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Anomalous Flow of Cellulose Solutions During Spinning

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Synopsis

An apparatus is described for measuring the flow rate of cellulose xanthate solutions at known pressure gradients across spinning jets. It is shown that the jet hole behaves more like a capillary than an orifice in its effect on flow and that similar relationships exist for flow through spinning jet holes and a narrow pipe characterized by an equation of the form $Q = Ap^b$, where b has a value of approximately 1.5 for the high rates of shear strain found in spinning jets and 1.4 for the lower rates found in the pipe. A fundamental approach shows a similar discontinuity when comparing the relation between apparent viscosity coefficient and rate of shear strain, between low and high rates of shear. Experimental results are given for the effect of certain chemical factors in viscose solutions and the effect of spinning jet hole size, on the flow rate/pressure gradient curve.

INTRODUCTION

These measurements were made mainly to provide data on the different flow characteristics of a number of viscose solutions and to find the effect of various conditions of shear. The data obtained have also been used to test the validity of certain fundamental studies at low rates of shear and to note whether they can be applied to the high rates of shear present in spinning jets.

APPARATUS

The apparatus is shown, in section, in Figure 1. The parts denoted as A, B, C, D, and E are made of ebonite. The remainder is of glass, except for the spinning jet itself and two steel tubes sealed into E, which take rubber pressure tubing leading to a compressed air supply and a manometer respectively.

A is a cap, which secures the glass reservoir to the body B of the main chamber, a good seal being obtained by means of a soft rubber gasket. The spinning jet is secured in the chamber C by means of the plug D, a good seal being obtained by means of a polyvinyl chloride gasket. This part is then screwed into B, a textile-reinforced rubber gasket being used to obtain a good seal. The cap E screws into the top of B, also with a rubber gasket. The pressure supply is connected through a suitable two-way cock. The apparatus was used at constant temperature conditions,

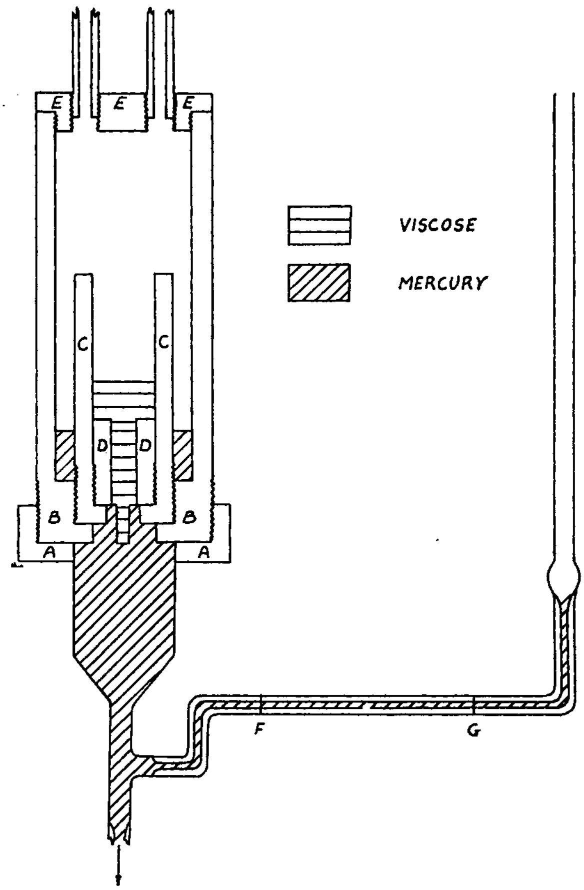

Fig. 1. Apparatus for measuring rates of flow.

obtained by immersing the whole apparatus in a water bath, the temperature of which was controlled to ±0.1 °C.

EXPERIMENTAL TECHNIQUE

The spinning jet was sealed into subchamber *C* as described above, and the subchamber half-filled with the sample to be studied, care being taken that no air was trapped below the surface. With the body *B* secured to the glass reservoir and capillary, the latter was carefully filled with mercury, which, of course, passed along the capillary to the right-hand tube. The mercury level was allowed to rise to a point such that when the subchamber *C* was screwed into *B*, mercury was displaced upward about *C* and a good air seal obtained.

