The Application of a Capillary Extrusion Rheometer to the Determination of the Flow Characteristics of Lard[1]

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Abstract

A capillary extrusion rheometer was designed, and techniques were developed to obtain basic information on the flow behavior of lard. Since constant and adjustable shearing rates are a necessary feature for the study of the basic rheological behavior of a material, the rheometer was designed for use in conjunction with a tensile tester having a wide selection of crosshead rates. Techniques were developed for accurately measuring the flow variables and for treating the data. A comparison study on a Newtonian liquid sample proved the validity of the approach. Lard at 23.4C was tested, and its flow parameters (fluid-consistency index and flow-behavior index or degree of non-Newtonian behavior) were obtained.

Introduction

RHEOLOGICAL STUDIES of lard and butter have been made but are too few in number (1–4) and of questionable value for obtaining basic flow-behavior information. Since it can be expected that lard at ambient temperatures would exhibit non-Newtonian behavior, the procedure and equipment employed to measure its rheological properties are of prime importance.

Of the several types of rheometer available, the rotational and extrusion types lend themselves most readily for a meaningful study of this kind. A rotational viscometer (coaxial cylinder, rotating disc, or cone and plate) (5) could be used if the sample can be prevented from rising out of the gap between the rotating and stationary elements at the higher shearing rates. Extrusion rheometers are of two types, the constant loading (fixed pressure) type (6) and the constant shearing type (7). Both have the wide range of shearing rate necessary for measuring non-Newtonian behavior. The main limitation of the capillary extrusion rheometer is its inability to distinguish between energy losses because of measurement errors that are inherent to the instrument and energy losses because of pressure drop across the capillary.

The purpose of this paper is to show that, with the proper selection of instrumentation and techniques (experimental and mathematical), more meaningful and useful rheological parameters of lard and similar materials can be obtained.

Instrumentation and Techniques

Rheometer and Associated Instrumentation

In addition to the theoretical considerations it was practical to use a capillary extrusion rheometer because this laboratory possesses an Instron Tensile Tester Model TT-B. This instrument has incremental crosshead rates from 0.02 to 10 in. per minute and a variable load-weighing capacity from 0 to 1,000 lb

with an accuracy better than $\pm\frac{1}{4}\%$ for all load ranges. A special compression bridge was built, which adapted the Instron to the rheology instrument design contained herein. It is not to be inferred that only sophisticated instrumentation as described above can be used to perform this test. Any device that can apply a range of constant plunger rates to the rheometer should be suitable.

The rheometer (Fig. 1) used in this study follows the Mertz-Colwell design (7). It has four basic parts: the barrel, the plunger, the capillary, and the thermal jacket. The barrel was made from low carbon lead-bearing, screw-stock steel bar 1.000 × 6.625 in. An axially located bore 0.500 in. in diameter was drilled through the bar. The nominal size of the bore is not significant, but it was measured to within ±0.0025 of an inch. The uniformity and "finish" of the bore are extremely important. It should not be bell-mouthed, barrel-shaped, or tapered; and the finish should be "mirror-like."

The plunger was a 0.5-in. diameter steel bar threaded at one end, for attachment to the bridge, and capped at the opposite end with a Teflon plug (piston), which could be adjusted to the barrel diameter for the most advantageous fit. The efficiency of the piston-to-barrel fit and the capillary-barrel seals was tested by noting the difference between an extruded volume of material and its volume calculated on the basis of the rheometer geometry. This was done for four cross-head rates. The average discrepancy was 0.25% volume difference. The calculated volume in all cases was 0.6010 cu in. In this test and subsequent testing the pressure drop across the end of the capillary did not exceed 235 psi.

The capillaries were made of stainless steel hypodermic needle tubing and were attached to the barrel by a threaded arrangement which required no gasketing material to make a reliable seal. The capillary radius, which has to be measured accurately since it enters into the shearing rate calculation as the cube and the viscosity calculation as the fourth power, was determined by direct measurement when the radius was greater than 0.020 in.; below this value the "effective capillary radius" was measured by a method similar to the one described in the ASTM Standards (8). Capillary radii measured by this method proved to be quite accurate. A statistical study of capillary radius data for two capillaries showed that 95 times out of 100 the radius measurements were respectively 0.29% and 0.32% within the average of two sets of measurements.

