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Schofield, R. K. and Blair, G. W. S. 1933. The relationship between viscosity, elasticity and plastic strength of a soft material as illustrated by some mechanical properties of flour dough.—II. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*. 139 (839), pp. 557-566.

The publisher's version can be accessed at:

- <https://dx.doi.org/10.1098/rspa.1933.0038>

The output can be accessed at: <https://repository.rothamsted.ac.uk/item/95x75/the-relationship-between-viscosity-elasticity-and-plastic-strength-of-a-soft-material-as-illustrated-by-some-mechanical-properties-of-flour-dough-ii>.

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bulb, falling to 0.49 in a 2 cm. cylindrical tube. Temperatures up to 300° C. do not influence γ . Molecular hydrogen has no effect, but atomic hydrogen decreases γ . The mechanism of the reaction is discussed and compared with the photo-sensitized reaction.

On adding oxygen to PH_3 and illuminating, a stable chain reaction occurs, and with pressures of $\text{PH}_3 = \text{O}_2 = 0.05$ mm. $\gamma = 200$. The significance of the value is discussed in relation to the theory of branched chains. It is concluded that the probability of branching at any one cycle is not more efficient than 5×10^{-3} .

A compact and sensitive form of combined reaction tube and McLeod gauge is described.

The Relationship between Viscosity, Elasticity and Plastic Strength of a Soft Material as Illustrated by some Mechanical Properties of Flour Dough.—II.

By ROBERT KENWORTHY SCHOFIELD and GEORGE WILLIAM SCOTT BLAIR.

(Communicated by Sir John Russell, F.R.S.—Received October 1, 1932.)

In an earlier communication* Maxwell's "time of relaxation" was given an extended and more general definition so that the conception might be used in describing the behaviour of plastic substances. From a study of such materials in steady flow it is well known that the viscosity defined as the ratio of the shearing stress, S , to the velocity gradient or rate of shear, G , is not a constant but usually decreases as S increases. The time of relaxation, t_r , is related to the viscosity, η , thus

$$t_r = \eta/n,$$

n being the rigidity modulus. Since n is normally independent of the stress,† t_r and η show parallel variations.

For ordinary fluids t_r is very small and no way has yet been devised for measuring it. In flour dough, however, we have a material in which high viscosities are combined with a low rigidity modulus, and consequently the relaxation of internal stress is slow enough to be easily followed experimentally.

* 'Proc. Roy. Soc.,' A, vol. 138, p. 707 (1932).

† p. 566.

Observations made on cylinders of dough which had been stretched to various extents showed that the relaxation time (and hence the viscosity) depends on the degree of stretching as well as on the stress.

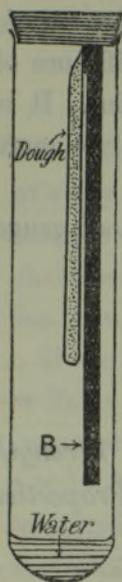


FIG. 1.

It appeared desirable to obtain confirmation of this double dependence of viscosity on the extent of shearing as well as on the shearing stress by a more direct method. For this purpose, a study was made of the rate of elongation of cylinders of unyeasted dough hung vertically by their upper ends and allowed to extend under the action of gravity. In carrying out these observations it has been found convenient to mark on the dough cylinders a series of fine parallel lines accurately spaced 1 mm. apart. The marks were made by successive turns of a fine wire wrapped round a frame, which are wetted with enamel, the marks remaining wet long enough to be subsequently printed off on to a strip of duplicator paper. This method has the advantage of enabling an instantaneous record to be made of the deformation of a series of elements throughout the length of the dough cylinder. The recording is rapid and permanent, and the print (which may be called a rheogram) is available for whatever analysis appears suitable, fig. 2.