Mercury was then run off through the drain tap, until the thread in the capillary reached the initial flow mark *F*. The cap *E* was then screwed on and the apparatus immersed in the water bath and allowed to reach thermal equilibrium, when mercury was again run off until the thread reached the point *F*. The pressure connections were then applied to *E*, and pressure applied to note that the mercury was running freely along the capillary. Mercury was again drained through the tap until the thread was at *F*, and the apparatus was then ready for an experimental run.

A number of pressures were applied and on each occasion the time noted for the mercury meniscus to move from *F* to *G*, the air pressure being read from the manometer.

Between tests on the various samples, subchamber *C* was removed and the jet taken out. The latter was thoroughly cleaned with water and then placed in chromic acid at 60°C. for 30 min. It was then dried and inspected under a microscope before further use. The subchamber *C* was also cleaned, and excess viscose was removed from the surface of the mercury in the reservoir.

The assembly of the apparatus was accomplished by specially-shaped keys which fitted into slots at the top of *C* and *D*. The bore of the capillary was found by measuring the weight of a thread of mercury placed in the capillary and observing its length. The distance between the two marks *F* and *G* was found by means of a traveling microscope.

EXPERIMENTS

1. Effective Length of a Jet Hole

In these experiments both five-hole and single-hole spinning jets were used. In all cases, the shape of the holes resembles that of a cylinder surmounted by a truncated cone so that an effective capillary length had to be obtained. To test whether the jet holes did behave as effective capil-

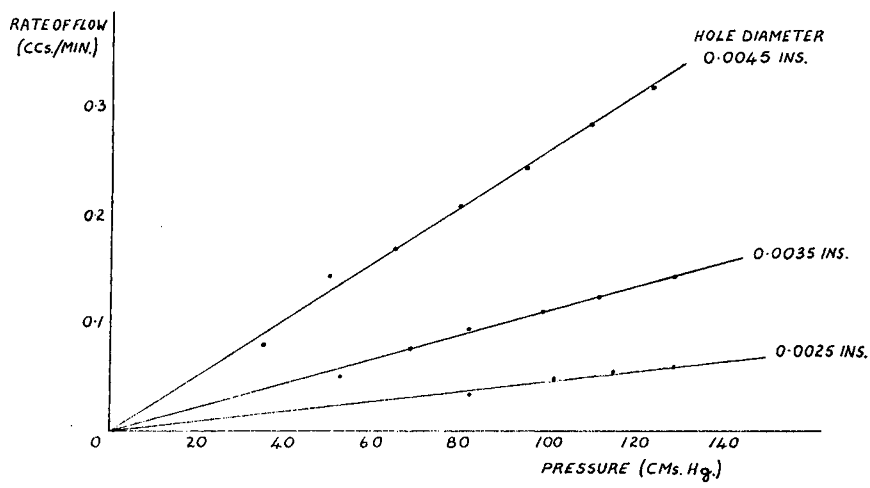

Fig. 2. Flow of glycerol through single-hole spinning jets at 25°C.

laries, measurements were made with glycerol at a number of jet hole sizes. The results, showing a linear relationship between flow rate and pressure are given in Figure 2.

These results were then used to find the effective capillary length L of the jet holes, assuming that the effective radius was that given by the nominal diameter of the jet holes, and using Poiseuille's law. Strictly, of course, since the jet hole radius was not constant along its length, only the ratio L/r^4 should be calculated. The calculated values of L are given in Table I.

TABLE I
Effective Capillary Length of Various Spinning Jet Holes

Jet hole size	5 Holes 2.5 thou.	1 Hole 2.5 thou.	1 Hole 3.5 thou.	1 Hole 4.5 thou.
L., thou	5.25	4.50	7.35	8.46

In general, the effective capillary length is about twice the jet hole diameter.

2. Effect of Salt Figure and Cellulose Content

A number of viscose solutions were examined to find whether their flow characteristics were related to salt figure (a measurement of the character of the xanthate obtained by precipitation of the cellulose) and cellulose content. The viscoses were produced from the same basic raw materials.

The rates of flow for four of these samples, characterized in Table II, have been plotted against the pressure across a five-hole jet of hole size 2.5 thou. in Figure 3. Particulars of the salt figures, cellulose contents, and viscosities measured by a falling sphere method are also given in Table II.

