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ORIGINAL CONTRIBUTION

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Imperfect lubricated squeezing flow viscometry for foods

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Abstract Commercial mayonnaise and mustard samples placed in a wide, shallow Teflon container were compressed by a wide Teflon plate to induce an 'imperfect' lubricated squeezing flow. A dominant squeezing flow regime could be clearly identified as a linear region in the log $F(t)$ vs log $H(t)$ relationship, $F(t)$ and $H(t)$ being the momentary force and specimen height respectively. The slope of the relationship enabled the estimation of the flow index, n, and the consistency coefficient K. The n values of the mayonnaise were on the order of 0.6–0.85 and those of the mustard about 0.7. The corresponding K values were on the order of 6–13 and 4–5 kPas^n respectively. Considering the crudeness of the array the measurements were highly reproducible and sensitive enough to detect differences (mayonnaise) or establish similarities (mustard) in products of different brands. The calculated flow index was practically independent of the plate's radius and of the consistency coefficient, which had a weak dependency on the latter. The calculated elongational viscosity vs biaxial strain rate relationship could also be used to compare the different products and brands. At 0.01 s^{-1} the elongational viscosity of the maynonnaise was on the order of 150 kPas, and of the mustard 60 kPas.

Key words Lubricated squeezing flow – elongational viscosity – slip – mayonnaise – mustard

Introduction

Most, if not all, semi-liquid food materials have the tendency to slip when tested in conventional viscosimeters – a problem that can cause a considerable and unacceptable error in their rheological properties determination. There are, of course, methods to correct for slip effects. But, they require either a modification of the sensor surfaces, which is relatively simple, or, and what constitutes the ultimate test of any correction method, that the measurements be repeated with sensors of different surface finish and/or geometry. Such procedures are impractical for routine quality control, and any industrial application where a large number of samples are involved. Moreover, since in most foods the sample has to "rest" in the sensor for a considerable time before a meaningful measurement can be taken, the use of expensive, accurate viscosimeters for routine applications becomes unattractive for economic reasons.

To add to the problem, many foods have rheological properties that are not only non-Newtonian but also time and shear history dependent. Thus, the mere insertion or pressing of the sample into the narrow gap of the viscosimeter's sensor can significantly affect the measurement outcome. This is a particularly serious problem when the textural changes in the specimen are irreversible, or when it takes a very long and usually unknown time to restore the original microstructure. The supposedly obvious solution to this problem is to produce the material in the viscosimeter's sensor itself, but in most cases this is not a viable option for variety of technical reasons and cost-related considerations.