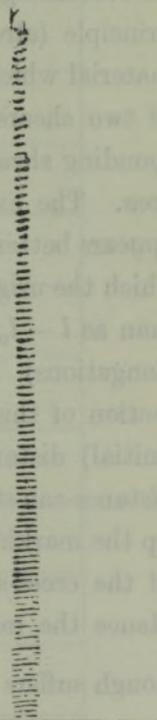
In obtaining the data which is about to be discussed, a number of precautions were taken:—

- (1) *Moisture Content.*—The doughs used had a moisture content such that when they were pressed firmly on to a glass plate they just did not stick appreciably to it. At the first mixing, slightly insufficient salt solution—strength 2·5 per cent.—was added to the flour, and after half an hour a little more was kneaded in so as to produce the desired condition. If, on thoroughly kneading at the end of a further half-hour, the moisture condition was judged to be correct, the dough was considered to be satisfactory for testing. In order to prevent drying during the test, the shaping, and marking of the cylinders was carried out as quickly as possible and while extending under gravity they were hung inside large boiling tubes containing a little water, see fig. 1.
- (2) *Shaping the Cylinders.*—Each of the pieces into which the dough was divided was forced through a short metal tube attached to a wider tube fitted with a plunger. In order to prevent avoidable straining, the “gun” was held vertically, the cylinders being extended downwards. Each cylinder as it was formed was detached and held by its

upper end which was then pressed firmly on to the support provided, see fig. 1. The diameter of these cylinders, about 7 mm., was surprisingly uniform except for a "blob" at the end, which was always cut off. The length used for the test depended on the nature of the dough, and on the time allowed for the extension, the aim being to use the longest cylinder that would not break off prematurely. In practice the lengths varied from 4 to 12 cm.

(3) *Elastic Deformation.*—So long as the dough cylinder is hanging vertically, it is subject not only to plastic flow, but also to an elastic deformation which could be recovered by placing it horizontally. This recovery will only proceed to completion on a frictionless support like a mercury bath. It was found convenient to apply the marker and also to print off the rheogram with the dough lying horizontally on the wooden back B, but in neither case is the elastic deformation recovered owing to the tendency of the dough to stick lightly to the back as soon as it touches it. The elastic deformation was generally small compared with the plastic deformation, and as it was present both at marking and at printing, it cannot appreciably have affected the results.

(4) *Printing the Rheograms.*—As a certain degree of pressure is needed when printing the rheograms which somewhat flattens the cylinder it was feared that the marks on the print might not faithfully reflect the state of affairs before the paper touched the dough. Such fears are, however, allayed by an examination of the rheograms themselves, fig. 2. The clearness of the impression makes it evident that no movement of the *surface* of the dough occurs once it has touched the paper. Before printing off, a line is ruled on the paper which is placed against the lower end of the dough cylinder when taking the print. This line provides the zero for computing the stress.



(1)
 $t = 8$
FIG. 2.

Although the apparatus and procedure are simple, care is needed in the manipulation lest the test cylinder be accidentally strained at some stage. So easy is it to spoil an experiment that the cylinders were usually tested in

triplicate. In the case of the data about to be discussed, one of each of the three sets had to be discarded leaving only duplicate rheograms for the final analysis.

When analysing the rheograms it is not necessary to know the dimensions of the cylinders, provided that they were initially of uniform cross-section. Except within a few millimetres of the support, the behaviour of any element is governed exclusively by the original length of the dough which hung below it. The top two or three marks are usually distorted and are always disregarded. In evaluating the shear and the shearing stress, use is made of the well-known principle (already employed in the earlier paper) that an elongation of a material which does not change its volume may be expressed as the resultant of two shears each equal in magnitude to the elongation, while the corresponding shearing stresses are equal to one-third of the tensile stress per unit area. The extensions involved here are sometimes several-fold, so that it appears better to express the elongation, and hence the shears, in an element of which the original length, l_0 , has increased after time, t , to l ; as $\log_e l/l_0$ rather than as $l - l_0/l_0$ (although, of course, the two expressions are equal for small elongations). The tensile stress per unit area acting over the meridian cross-section of this element was equal to $g\rho L_0$ where ρ is the density and L_0 the (initial) distance of the section from the lower end of the cylinder. This distance can still be ascertained after the elongation has taken place by counting up the markings. The elongation is accompanied by a proportional shrinkage of the cross-sectional area, so that the tensile stress at time t is $g\rho L_0 l/l_0$. Hence the mean shearing stress equals $\frac{1}{3} g\rho L_0 (1 + l/l_0)$ under which the dough suffers a mean rate of shear of $\frac{1}{t} \log_e l/l_0$.