TABLE II

Sample no.	Relative viscosity	Salt figure	Cellulose content, %
11	100	9.4	7.38
12	111.2	8.7	7.60
13	100.9	8.8	7.42
14	115.9	8.7	7.36

Comparing viscose solutions of similar static viscosities and salt figures, it may be seen that the rate of flow is higher for a given pressure when the cellulose content and presumably molecular entanglement is lower (see samples 12 and 14); in this particular case, it is to be noted that the sample with the lower cellulose content also has the higher static viscosity, thus re-emphasizing the effect of the lower cellulose content. Comparing samples of similar static viscosity and cellulose content (samples 11 and 13),

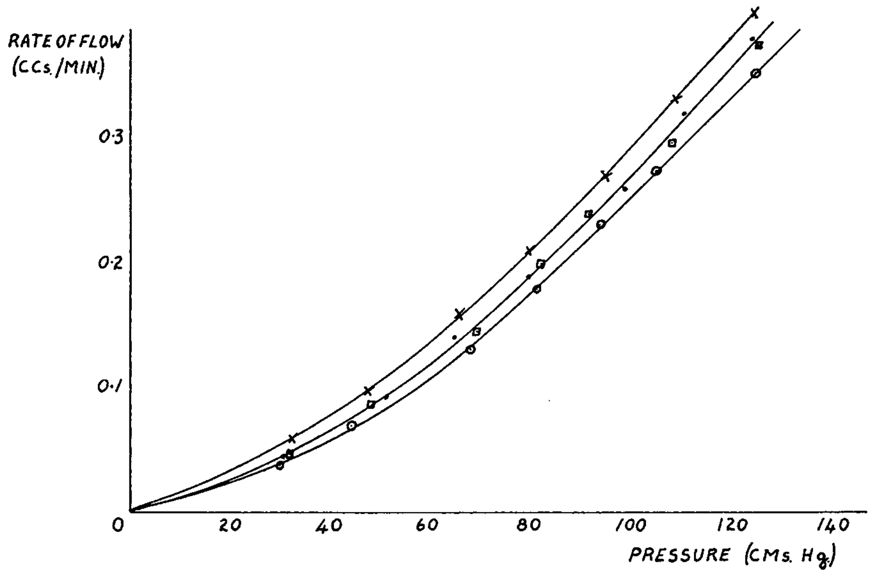

Fig. 3. Flow of viscose solutions through five-hole spinning jet (hole diameter 0.0025 in.) at 25°C.: (●) sample 11, (O) sample 12; (×) sample 13; (□) sample 14. See Table II.

it may be seen that a lower salt figure also leads to a higher rate of flow at a given pressure.

3. Effect of Jet Hole Size

The same viscose solution was passed through single-hole jets of different size, at five temperatures between 25 and 45°C.

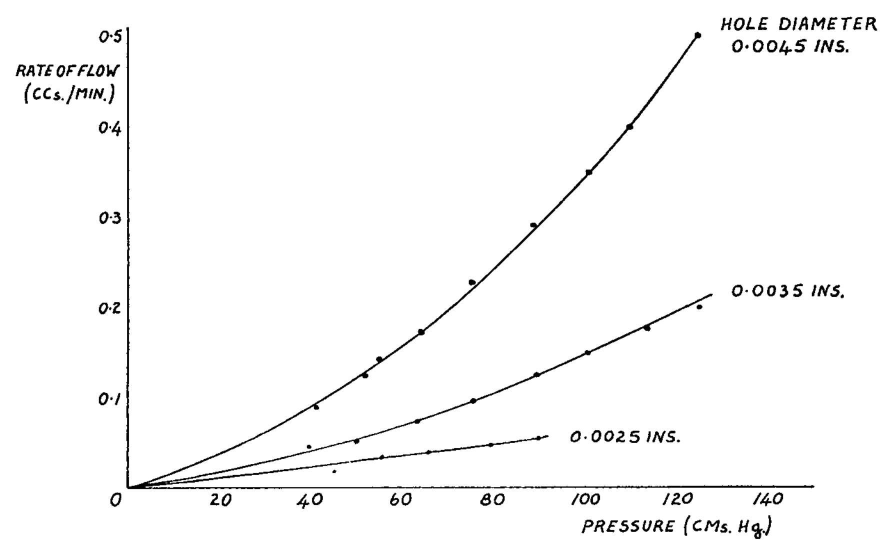

Fig. 4. Flow of viscose solution through single-hole spinning jets at 25°C.