The design length of each capillary was calculated from the selected nominal radius and aspect ratio and was measured with a micrometer caliper. A tolerance on the length as large as ±0.010 of an inch was permissible because of the long capillaries used in this study. A threaded brass fitting was permanently soldered to the end of each capillary, and this provided the means by which the capillaries were attached to the rheology barrel. All these fittings had 90° entrance-angle tapers.
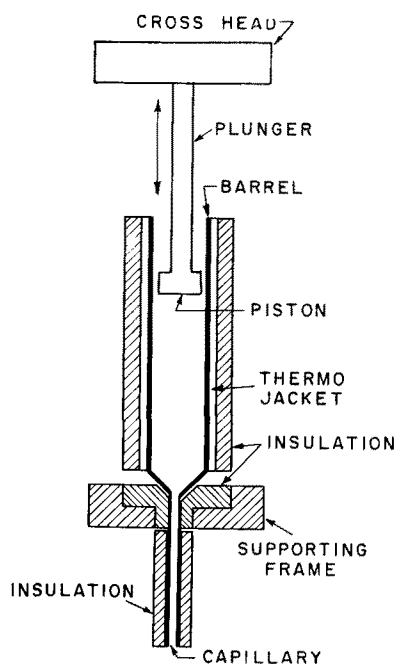
The thermostat consisted of an ethyleneglycol-water

FIG. 1. Schematic diagram of the rheometer.

bath adjustable from −10 to +60C ±1C, a thermal jacket that fitted the rheology barrel snugly, and a circulating pump and lines for circulating the bath medium through the jacket. Temperature control was maintained in the bath, and thereby in the rheology barrel and capillary because of the efficiency of the insulation (Fig. 1). An adjustable mercury thermoregulator coupled to a sensitive electronic relay was used for temperature control. Electric heaters and an ice compartment supplied the energy that the thermoregulator controlled. Barrel temperature was monitored by a thermocouple placed in a well provided for it in the rheology barrel.

Operation of the Rheometer

To compensate for the instrumental error, which is made up of the fluid friction (shearing of the sample in the rheology barrel) plus the mechanical friction of the piston-to-barrel contact, a table relating crosshead rate to consecutive piston travel increments was employed whereby the operator could refer a measured load-point to the corresponding piston position point in the rheology barrel for each crosshead rate used.

Instrument blank data were obtained by running a sample through an orifice of the same diameter as the test capillary; a selected blank datum was then subtracted from an experimental load-point obtained from the capillary run, taking care to refer each blank and capillary number generated by a particular crosshead rate to the same point along the rheology barrel.

The piston travel increments were determined by the gauge length dials of the Instron. After each increment was run, the lower gauge length dial was reset and the transmission shifted to the next crosshead rate.

Noise was always present in the force trace regardless of the material being extruded (liquid or solid). Optimum conditions were obtained by using a Teflon piston head 0.002 in. larger in diameter than the barrel average diameter. A slight "back rake" was given to this piston head so that it was the frustum of a cone and not cylindrical. This was done to

minimize the contact area of the piston head with the barrel wall. Cyclical forces generated by the piston-to-barrel friction and a fluid characteristic could be the cause for the short-term noise in the force trace.

Theory and Experimental

Theoretical Considerations

A Newtonian fluid undergoing laminar flow exhibits a linear relationship between shearing stress and shearing strain; the ratio of stress to strain is the viscosity of the fluid. Thus the rheological behavior of the fluid is completely defined by a single value. Non-Newtonian fluids, on the other hand, exhibit a nonlinear relationship between shearing stress and shearing strain; consequently the ratio or so-called viscosity of such fluids varies as a function of shearing rate.

A general method has been developed which is applicable to all fluid systems whether Newtonian or non-Newtonian. Mathematically this method is described by the empirical "power law," which may be written as follows:

$$S = KD^n \qquad [1]$$

where   S = the true shearing stress

      K = a rheological parameter generally referred to as fluid consistency index

      D = the true shearing rate

      n = a rheological parameter called the flow-behavior index

It is obvious that when $n = 1$, K becomes simply the ratio of stress to strain which is the definition of viscosity; hence the fluid is Newtonian. Thus it can be seen that the value of n provides a measure of the deviation of a fluid from Newtonian behavior.