In fig. 3 the mean rate of shear is plotted against the mean shearing stress for three times-of-hanging of cylinders all formed from pieces of the same dough and tested in rapid succession. The flour was a good bakers' mixture. A value of 5 mm. was selected for the length, l_0 , of an element, and l was found by determining the distance between each mark and its fifth neighbour. Each point is the result of a measurement of this kind. Two rheograms were obtained for each of the three times-of-hanging, and the points from all six are shown.

The lack of coincidence between the three sets of points shows that the rate of shear under a given stress is not uniform but diminishes as the shearing proceeds. It is very important to bear this fact in mind when seeking to interpret the curves. Apart from this fact their negative curvature, which is

particularly marked for the 8 min. curve, would be very surprising, involving an increase of viscosity with increase of stress. This curvature is the result of the double dependence of viscosity on stress and shear. The position emerges more clearly if sets of points corresponding to equal shears are picked out on the three curves. One such set is connected by the broken curve. This evidently leaves the axis of the abscissa at about $S = 800$ (since below this

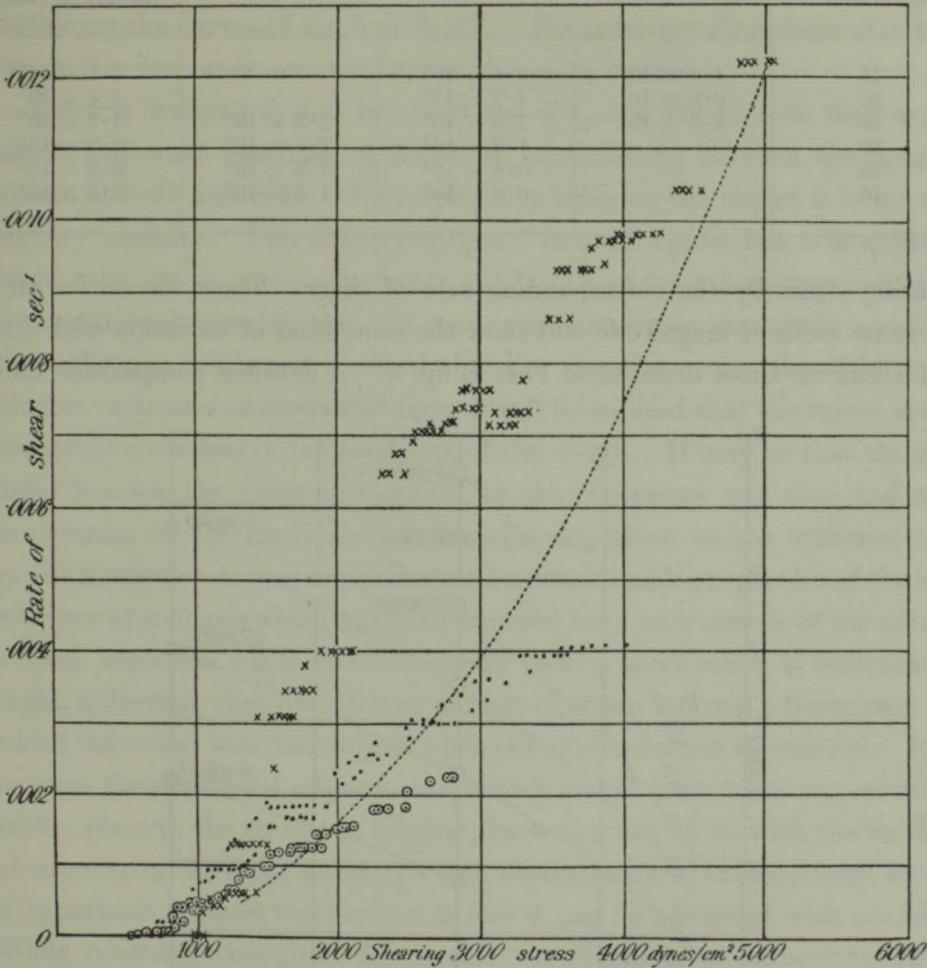


FIG. 3.—○ $t = 64$; • $t = 32$; × $t = 8$ minutes.

stress the shear is zero for all three times) and thence shows a consistent upward curvature. A family of such curves could be drawn each having the same form.