Some typical results are given in Figure 4.

These results have also been used and replotted on a log-log scale and will be discussed below.

DISCUSSION

The results given in Figure 4 have been replotted in Figure 5 on a log–log basis, where the relationship is clearly linear, leading to the equation

$$Q = aP^b \qquad (1)$$

Values of b for the three hole-sizes and five temperatures used, have been extracted and are given in Table III.

TABLE III
Values of b in Equation (1) for Various Temperatures and Jets

	b				
Jet hole size	25°C.	30°C.	35°C.	40°C.	45°C.
2.5 thou.	1.70	1.47	1.72	1.54	1.54
3.5 thou.	1.48	1.52	1.53	1.60	1.54
4.5 thou.	1.44	1.48	1.53	1.52	1.48

It will be noted that apart from two anomalous values, the value for b is about 1.5. Other work[1] has shown a similar relationship between rate of flow and pressure drop for flow through an orifice, using viscose, but the value of b in this case depended on the orifice diameter as shown in Figure 6 ranging between $b = 0.58$ and $b = 0.39$, again demonstrating that flow through a spinning jet is capillarylike rather than orificelike.

An approach due to Rabinowitsch[2] has been used to obtain the fundamental flow curve relating shear stress, s with the rate of shear strain in a

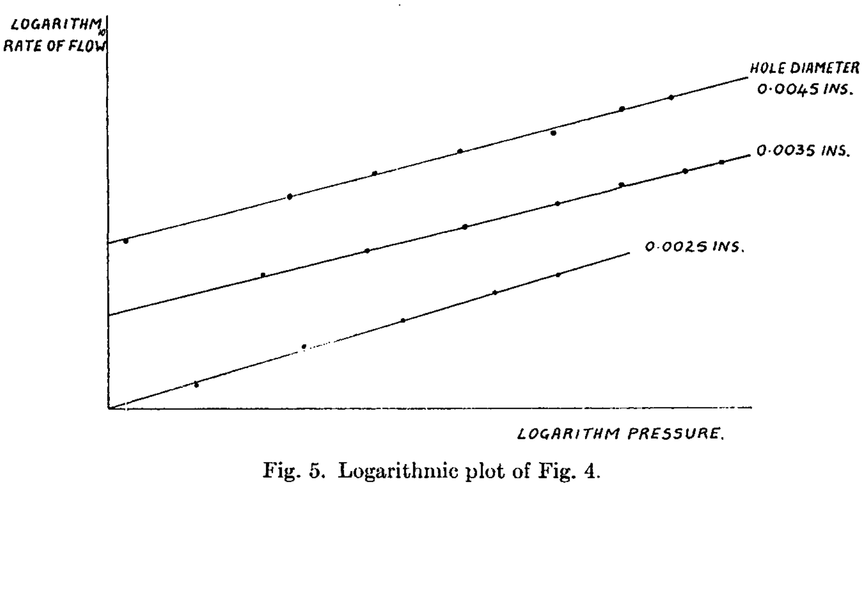

Fig. 5. Logarithmic plot of Fig. 4.

capillary, du/dr, u being the linear velocity in the direction of flow at radius r.

