.00	0.0	0.6048	0.000	0.000	0.0
5.10	26.0	0.6565	0.085	0.085	0.0
11.21	125.7	0.7261	0.200	0.193	3.5
15.38	237.2	0.7707	0.274	0.272	0.7
19.29	372.5	0.8263	0.366	0.349	4.6
23.98	576.0	0.8764	0.449	0.446	0.6
26.93	723.6	0.9178	0.517	0.508	1.7

6. EFFECT OF ASPECT RATIO

In the derivation of equation (6) in Section 3 two extreme cases were distinguished, namely (i) that of the rod-like vortex in which the length is big compared with the radius and (ii) that of the disk-like vortex in which the radius is large compared with the length. Equation (6) was shown to be equally applicable to both, though it was noted that the "form" factor differed in the two cases.

In view of the considerable use made of viscosity measurements in the study of high polymers it is of interest to consider whether macromolecules in which one dimension is large compared with the others would lead under the flow conditions under discussion to the formation of rod-like or disk-like vortices. That is, would macromolecules of large aspect ratio κ/d , κ being the longest and d the shortest dimension, orientate themselves so that κ is parallel to or perpendicular to the vortex axis?

Making the assumption that under the circumstances visualized entraining of the solvent by the solute molecules occurs only to a limited extent, the dimensions κ and d can be identified approximately and respectively with h and $2a$ in the former case and with $2a$ and h in the latter.

The ratios of the actions involved in the two cases is the same as the ratio of the form factors and therefore equal to $3\pi h/8a$. Since κ is much greater than d , the action involved when the molecule is orientated parallel to the axis of rotation is big compared with that for perpendicular rotation. In accordance with the principle of least action, therefore, the molecule will rotate so that its longest dimension is normal to the axis of rotation, the molecular vortex consequently being disk-like.

This conclusion is consistent with the observations of Binder.⁽¹¹⁾ He investigated the relationship between the orientations of sections of human hair and their aspect ratios and found that for very large values of the latter the specimens orientated themselves in the perpendicular position.

In the light of the above it becomes apparent that the direct association of specific or intrinsic viscosity with molecular weight, M , in the manner suggested by the equations of Staudinger and Huggins and their co-workers may be misleading.

The index of M in an empirical formula of the type $\eta_{int} = kM^x$ must clearly depend upon the form factor of the vortices and hence on the aspect ratio of the molecules themselves. The index would, however, appear to provide an indirect measure of the shape, degree of branching, etc., of molecules of given molecular weight.

7. GENERAL OBSERVATIONS

The mechanism outlined in the previous sections involves the motion of vortices along streamlines adjacent to the tube axis, as shown in Section 3. The action per unit volume is hence greatest in those regions where the velocity gradient is least; that is, the coefficient of viscosity decreases with increasing velocity gradient. This qualitative result is consistent with the variation found experimentally and investigated quantitatively by Tyler and Richardson,⁽¹²⁾ and hence, indirectly, with the well-established fact that the volume of solution discharged per second increases with applied pressure to a value in excess of that to which simple proportionality would lead.

Further work is necessary to reduce to quantitative terms these and other qualitative relationships suggested by the analysis.

8. CONCLUSIONS

The flow mechanism described follows directly from the conception of viscosity as action per unit volume. It leads simply to the laws of Einstein, Staudinger and Huggins and their co-workers as special cases of a general law which is derived. The quadratic law connecting specific viscosity with concentration, which is also derived, is shown to be in good agreement with typical experimental results. The mechanism is observed to be qualitatively consistent with other experimental data.

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