Practical Considerations

Of primary importance to a rheological measurement with capillary extrusion instrumentation is a steady-state condition (equilibrium) and laminar flow of the sample. The criterion for steady-state condition in the absence of turbulence and time-dependent effects should be steady extrusion rate. Instruments such as those employed in this work which have a constant plunger (piston) rate and therefore a constant extrusion rate, are steady-rate-of-flow instruments. Conversely it is not possible to have steady-state flow with a constant load instrument because in this instrument there is a dynamic pressure gradient within the sample in the rheometer barrel which increases from the top-to-bottom position of the plunger. This pressure gradient is the result of the lessened fluid resistance to the applied constant pressure as the unextruded sample volume decreases. It follows that the flow rate must increase if the dynamic forces are to remain in balance with the static head. Charley (9,10) showed that the pressure gradient in the rheometer barrel of a constant-load rheometer was responsible for an increase of the flow rate along the length of the barrel, which in the case he cited could increase by a factor of 3.6 in the course of a run. However, even though a constant plunger rate ensures a steady-state flow condition, a pressure gradient still exists within the sample in the rheometer barrel, but now it manifests itself as a shear stress gradient inverse to the flow-rate gradient of the constant-load instrument. This gradient shows up in the force trace (shearing stress), depending on the
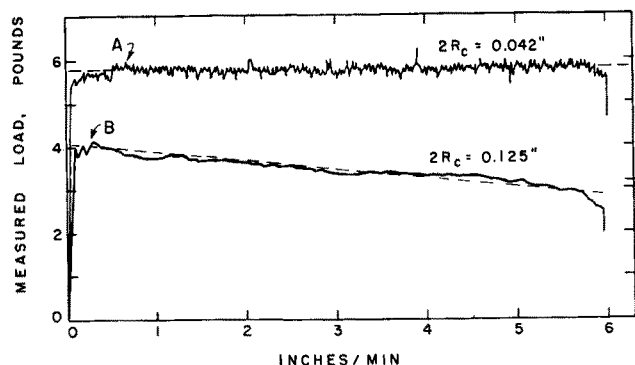
FIG. 2. Force trace "A"; capillary aspect ratio 30:1. Force trace "B"; capillary aspect ratio 5:1.

aspect-ratio of the capillary and the diameter ratio of the barrel and capillary. Figure 2 illustrates the effect of capillary-aspect ratio and barrel/capillary diameter ratio on the force trace. The difference in aspect ratio has the more pronounced effect. The cause of the slanting force trace "B" becomes apparent when it is realized that the fluid resistance in the barrel and capillary represents a series or additive resistance load and therefore will show up as a slanting line if the shearing stress in the barrel is large relative to the shearing stress in the capillary. The pressure drop in the barrel relative to the pressure drop in the capillary for a Newtonian material under steady-state and isothermal conditions is expressed mathematically by equation 2 (11):

$$\Delta P_B/\Delta P_c = (L_B/L_c)\,(R_c/R_B)^4 \qquad [2]$$

where

ΔP_B = pressure drop in the barrel in psi

ΔP_c = pressure drop in the capillary in psi

L_B & R_B = "effective length" and radius respectively of the barrel in inches

L_c & R_c = length and radius respectively of the capillary in inches

In equation 2, L_B varies as the measurement proceeds whereas the other factors in the right-hand side of the equation and ΔP_c remain constant. This leaves ΔP_B solely dependent on ΔL_B.

In trace "A" of Figure 2 the pressure drop in the barrel has been overshadowed but not eliminated. This and other error factors must be taken into account if accurate results are to be obtained. There are five components in the force trace which must be accounted for before the shearing stress measurement and calculation can be considered a true shearing stress. These components are as follows: a) the shearing of the sample within the barrel plus the piston-to-barrel friction; b) the shearing stress of the sample within the capillary, which stress is capillary length/diameter-dependent and consequently subject to random error; c) the energy because of entrance-and-end effect (12); d) the elastic energy present in a flowing liquid; and e) the pressure drop on account of the kinetic energy of the sample at higher flow velocities. The instrumental factor can be measured directly and can therefore be subtracted from the raw data before any application of the data is made. Mathematical techniques are used to compensate for the remaining four errors. There is also a shearing rate error whenever non-Newtonian materials are tested, and this too must be corrected before accurate rheological parameters can be obtained.