Thus, since

$$du/dr = f(s)$$

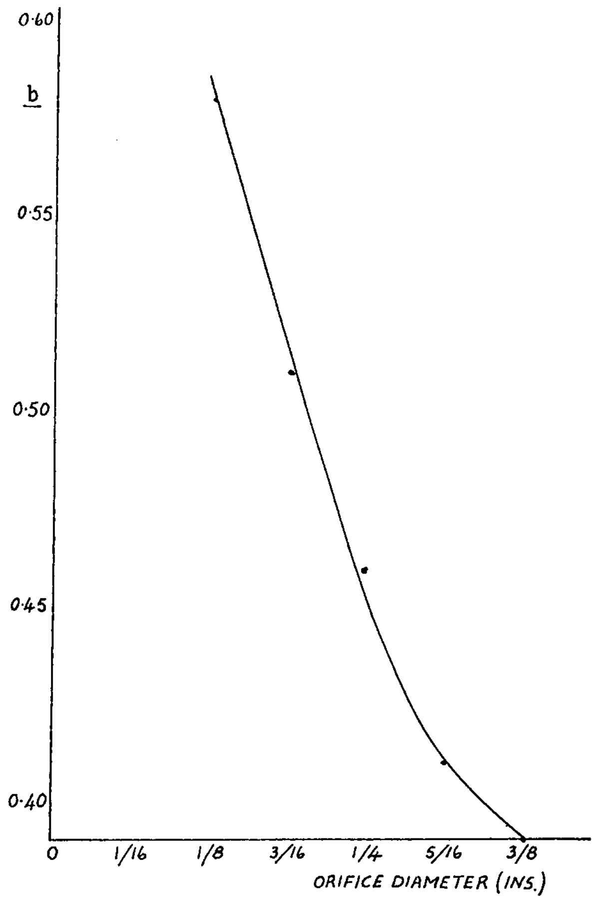

Fig. 6. Value of b in eq. (1) for flow of viscose through orifices.

which, in the case of a capillary is

$$du/dr = f(Pr/2)$$

where P is the pressure gradient, the rate of discharge from the capillary will be

$$Q = 2\pi \int_0^R r \int_r^R f(Pr/2)\, dr\, dr, \text{ which leads to}$$

$$Q = 2\pi \int_0^{S_1} 8S/P^3 \int_S^{S_1} f(\text{S})dS\, dS$$

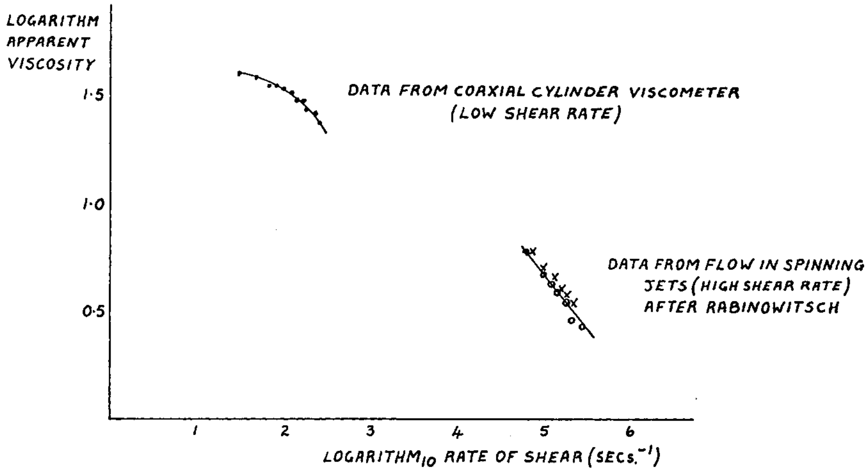

Fig. 7. Fundamental flow relation from experimental data at low and high rates of shear.

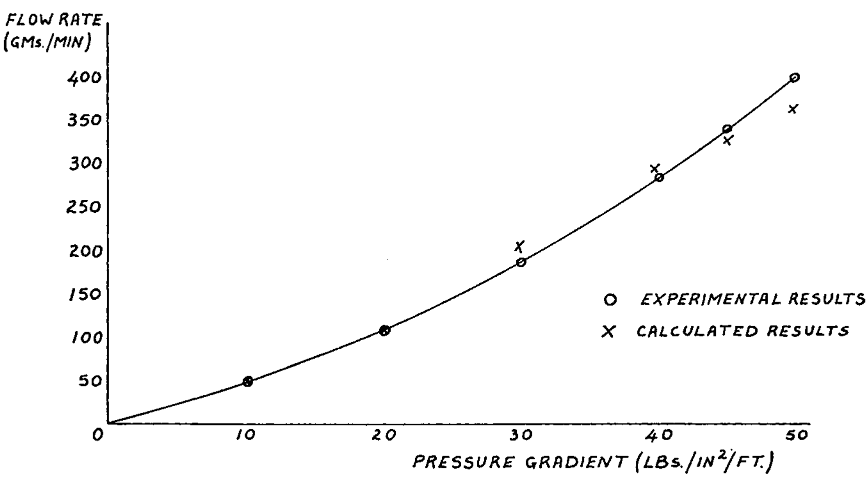

Fig. 8. Flow of viscose through pipe, diameter 0.125 in.

where S_1 is the shear stress at $r = R$, and since P is a constant with respect to integration, it is possible to substitute $P = 2S_1/R$ so that

$$Q = 2\pi R^3/S_1{}^3 \int_0^{S_1} S \left[\int_S^{S_1} f(S)dS \right] dS \qquad (2)$$

By integrating eq. (2) by parts, it is possible to obtain the equation,

$$f(S_1) = 1/\pi R^3(3Q + S_1\, dQ/dS_1) \qquad (3)$$

If now, Q is plotted against S_1 ($= PR/2$), then dQ/dS_1 may be found, and hence $f(S_1)$, which is merely the function $f(S)$ for $S = S_1$.