In order to evaluate the viscosities, the variation of shear with time for a series of constant mean stresses has been traced in fig. 4, the positions of the points being obtained by interpolating on fig. 3. The slope of one of these

at any particular point, gives the rate of shear under that stress after a shear measured by the ordinate has taken place. The figures given in the table for shears of 2.5, 4.1, and 7.0 were obtained in this way by dividing each

Shear.	0.25	0.41	0.70	0.58
Stress dynes/mm. ² .	Viscosities from rheograms.			Viscosity from relaxation times.
15	1.6×10^7	2.4×10^7	—	2.1×10^7
20	0.9×10^7	1.7×10^7	—	1.4×10^7
25	0.7×10^7	1.6×10^7	2.3×10^7	1.1×10^7
30	—	1.2×10^7	2.1×10^7	0.8×10^7
35	—	0.9×10^7	1.8×10^7	0.6×10^7
40	—	0.8×10^7	1.6×10^7	0.5×10^7

shearing stress by the corresponding rate of shear. These viscosities are of the same order of magnitude and show the same kind of variation with stress and shear as those deduced in Part I, fig. 5. A detailed comparison shows,

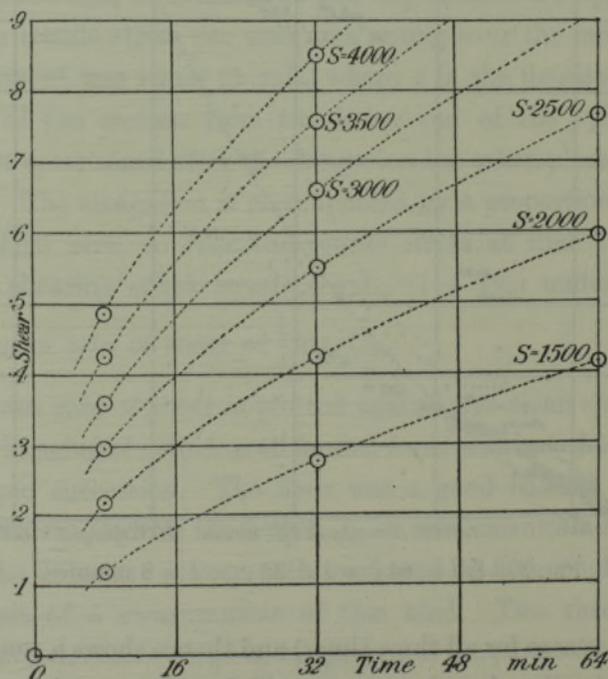


FIG. 4.

however, that the values obtained by the earlier method are consistently lower. As the new flour, though very similar, was not identical with that used in the former experiments, a set of observations of the relaxation of stress was

carried out on a dough cylinder made from the new flour (the old flour being no longer available). The measurements were carried out in the way described in Part I (*loc. cit.*) and the viscosities which are given in the last column of the table were obtained by finding the relaxation times $\left(= \frac{1}{d(\log_e S)/dt} \right)$ for a series of stresses, and multiplying by the rigidity modulus $(= \frac{1}{3}$ Young's modulus). An extension of 80 per cent. was used, which falls between those used in obtaining the curves of fig. 5 of Part I. The corresponding shear of 0.58 also lies within the range covered by the rheogram figures.