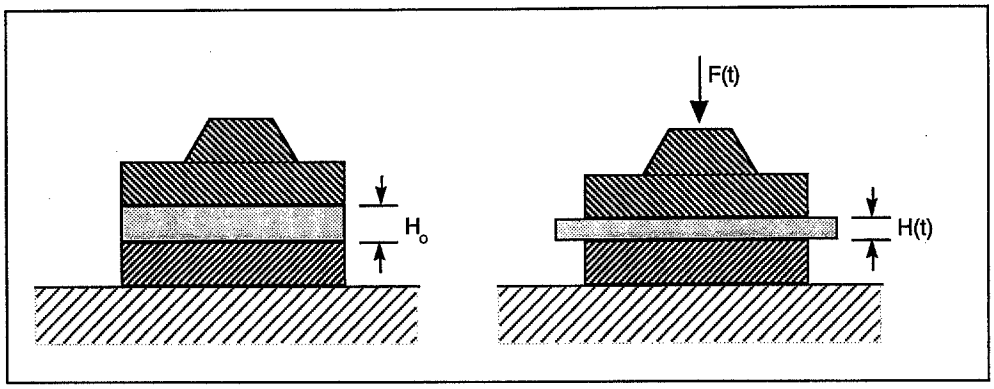

Fig. 1 Schematic view of a perfect squeezing flow sensor

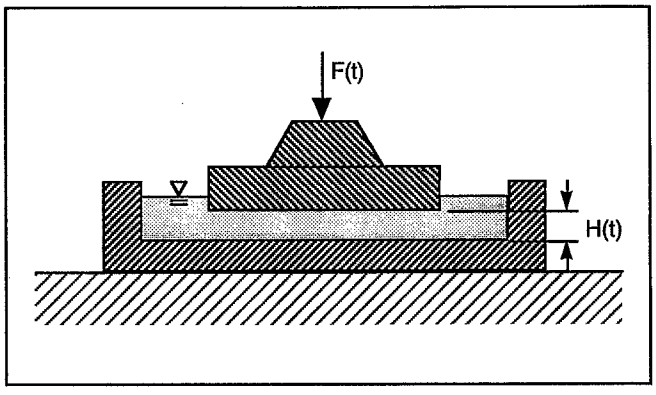

Fig. 2 Schematic view of an imperfect squeezing flow sensor

A potentially feasible solution to most, if not all, of these problems is the lubricated squeezing flow viscosimetry described by Chatraei et al. (1981) and Soskey and Winter (1985). Since it is basically a compression test (Fig. 1) the measurements can be taken with any sensitive enough universal testing machine or a creep tester. Either can be a much less expensive alternative to an accurate viscosimeter. Preparation of a food specimen for proper squeezing flow viscosimetry, although possible (Casiraghi et al., 1985; Campanella and Peleg, 1987; Campanella et al., 1987; Bagley et al., 1990; Huang and Kokini, 1993), remains a problem. This is because forming an undisturbed flat disk with controlled dimensions from a semi-liquid material is not an easy task. The difficulty can be bypassed by using what will be called "imperfect lubricated squeezing flow viscosimetry" (Fig. 2). It differs from the original squeezing flow array in that the bottom plate of the sensor is replaced by a wide flat container (Lee and Peleg, 1992).

In such a geometry the flow is "imperfect" in the sense that the pressed fluid does not exit to the atmosphere, and hence buoyancy can play a role (Damrau and Peleg, 1997). There are also entry and end effects, and annular flow in the gap between the upper plate and the container wall (see below). Thus, the imperfect array shown in Fig. 2 is a compromise configuration where the accuracy of a perfect geometry is traded for convenience and simplicity.

But moreover, since, at least in principle, specimens can be formed in separate containers each being filled directly in the production line, the method also enables testing a large number of practically undisturbed specimens. Also, since many food products are notorious for their variable consistency, the loss of accuracy that is caused by the mentioned secondary effects need not always be a serious handicap (see below). The objectives of this work are to demonstrate the concept of imperfect squeezing flow viscosimetry and to test the method with selected food products.

Theoretical background

The principles of squeezing flow of polymers have long been established (Leider, 1974; Leider and Bird, 1974; Chatraei et al., 1981; Lee et al., 1982; Soskey and Winter, 1985). It has two basic geometric configurations – constant area and changing volume, the one shown in Fig. 1, and constant volume and changing area which will not be discussed. The flow regime is determined by whether the plates are lubricated to eliminate friction (see below) or are non-lubricated to induce shear. In both cases when the tests are performed at a constant displacement rate the source of information is the force vs height relationship. When the tests are performed under a constant load or stress (creep) it is the height-time relationship which provides the information. This test configuration will not be further discussed.

For squeezing flow measurements to be valid the radius to height ratio has to be larger than about 10 to avoid end effect (Lee et al., 1982). The main differences between the lubricated and non-lubricated flows are the overall magnitude of the generated forces, and the nature of their dependency on the specimen height. For a Newtonian liquid with a viscosity, μ, squeezed at

a constant displacement rate, V, the momentary force $F(t)$ in ideal frictionless plug flow is:

$$F(t) = 3\pi\mu R^2 V/H(t), \quad (1)$$

while in an ideal frictional flow without slip (non lubricated) flow it is given by Stefan's equation:

$$F(t) = (3/2)\mu R^4 V/H(t)^3 \quad (2)$$

where R is the specimen's radius and $H(t)$ the momentary specimen height.