$f(S_1)$ has been calculated for the 4.5 thou. spinning jet and the apparent viscosity coefficient η, has been obtained from the quotient $S_1/f(S_1)$. η has been plotted against $f(S_1)$ in Figure 7. Values obtained by Toms[3]

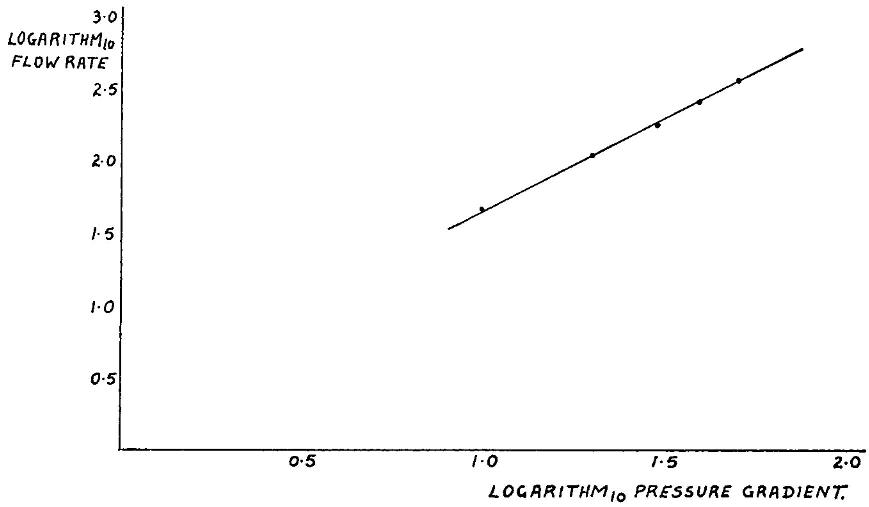

Fig. 9. Logarithmic plot of Fig. 8. Experimental results.

at low rates of shear, in a coaxial viscometer, have also been included, but there is an absence of continuity in the two sets of results, indicating that other factors become important at high rates, possibly a "wall" effect.

Other data, provided by Toms, have been used in a different way to predict the rate of flow of viscose through a narrow pipe (relatively low rates of shear) at various pressure gradients, and these flow rates have been confirmed in an experiment, where the same viscose was passed through a pipe, 2 ft. in length and $^1/_8$ in. in diameter. The data obtained are given in Table IV and are plotted in Figures 8 and 9. The calculated flow rates shown in Table IV have been obtained by starting with the relationship

TABLE IV
Experimental and Calculated Data for Flow of Viscose Through $^1/_8$ in. Pipe

Coaxial viscometer data		Actual flow measurements		
Rate of shear, sec.$^{-1}$	Apparent viscosity, poises	Pressure gradient, lb./in.^{2}/ft.	Flow rate, g./min.	Calculated flow, g./min.
0–5	48.7	10	49.53	50.34
10	48.5	19.9	117.08	116.74
15	48.2	29.7	189.85	202.33
20	47.1	39.6	289.26	294.01
30	45.8	44.6	343.54	333.76
40	44.1	49.7	401.50	362.16
60	41.5			
80	39.8			
100	38.1			
150	34.2			
200	31.6			
300	28.0			

between the apparent viscosity coefficient η and the rate of shear strain, provided by the values given in columns 1 and 2 of Table IV.

Thus,

$$\eta = g(du/dr)$$

and hence,

$$Pr/2 = (du/dr)g(du/dr)$$

For any value of P it is then possible to find du/dr as a function of r, integrate from r to R, the pipe radius, and so obtain u, the linear velocity of flow, as a function of r. Finally, the rate of flow Q may be obtained from the integral,

$$Q = 2\pi \int_0^R rudr$$

It will be seen from Table IV and Figure 8, that the observed rates of flow deviate from the calculated values only at the high pressure gradients. This is not surprising as the calculated values were obtained from extrapolating the coaxial cylinder viscometer data—the highest rate of shear used in the viscometer corresponded to only 24.6 lb./in.^{2}/ft. in the pipe. The log–log plot of the observed rates of flow in Figure 9 is linear, and therefore, of the same form as eq. (1), but in this case, the value for b is 1.40, somewhat less than the value obtained from the results with spinning jets at higher rates of shear strain.