The true or corrected shearing stress, S_T, at a constant rate of shear is obtained from a plot of the corrected measured load, $L_{M(CR)}$, *versus* capillary length/diameter ratio. The slope of this plot when divided by a constant gives the true shearing stress directly. Because several capillaries of significantly different length/diameter ratio are used to obtain the measured loads, the effect of capillary geometry (random error) on the shearing stress is minimized. The formulation which expresses the above function and includes all the measurement errors, except the first, is shown in equation 3. This is the basic equation of the Philippoff-Gaskins paper (13), adapted to a constant rate of shear rheometer.

$$L_{M(CR)}/\pi R_B^2 = S_T[\,(2L_c/R_c) + 2N + S_{Rc}] + M\,\rho V^2 \qquad [3]$$

where

$L_{M(CR)}$ = measured load with instrumental error removed

R_B = radius of rheology barrel

S_T = corrected or true shearing stress

L_c & R_c = length and radius of the capillary respectively

N = entrance-exit correction; "Couette Correction"

S_{Rc} = recoverable shear at radius R_c

$M\,\rho V^2$ = kinetic energy correction

where

$M = 1.12$ for the normal parabolic distribution of velocity across the capillary radius

ρ = density of sample

$V = (R_B^2/R_c^2)$ (crosshead rate in in./sec); the velocity of the sample in the capillary

The kinetic energy (times the area of the rheology barrel) of lard at 23.4C was calculated; the crosshead rate was 10 in./min and $R_c = 0.017$ in. (It is necessary to multiply the kinetic energy by the area of the barrel so as to get lbs-force, the units of the force trace.) When the calculated kinetic energy times area was divided by an experimental force-trace value generated by the same instrumental parameters that were used in the kinetic energy calculation, the percentage figure turned out to be 0.112%, a negligible amount. Similarly when the recoverable shear was calculated, it also was found to be negligible. Equation 3 can then be written

$$L_{M(CR)}/4\pi R_B^2 = S_T(L_c/2R_c) + S_T(0.5N + 0.25\,S_{Rc}) \qquad [4]$$

where

$4\pi R_B^2$ is equal to $4(3.1416)\,(0.2525)^2$.

Philippoff and Gaskins as well as Bagley have proved that $L_{M(CR)} = f(L_c/2R_c)$ is a linear function and in no way is the shearing stress, when determined by this equation dependent on capillary geometry (12,13); therefore, if the corrected measured load vs. incremental capillary length/diameter ratios are plotted for equivalent shearing rates, one is then able to obtain the true or corrected shearing stress for that particular shearing rate. The true shearing stress is obtained by dividing the slope of this plot by $4\pi R_B^2$. Since the value of a slope of a straight line was used throughout this study, the least-squares equations were applied to the data for greater accuracy. Since the data for this plot must be obtained under the condition of equivalent shearing rates, it should be pointed out that a direct method of getting
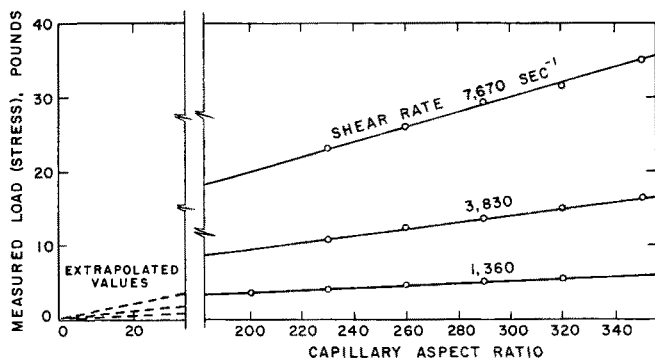
FIG. 3. Measured load vs. capillary-aspect ratio; slope gives shearing stress directly. Flexol 8N8.

an equivalent shear rate out of capillaries of different length/diameter ratios is to make all the capillaries the same diameter, thereby intrinsically referring the different loads generated by the various capillary lengths to the same shearing rate. This is true because in the shearing-rate equation

$$D = 4Q/\pi R_c^3 \qquad [5]$$

where

D = shearing rate sec.$^{-1}$

$Q = \pi R_B^2$ (crosshead rate in in./sec) or volumetric rate of flow

R_B is a fixed value and D will only be dependent on R_c and the crosshead rate. When the values of the instrument parameters are substituted in this equation, a D (shearing rate) constant can be calculated for each capillary radius, and it is then only necessary to multiply the crosshead rate by this number to find the shearing rate in sec^{-1}.