For a pseudoplastic liquid with a consistency coefficient, K, and a flow index, n, Eqs. (1) and (2) are replaced by:

$$F(H(t)) = \pi R^2 K^{3(n+1)/2} (V/H(t))^n \quad (3)$$

and

$$F(H(t)) = 2\pi K R^{n+3} V^n \cdot [(2n+1)/n]^n/[(n+3)H(t)^{2n+1}] \quad (4)$$

respectively.

The rheological constants, μ, K and n, can be determined from the plot of log $F(t)$ vs log $H(t)$. In an ideal lubricated flow the slope of the resulting line is –1 if the fluid is Newtonian, and $-n$ if ist is a power law liquid. In an ideal frictional (non-lubricated) flow it is –3 if the fluid is Newtonian, and $-(2n+1)$ if it is a power law liquid (see below).

In an ideal frictionless lubricated flow one can also calculate the elongational biaxial viscosity (μ_b as a function of the biaxial strain rate, $d\varepsilon_b/dt$ (Chatraei et al., 1981; Casiraghi et al., 1985; Campanella et al., 1987; Bagley et al., 1990; Huang and Kokini, 1993) i.e.:

$$\mu_b(t) = 2F(t)H(t)/(\pi R^2 V) \quad (5)$$

where $d\varepsilon_b/dt = V/[2H(t)]$. The elongational viscosity is probably the most relevant rheological property that controls the "spreadability" of food products (Casiraghi et al, 1985; Campanella et al., 1987) and hence should be considered as a prime textural quality factor.

Imperfect squeezing flow

Equations (1–5), as already mentioned, have been developed for ideal flow regimes based on the premise that the pressed liquid exits to the atmosphere. This condition is clearly violated in the imperfect squeezing flow array shown in Fig. 2. In this case the fluid exists against an increasing hydrostatic pressure produced by the rising fluid level in the annulus. If the fluid rise is uniform, this pressure, or buoyancy effect, can be described by (e.g., Steffe and Osorio, 1987; Damrau and Peleg, 1997)

$$F_B(H(t)) = \pi\rho g[H_0 - H(t)] R^2 R_{cont}^2/(R_{cont}^2 - R^2) \quad (6)$$

where $F_B(H(t))$ is the momentary buoyancy force, H_0 and $H(t)$ the initial and momentary specimen heigt respectively and R and R_{cont} the upper plate and container's radius respectively, and ρ the fluid density. The relative magnitude of the error produced by the buoyancy effect diminishes as the viscosity or consistency of the tested liquid increases. It can also be reduced by using a sensor with a relatively wide gap, and a very shallow specimen with an aspect ratio, higher than the minimal required in a perfect squeezing flow experiment. It can be shown that with semi-fluid foods (whose density rarely exceeds 1200 $kg \cdot m^{-3}$) tested with sensors of the geometry used in this work the buoyancy effect is only on the order of 0.1 N per mm displacement (Damrau et al., 1997). Thus although buoyancy is a source of an error, it need not be a critical one if the generated forces are comparatively large (see below). The same can be said about the induced annular flow. With a gap on the order of 10 mm or more and a length of only a few mm the pressure gradient is negligible. Consequently, the role of the annular flow as a source of an error can be safely ignored as long as the gap is not too narrow.

The effects of entry and a subsequent transient flow regime are not easy to quantify. Their existence is evident in the smooth shape of the flow curve as shown in Fig. 3. It can be argued that their role progressively diminishes as the specimen height decreases and the overall force increases. The range of heights, where squeezing flow is the dominant regime, can easily be identified from the log $F(t)$ vs log $H(t)$ plot as shown in Fig. 3 (Lorenzo et al., 1997). It has a characteristic linear or almost linear region as shown in the figure (bottom, right).(The slight curvature at the left end of this region is an artifact – see below.) The slope of the relationship in the linear region is indicative not only of the tested fluid properties, but also of whether the flow can be considered as "fully" lubricated – a condition for the calculation or estimation of the sample's elongational viscosity.

Materials and methods

Materials

Jars of commercial mayonnaise and mustard, of two national brands each, were purchased at a local supermarket and were tested soon after their container had been opened. Since no attempt was made to establish how representative they were of their respective manufacturer product they are only identified by a code letter.