CONCLUSIONS AND SUMMARY OF RESULTS

The anomalous flow behavior of viscose solutions (cellulose xanthate) has been demonstrated in a spinning jet by means of a specially designed apparatus. The results show that for viscose solutions made from the same raw materials, and with identical viscosities at zero rates of shear, the rate of flow at any given pressure gradient is higher for viscose solutions with lower cellulose content and salt figure. When plotted on a log–log scale, the results show a relationship between rate of flow and pressure gradient of the form $Q = aP^b$, where b is of the order 1.5.

Results obtained from the flow of glycerol through the same spinning jets have shown the flow to be capillarylike rather than orificelike.

Calculations from data obtained with a coaxial viscometer have been made on the rate of flow of viscose solution in a narrow pipe and agree, except at the higher shear rates with the observed rates of flow. The rate of flow was again related to the pressure gradient in the same way as above, except that in this case, b is 1.4.

By using an approach suggested by Rabinowitsch, the experimental results obtained with spinning jets have been used to obtain a theoretical relationship between the apparent viscosity coefficient and the rate of shear strain. There is a discontinuity between the theoretical results and those obtained at low rates of shear with a coaxial viscometer, which is presumably connected with the different values of b noted above.

The author wishes to thank the Directors of Courtaulds Limited for permission to publish this information.

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1. Grafton, R. W., unpublished information.
2. Rabinowitsch, B., *Z. Physik. Chem.*, **A145**, 1 (1929); see T. Alfrey, Jr., *Mechanical Behavior of High Polymers*, Interscience, New York, 1948, p. 41.
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Résumé

On décrit un appareil pour mesurer la vitesse d'écoulement de solutions de xanthate de cellulose à des gradients de pression connus à travers des filières. On montre que le trou de la filière se comporte plus comme un capillaire que comme un orifice dans ses effets sur l'écoulement et qu'il existe une relation pour l'écoulement par l'orifice d'une filière et un tube étroit caractérisée par une équation de la forme $Q = Ap^b$, où b a une valeur d'environ 1,5 pour les vitesses élevées de tension dans les filières et de 1,4 pour de plus faibles vitesses trouvées avec le tube. Un examen fondamental montre une discontinuité similaire lorsqu'on compare la relation entre le coefficient de viscosité apparente et la vitesse de tension, entre les vitesses faibles et élevées de tension. On donne des résultats expérimentaux pour l'effet de certains facteurs chimiques sur les solutions de viscose et l'effet de la dimension de l'orifice de la filière sur la courbe de gradient d'écoulement vitesse/pression.

Zusammenfassung

Ein Apparat zur Messung der Fliessgeschwindigkeit von Cellulose-xanthatlösungen bei bekanntem Druckgradienten über die Spinndüsen wird beschrieben. Das Düsenloch verhält sich bezüglich seines Einflusses auf das Fliessen mehr wie eine Kapillare als wie eine Öffnung und für das Fliessen durch Spinndüsenlöcher und eine enge Röhre bestehen ähnliche Beziehungen, für die eine Gleichung von der Form $Q = Ap^b$ charakteristisch, ist, wo b für die hohe Schergeschwindigkeit in der Spinndüse einen Wert von etwa 1,5 und für die niedrigere Geschwindigkeit in der Röhre einen solchen von 1,4 hat. Eine grundlegende Untersuchung führt zu einer ähnlichen Diskontinuität beim Vergleich der Beziehung zwischen dem scheinbaren Koeffizienten der inneren Reibung und der Geschwindigkeit der Scherungsverformung zwischen niedriger und hoher Schergeschwindigkeit. Versuchsergebnisse bezüglich des Einflusses gewisser chemischer Faktoren bei Viskoselösungen und des Einflusses der Grösse der Spinndüsenöffnung auf die Kurve Fliessgeschwindigkeit gegen Druckgradient werden mitgeteilt.

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