To obtain the lb-force and rational values of the intercepts, $L_c/2R_c$ and $L_{M(CR)}$ are extrapolated to zero respectively. The y intercept value represents the sum of the recoverable elastic energy, the kinetic energy, and the Couette effect, but it is not needed to obtain the true shearing stress. The Couette effect is not pronounced at the lower shearing rates when large values of capillary-aspect ratio are used. According to Bagley, the Couette effect is almost solely a function of the shearing rate. Therefore one can expect the straight lines to come close to passing through the origin at the lower shearing rates. This proved to be the case, as Figure 3 illustrates.

Experimental Check of Instrumentation

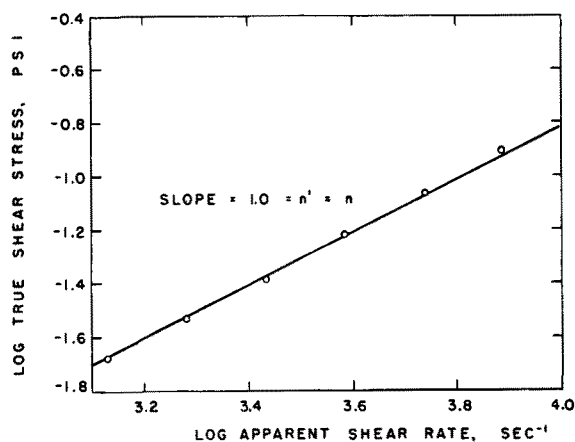These curves were constructed with data obtained

from an experiment on Flexol 8N8,[3] a fluid of known viscosity. All measurements were made at 23.4C. The shearing rates used ranged from 1,000 to 8,000 sec^{-1}. Two sets of five capillaries were used; one set had a radius of 0.01161 in., the other a radius of 0.01035 in. They ranged in length from 4.6 to 7.3 in. and in length/diameter ratio from 200 to 1 to 350 to 1. In this figure it may be observed that the measured stress increases with capillary length/diameter ratio, which is equivalent to saying that the measured load is capillary-length dependent. Both the measured loads and the slopes of the curves increase with increased shearing rate. It will be noted that the curves are straight lines and that they pass through the origin, indicating that the end effect is negligible.

Figure 4 is a flow curve of log-true shearing stress, S_T, vs. log-apparent shearing rate, D_A. From the slope of this plot n' is obtained. The value of n' for Flexol 8N8 at 23.4C is 1.0. This indicates that the fluid is Newtonian and that the apparent calculated shearing rate is also the true shearing rate. The true shearing rate is calculated by using the Rabinowitch correction equation (14).

$$D_w = [(3n' + 1)/4n'] \cdot D_A \qquad [6]$$

It is apparent that when n' = 1, D_w equals D_A. The value of K can now be calculated from the logarithmic form of the empirical power law.

$$\log K = \log S_T - n \log D_w \qquad [7]$$

For Flexol 8N8 the value of K is 154×10^{-7} (lb-force) (sec)/(in.)^2 or 106 centipoises. This compares favorably with the Ostwald value of 105.1 centipoises for Flexol 8N8 at 23.4C. Figure 5 is a linear plot of true shearing stress *versus* true shearing rate for the same data. The curve is a straight line, and it passes through the origin, which is characteristic of a Newtonian fluid. These results are offered as proof of the validity of the instrumentation techniques and experimental procedure for this capillary rheometer.

Application of Techniques to Lard Sample

This rheometer and these techniques and experimental procedures were then applied to a lard sample. Since excessive handling would cause melting or entrapping of air when the lard is loaded into the rheometer barrel, the following special loading procedure was used.