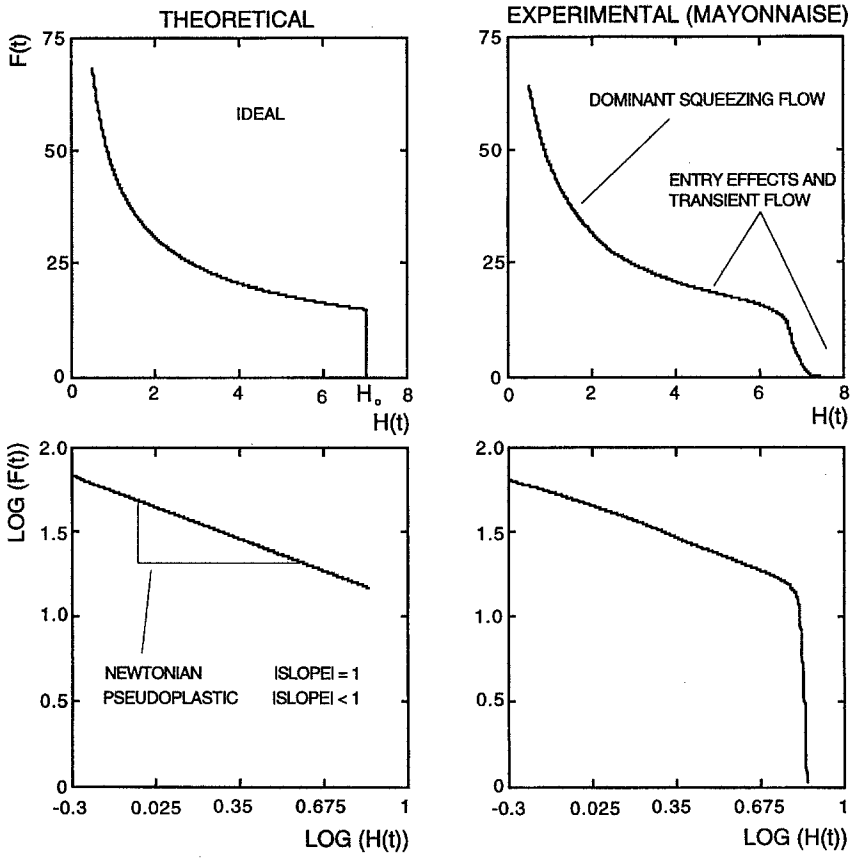

Fig. 3 Ideal flow curves in perfect lubricated squeezing flow (right) and experimental flow curves in imperfect lubricated squeezing flow (left)

Mechanical testing

In a set of preliminary tests the products were poured into wide Petri dishes 140 mm in diameter, on the bottom of which there was a thin metal disk 120 mm in diameter with a polished or grooved surface. The liquid's initial height was about 6 mm, and it was compressed with a metal plunger (upper plate) 120 mm in diameter having a matching surface finish, polished or grooved respectively.

In the main tests the products were poured into a smooth Teflon container 140 mm in diameter (Fig. 4), also to an initial height of about 6 mm. They were subsequently compressed using a Teflon plunger having a diameter of 120 mm. Additional 100 and 64 mm Teflon plungers were used to evaluate the role of the upper plate radius.

The compression tests were performed with a TA.TX2 Texture Analyzer, a table top universal testing machine (Texture Technologies Corp., Scarsdale, NY) interfaced with a Gateway 2000 microcomputer. A displacement rate of 0.1 mm min^{-1} was used in all the experiments with a data retrieval rate of 10 Hz. The digitized date file so created was imported to and processed by the Systat 5.0 package (Systat, Inc., Evanston, IL). Each squeezing flow test was performed in four or five replicates unless otherwise stated.