The new values fall into line with the old. As will be seen they are only about half what would be obtained by interpolating between the figures for shears of 0.41 and 0.70. They rest, it is true, on the use of 2×10^4 as the rigidity modulus. This is admittedly a "round" figure, but it is unlikely to be in error by as much as a factor of 2*. There are other experimental and computational uncertainties, but none appears serious enough to account for the difference. When it is noted that the viscosities vary threefold for only modest variations of stress and shear it will be realized that the values are very sensitive to changes in the condition of the dough. It may be that the results differ because the shearing recorded in the rheograms was slow and steady in contrast to the rapid preliminary shearing given to the cylinders in the earlier method. As was noted in Part I a considerable proportion of the elastic recovery of a dough which has been strained for a long time is of the nature of a creep while the whole of the recovery from a short strain is comparatively rapid, indicating that during long periods of strain internal adjustments occur which take some time to readjust when the external stress is removed. It may be that these internal adjustments, which would have more opportunity of taking place in the rheogram method, have as much to do with the variations of viscosity as the shear associated with the alteration of external form, but such a hypothesis requires further test before it can be advanced with confidence. While reserving this point for further investigation we appear justified in taking the general concordance of the two sets of figures as substantiating the correctness of the relationship

$$\eta = t_r \cdot n.$$

In a general way the results from the rheograms show an interesting correspondence with those of Trouton† for pitch, though there are contrasts in the

* *Note.*—p. 566.

† 'Proc. Roy. Soc.,' A, vol. 77, p. 426 (1906).

magnitudes concerned. The stresses used by Trouton were considerably larger, and the rate of shear for a given stress approached constancy after a much smaller degree of shearing. Trouton stated that after the lapse of about half an hour under a constant stress a steady rate of shear was reached, but the data shown in his fig. 2, p. 428, are consistent with the view that a decrease was going on during the whole of the experiment. It is also interesting to compare his fig. 3, p. 429, with our fig. 3. Although he draws a straight line in his graph, his points indicate a negative curvature just as do ours in the case of dough. In both cases the curvature is only negative above a certain stress limit. For the dough, the curves start from the origin and remain practically coincident with the axis of the abscissa until somewhere between $S = 600$ and $S = 1000$, they then take a very steep upward bend after which the gentle negative curvature sets in. The upward bend marks the yield value, the corresponding stress measuring the shearing strength of the material under the conditions of the experiment. Below this stress no detectable flow takes place during the time of the experiment, and the material behaves like a solid.

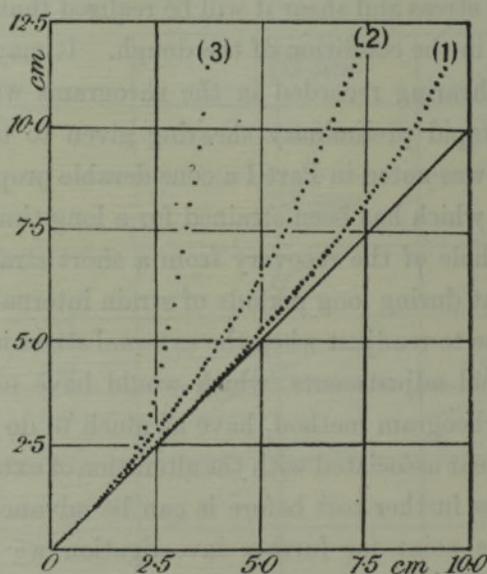


FIG. 5.—(1) Manitoba flour ; (2) Baker's mixture ; (3) English flour.

Although the foregoing analysis is needed to ascertain the factors upon which the rate of flow of a dough depends, a much less elaborate treatment suffices in many cases to distinguish doughs by their flow properties. Fig. 5 has been constructed by placing each of the three rheograms in turn along the axis of the ordinate and plotting the marks in turn against successive millimetre divisions along the abscissa. If no flow had taken place the result

would have been a line from the origin, of slope unity, and the aggregate flow in 20 minutes is shown by the departure of points from this line. This way of plotting easily differentiates the three flours in question, but the capacity of the method to distinguish the small differences which are of importance in baking has still to be determined. The outlook in this direction is, however, distinctly hopeful, as, from the point of view of the stresses used and their times of application, the conditions of the test correspond closely to those ruling inside a dough that is distending under the action of yeast.