The lard sample was a standard supermarket product purchased in pound blocks. For convenience of

[3] Union Carbide Chemicals Company plasticizer; 2-2'(2-ethylhexanamido)-diethyl di 2-ethylhexoate.
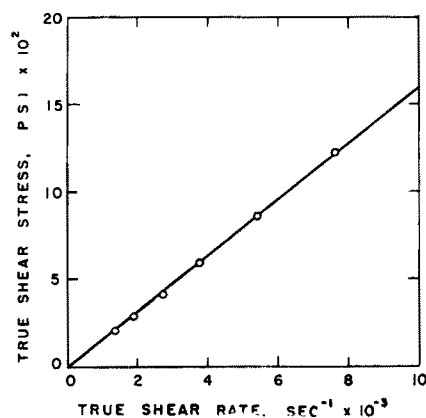

FIG. 4. Flow curve for Flexol 8N8.

FIG. 5. Linear plot of true shear stress vs. true shear rates for Flexol 8N8.

handling the pound block was first cut lengthwise into two portions. Next a steel tube 1.003 I.D. × 1.75 in. long with a wall thickness of ~0.03125 in. was used to core out a sample. A 0.250-in. piece of plate glass was found to be a good work base. The coring tool and sample were removed from the lard block with a twisting, sliding motion; the coring tube was always kept in contact with the glass base and always vertical. The rheology barrel was telescoped into the coring tube top first; care was taken to prevent the sample from extruding between the bottom edge of the coring tube and the glass base. By the time the rheology barrel was fully telescoped into the coring tube, the sample had extruded in the rheology barrel, filling it completely, including the threaded portion. Again, with a sliding motion, the loaded barrel was removed from the glass base. After sliding off the coring tube, the barrel, still in an inverted position, was placed in a special vise, which, in addition to holding it while the capillary was screwed on, also prevented the sample from extruding out of the top of the barrel. Since there was more than enough sample in the barrel, screwing on the capillary caused excess lard to extrude through it. The barrel was then ready for insertion in the compression bridge. The sample was permitted to reach "thermal equilibrium" in the rheology barrel before testing.

In the lard experiment which follows, both the capillary length and radius were varied; this experimental procedure was employed to provide further proof that the rheological measurement is completely independent of capillary geometry once steady flow has been achieved.

When capillaries of mixed geometry (both length and diameter are varied) are used, the experimental force-trace points are referred to an equivalent shearing rate by first plotting (Fig. 6) log-measured load $L_{M(CR)}$ against log-apparent shearing rate D_A (12). Then, choosing arbitrary shearing rates, the measured load-points corresponding to these rates are determined from the plot. The data produce a family of parallel straight lines, indicating no capillary effect on flow rate other than the change in radius. This procedure effectively ties in the measured force-trace points from a mixed capillary geometry with equivalent shearing rates.

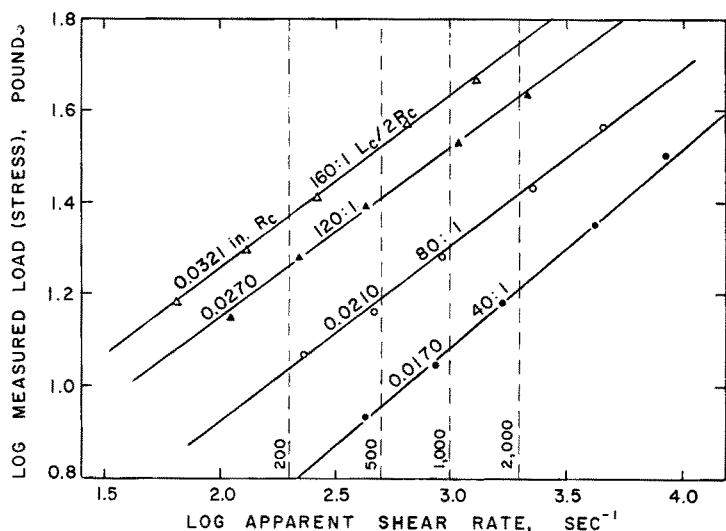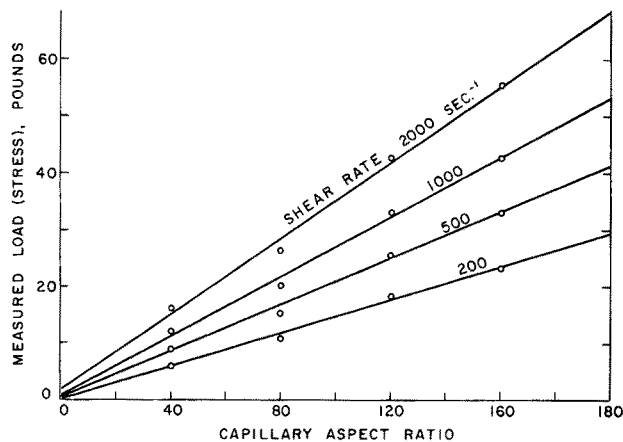Figure 7 is a plot based on the values of corrected