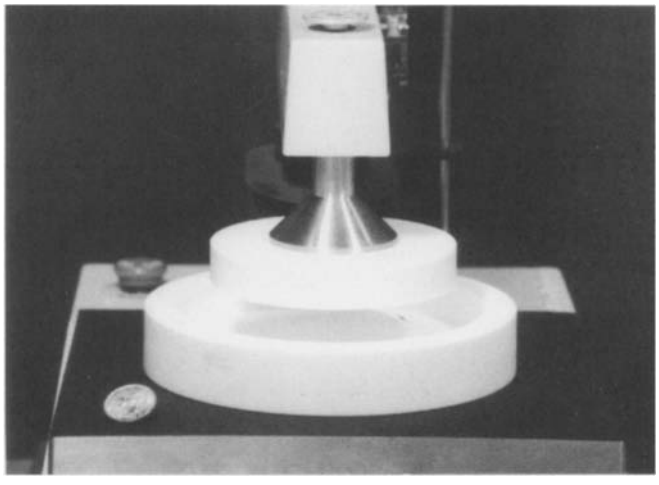

Fig. 4 All-Teflon sensor for imperfect lubricated squeezing flow viscosimetry

Results and discussion

Preliminary results

Typical flow curves of mayonnaise and mustard compressed between grooved metal plates are shown in Fig. 5. The curves were highly reproducible despite the system's crudeness. The slight curvature of the left part of the curves is a result of imperfect parallel orientation of the plates and a small compliance of the array. Similar flow curves were recorded when the metal plunger and bottom plate both had a polished surface. These curves also had the same degree of reproducibility. The shape of the plots shown in Fig. 5 demonstrates that despite the system's imperfections the region of a dominant squeezing flow could easily be identified. The same was observed in three different tomato products tested with the same sensor (Lorenzo et al., 1997). Although in this region the log $F(t)$ vs log $H(t)$ relationship was not perfectly linear – primarily because of the imperfect construction of the array – its average slope could still be determined by linear regression. In both products the mean absolute magnitude of the calculated slopes was on the order of 0.6–0.8. Since theoretically the absolute magnitude of the slope in frictional squeezing flow cannot be smaller than unity,

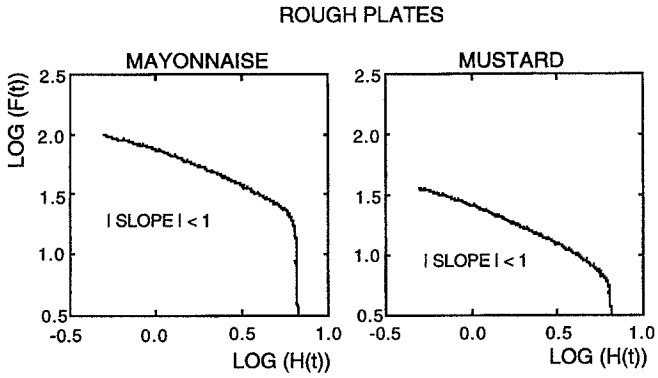

Fig. 5 Typical flow curves of commercial mayonnaise and mustard squeezed between grooved plates. Note that the slope indicates total slip despite the rough surfaces

even with a flow index $n=0$, one must conclude that the tested mayonnaise and mustard provided enough self-lubrication to overcome the effect of a rough surface. Support for this condition comes from the flat front of the flow which could be visually observed when the liquid was squeezed between the two plates. Nevertheless, in order to avoid a partial slip and reduce the role of the other artifacts, all subsequent experi-