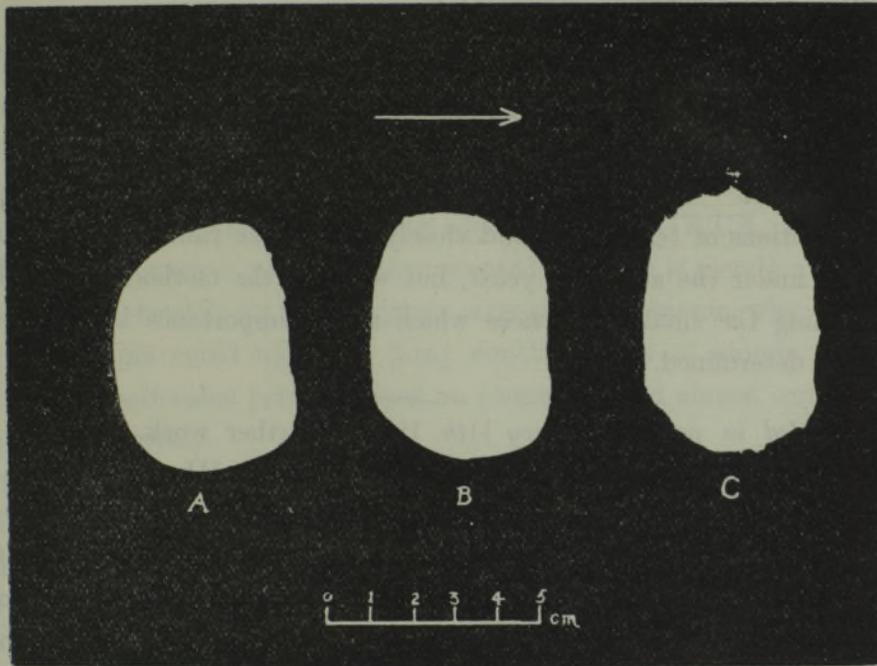


FIG. 6.

The hardening of a dough by shearing was independently demonstrated by a simple experiment. Three doughs were made up *with yeast* and kneaded well. Each was then pulled out and folded over a number of times, care being taken that the elongation and folding were always made in the same direction. The final fold was made so as to give the dough a form as nearly cubic as possible, finishing touches being given by slight pressure of the thumb and fingers. As the photograph, fig. 6, shows, the swelling was uneven, the smaller expansion occurring in the direction in which the dough had previously been stretched, as shown by the arrow. Dough B was at the standard moisture content, A being a little wetter, and C a little dryer.

Summary.

The dependence of the viscosity of a flour dough on the shear which has taken place as well as on the shearing stress is brought out by a series of observations on the rate of shear in cylinders of unyeasted dough hung vertically and allowed to elongate under the action of gravity.

The deformations were recorded by marking a millimetre scale in enamel on the surface of the dough cylinders, and, after elongation had proceeded for a measured time, printing the deformed scale off on to a strip of duplicator paper. The print has been called a rheogram.

The agreement between the viscosities determined directly in this way and those obtained indirectly in Part I is satisfactory as substantiating the theory that the viscosity is equal to the product of the rigidity modulus and the relaxation time.

The conditions of test correspond closely with those ruling inside a dough distending under the action of yeast, but whether the method is capable of distinguishing the small differences which are of importance in baking has still to be determined.

[*Note added in proof, January 11th, 1932.*—Further work, of which the results have been submitted for publication as Paper III of this series, has shown that, although n is far less variable than η , it is not exactly constant. (cf p. 557, lines 9 and 10.) Since writing the statement on p. 563 to the effect that the value of 2×10^4 for the rigidity modulus used to compute viscosity from relaxation time “is unlikely to be in error by as much as a factor of 2,” the authors have concluded that 2×10^4 , although a good mean value, is not that most appropriate for this computation. Using the value which the latest experiments have shown to be applicable to the conditions of stress relaxation, a close agreement is found between the viscosities calculated from the relaxation times and those obtained from the rheograms.]