FIG. 7. Flow curve constructed from the arbitrary equivalent values of shear stress and shear rate obtained from Figure 6.

measured load versus capillary length/diameter ratio which corresponds to the aforementioned arbitrary shearing rates. This plot yields a series of straight lines, indicating the validity of the procedure used. Again, as with the Newtonian fluid previously discussed, the measured stress increases with capillary length/diameter ratio and shearing rate, and, as before, the slope of these curves is directly related to the true shearing stress S_T at the wall of the capillary.

Figure 8 is a plot of log-true shearing stress S_T vs. log-apparent shearing rate D_A for the lard sample. The value of the slope, n′, of this curve is 0.36 as compared with a value of 1.0 for the Newtonian fluid previously discussed. With this value n′ it is possible to obtain the true shearing rate D_w of the fluid by multiplying the apparent rate D_A by the Rabinowitch correction factor $(3n′ + 1)/4n′$. Having determined the true shearing rates, another log vs. log plot of the data is made. This plot is shown in Figure 9, and from it is obtained a value for n equal to 0.36, which makes n′ equal to n. This is true for many non-Newtonian fluids reported in the literature (14). The value of K can now be calculated, as was done previously with the Newtonian fluid by using Equation 7. K equals 2.46 × 10⁻² (lb-force) (sec)/in² or 1.69 × 10⁵ centipoises.

The rheological parameters, flow-behavior index, and fluid consistency index needed to describe the flow characteristic of lard have been determined for the first time. The fact that the flow behavior index, n, of the lard sample studied was only 0.36 indicates a fluid system which is highly non-Newtonian in character. The other parameter, fluid consistency index, K, describes its "thickness" or "viscous" character by means of a single number. Previous descriptions refer to "apparent" viscosity numbers, which have no meaning except for a set of special conditions con-

FIG. 6. Method of referring the measured load obtained from capillaries of mixed geometry to the same arbitrary shearing rate. Lard at 23.4C.
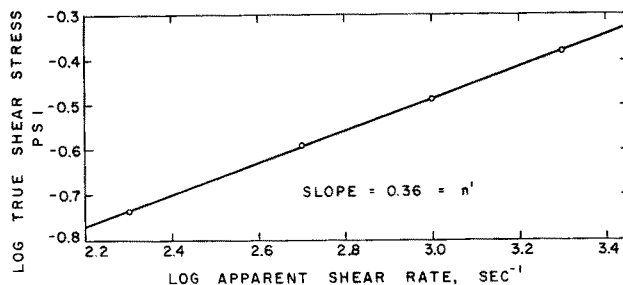

FIG. 8. Flow curve based on true shear stress vs. apparent shear rate for lard at 23.4C.
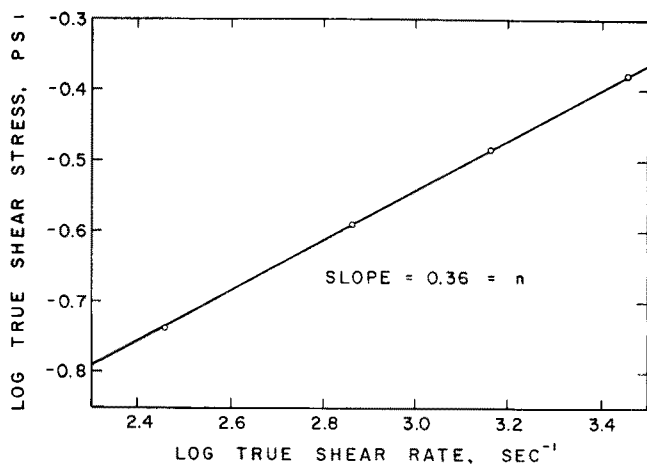
FIG. 9. Flow curve based on true shear stress vs. true shear rate for lard at 23.4C.

cerning instrumentation and mathematical calculations.