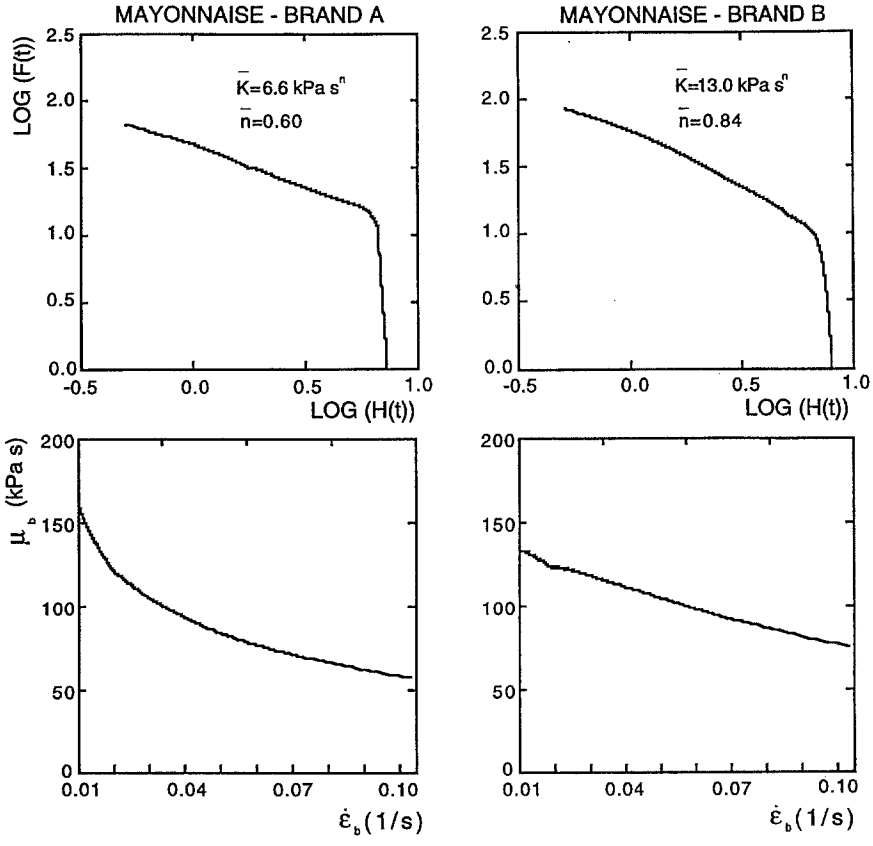

Fig. 6 Typical flow curves and elongational viscosity vs biaxial strain rate relationships of commercial mayonnaise squeezed in the all-Teflon sensor shown in Fig. 4

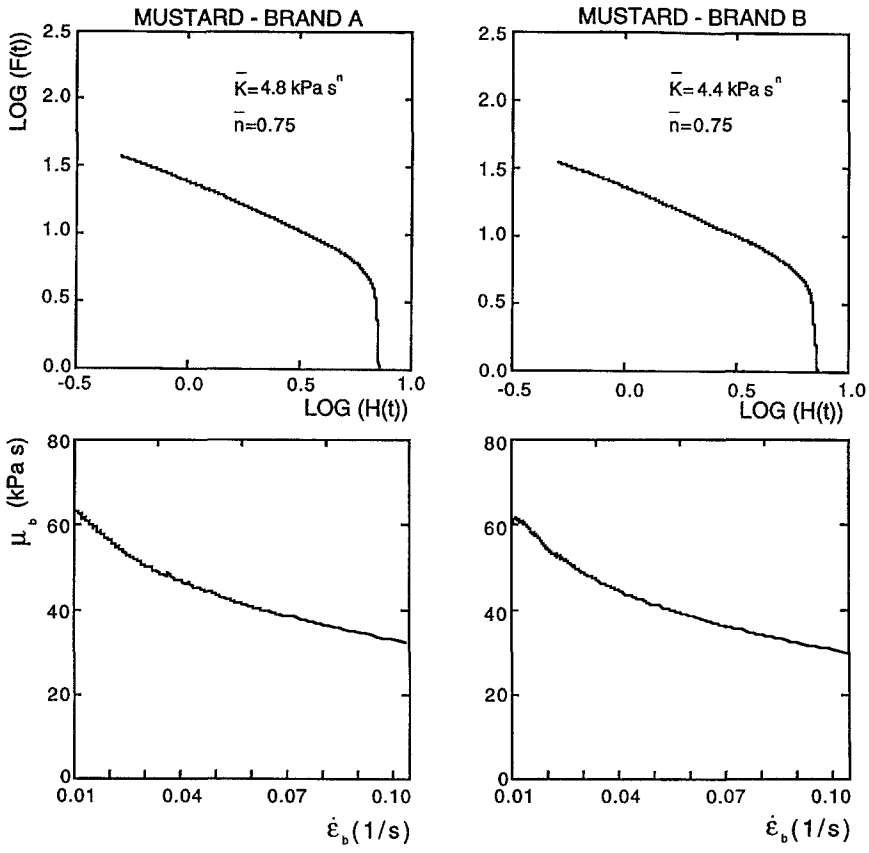

Fig. 7 Typical flow curves and elongational viscosity vs biaxial strain rate relationships of commercial mustard squeezed in the all-Teflon sensor shown in Fig. 4