Figure 10 is a linear plot of true shearing stress S_T vs. true shearing rate, D_w. As seen, the non-Newtonian nature of lard is quite obvious. The two parameters just discussed provide a means for describing this curve and permit a calculation of the true shearing stress for any selected shearing rate.
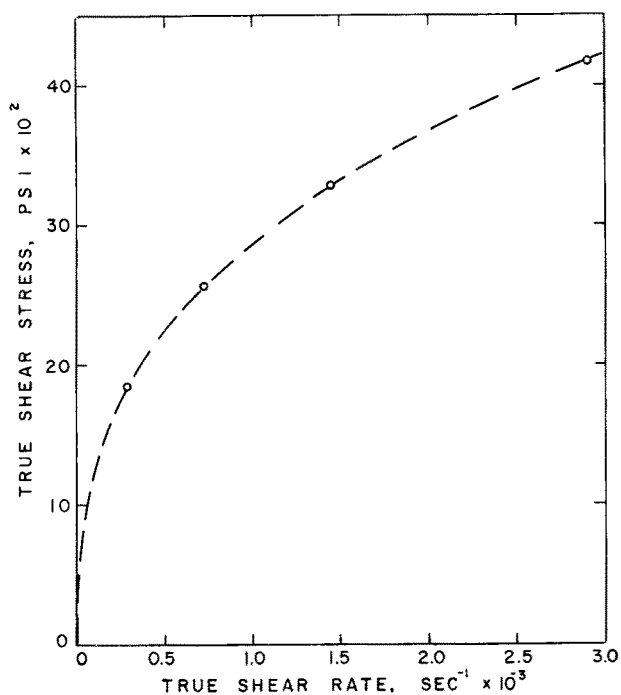

FIG. 10. Linear plot of true shear stress vs. true shear rate for lard at 23.4C, based on fluid parameters n and K.
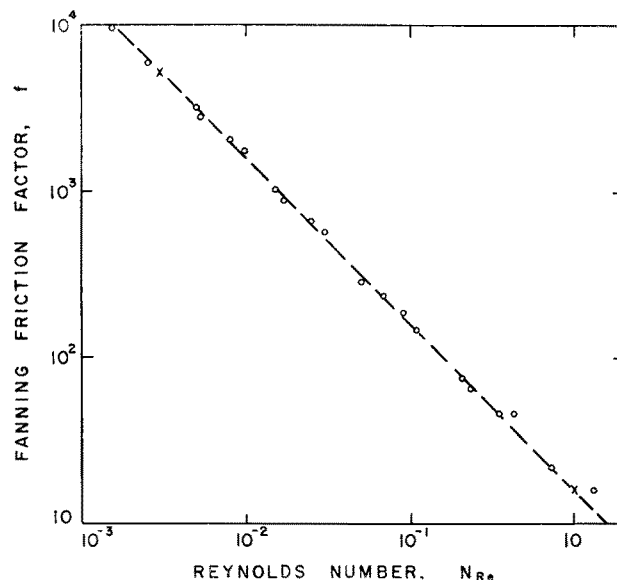

FIG. 11. Friction factor vs. Reynolds number correlation for non-Newtonian fluids. Thixotropy criterion.

The question of time-dependent viscous effects, or thixotropy, of the lard sample was answered by subjecting the data to a test for such effects worked out by Metzner and Reed (14). This test consisted of calculating a Fanning Friction Factor, "f" $f = (\Delta PR_c/2L_c)/(\rho V^2/2g_c)$ and a generalized Reynolds Number "N_{Re}" $N_{Re} = (R_c{}^{n'}V^{2-n'})(2\rho/g_cK'4^{n'-1})$ for each data point and plotting the same. If the sample was nonthixotropic, these points fell within experimental error on a theoretical curve, based on the relationship $f = 16/N_{Re}$. The lard sample did not exhibit thixotropic behavior during its flow through the capillaries used (Fig. 11).

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