Table 1 Rheological parameters of commercial mayonnaise and mustard determined by imperfect squeezing flow viscosimetry (Container's and upper plate's radius 70 and 60 mm, respectively)

Product	Consistency – K (kPa s^n)		Flow index – n (–)		Elongational viscosity – μ_b (kPa s), at $\dot{\varepsilon}_b = 0.01\ s^{-1}$		Elongational viscosity – μ_b (kPa s), at $\dot{\varepsilon}_b = 0.05\ s^{-1}$	
	Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation
Mayonnaise Brand A	6.6	0.4	0.60	0.03	152	8	82	2
Mayonnaise Brand B	13.0	1.4	0.84	0.03	130	4	105	7
Mustard Brand A	4.8	0.4	0.75	0.02	63	2	44	2
Mustard Brand B	4.4	0.2	0.75	0.01	59	1	41	1

ments were performed with the more robust all-Teflon sensors (Fig. 4).

Imperfect lubricated squeezing flow

Typical flow curves of mayonnaise and mustard recorded with an all Teflon sensor are shown in Figs. 6 and 7 (top). The sensor used had the same Teflon container, 140 mm in diameter, and an upper plate of 120 mm in diameter with a corresponding gap of 10 mm. The calculated rheological parameters, K and n, of the two products, using Eq. (3) as a model, are listed in Table 1. It demonstrates the reproducibility of the results, which in light of the sensor's and the procedure's simplicity can be considered as quite satisfactory. The

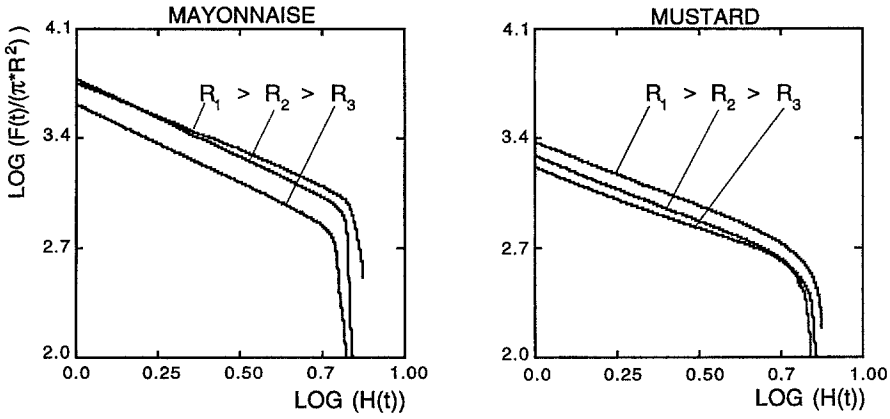

Fig. 8 Effect of the upper plate radius on the flow curves of mayonnaise and mustard squeezed in the all-Teflon sensor shown in Fig. 4 (R_1=60, R_2=50, R_3=32, $R_{container}$=70 mm)

Table 2 Effect of the upper plate radius on the calculated values of the rheological parameters of commercial mayonnaise and mustard determined by imperfect squeezing flow viscosimetry (Container's radius 70 mm)

Radius (mm)	Mayonnaise		Mustard	
	Consistency – K (kPa s^n)	Flow index – n (–)	Consistency – K (kPa s^n)	Flow index – n (–)
60	15.0	0.86	5.6	0.82
50	19.5	1.0	4.8	0.85
32	13.5	1.0	3.6	0.78

results also indicate that time effects and/or any changes produced during the transfer of the products into the container were negligible factors. The same can be said about the buoyancy force which was on the order of 1% of the total force. The method, despite its crudeness, was sufficiently sensitive to distinguish between the rheological properties of the two mayonnaise brands. The two mustard brands were clearly different from those of the mayonnaise of either brand. The very same general results were obtained with the metal sensors irrespective of whether the plates were smooth or rough. The observed reproducibility of the results and the method's sensitivity to rheological differences is in agreement with previous findings (Lee and Peleg, 1992; Damrau and Peleg, 1997; Lorenzo et al., 1997), and it allows expressing textural differences in terms of the estimated magnitude of established rheological parameters. The pertinent region of the flow curves was also converted into biaxial elongational viscosity vs biaxial strain rate relationship using Eq. (5). Typical results are also shown in Figs. 6 and 7 (bottom). Their general shape is very similar to that of other spreadable food products as obtained by a "perfect" array (Casiraghi et al., 1985; Campanella et al., 1987). The μ_b vs $d\varepsilon_b/dt$ relationship provided additional rheological parameters for comparison between the brands – the apparent elongational viscosity at a selected strain rate or rates. The actual values are listed in Table 1, and as could be expected, the differences that they indicate, or their absence thereof, are consistent with those calculated directly from the original flow curve, namely the consistency coefficient K and the flow index n.

Effect of the upper plate diameter

Normalized experimental flow curves, recorded with all-Teflon sensors having an upper plate of a diameter 120, 100 and 64 mm are shown in Fig. 8. (The corresponding gaps were 10, 20 and 38 mm, respectively.) They were computed by dividing the recorded force by the corresponding upper plate's area. As could be expected, the higher the diameter, the higher the flow curve, even after compensation for the plate's cross-sectional area, resulting in a higher value of the calculated K. The actual values, the mean of two replicates, are listed in Table 2. The table and Fig. 8 demonstrate that the magnitude of n was only slightly affected by the upper plate's radius. The dependency of K on the plate's area reflects the diminishing role of end effects as the latter increases. However, since for practical reasons the plate's diameter cannot be increased indefinitely, the values obtained by the method, even with a relatively wide plate, should be considered as low estimates of the true values. Obviously, if the method is only used for comparison, then using a single plate – wide as practically possible – will suffice. (An increase of the plate's diameter also increases the risk of a tilt

which can introduce an additional source of error. Hence, the 120 mm plate appears to be close to an optimal choice.) The same applies to the calculated elongational viscosity vs biaxial strain rate relationship. It too is affected by the plate diameter, but it can still be used as a consistent tool to detect rheological differences between food products or establish similarities between them.

Conclusions

Lubricated imperfect squeezing flow viscosimetry offers a simultaneous solution to two major problems that are encountered in conventional viscosimetric methods when applied to semi liquid food materials, namely uncontrolled slip and structural changes in the specimen during its insertion into the narrow gap of the rheometer's sensor. As shown, materials like mayonnaise and mustard provide enough self-lubrication that equations developed for frictionless squeezing flow can be used to estimate their rheological constants. Although buoyancy, entery effects, geometric imperfections and artifacts can distort the shape of the flow curve – a region of a dominant lubricated squeezing flow regime can be clearly identified when the data are plotted as a force vs height relationship in logarithmic coordinates. This region yields rheological parameter estimates such as the flow index, consistency coefficient and elongational viscosity (at a range of biaxial strain rates) which are reproducible and sensitive to detect textural differences and similarities between products of different brands. In principle, the role of the imperfections diminishes as the sensor diameter increases. However, the latter cannot be enlarged indefinitely for practical considerations including the danger of a tilt. It appears that at least for the materials tested a sensor with a plate diameter on the order of 100 mm and a gap of 10 mm or more is a practical compromise.

Because the main objective of the work was to test the methodology, the sensor – upper plate and container – was made of solid Teflon. For routine testing of a large number of samples, especially if collected directly in the production line, a less expensive container can be found or made. Although the presented results are of mayonnaise and mustard only, the method would probably be also applicable to other food and non-food materials of similar consistency.

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