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Observations on the Instrumental Measurements of Liquid Food Stickiness †

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Abstract: While we encounter sticky liquids in our daily life and are able to discriminate between them, instrumental measurements of stickiness are difficult to match to those which relate to our perception. In this paper we examine some of the factors that influence instrumental measurements of stickiness in liquid foods. Shortcomings of using the maximum peak or the area under the curve are discussed and a hitherto unused measure, the gradient of the force-distance curve is suggested as a measure of tension per unit contact area. The zero-perimeter virtual probe which compensates for the changing meniscus and mass of liquid below it, is introduced. This zero-perimeter approach allows us to extrapolate measures of stickiness such as the gradient of the force-distance curve or the area below that curve. Despite the zero-perimeter correction, there is still a speed dependency on results from instrumentally measured stickiness (for all indexes considered). The speed of the test is responsible for the type of failure (cohesive or adhesive) reported by other authors.

Keywords: stickiness; zero-perimeter virtual probe; initial gradient; maximum peak; area under the curve

1. Introduction

Stickiness is an important characteristic of many food materials and it has a major influence on numerous industrial processes. While in some products it is a desirable property for example in bonding oats together in a breakfast bar, in many it causes problems such as doughs sticking to conveyor belts or material not separating from a depositing mould. Related scientific terms include adhesiveness and cohesiveness, though again the meanings of these terms are poorly defined. This is compounded by some highly influential papers on texture measurement which have (perhaps inappropriately) attributed parts of a texture analyser curve to a particular property [1]. Ultimately understanding what contributes to stickiness is a worthy aspiration and publications such as Adhikari and co-workers [2] attempt to quantify the different forces involved, they report the study of Brennan and Mohamed attempted to correlate sensory stickiness of sugar solutions with a number of physical measures, finding viscosity and surface tension gave the best relationships.

Fiszman and Damasio [3] surveyed the variety of instrumental food stickiness tests illustrating the diversity of measures which researchers have used. The normal approach taken is to press a probe on to the food surface and then pull it away while measuring the force. The two most widely used measures being the area under force-time/distance curve and the peak force to detach the probe from the surface [3].

Possibly as a result of the engineering limitations of the early texture analysers, many of the publications on stickiness measurement deal with force-time curves. Hoseney and Smewing [4] show the effect of varying the withdrawal speed on the force required to pull a probe away from a surface (Figure 1).

When a probe is slowly raised up as if to try and separate it from the surface of a sticky liquid, a column of liquid clings on below. Close to the liquid surface the meniscus of the liquid spreads out beyond the perimeter of the probe and the force pulling downwards on the probe is due to the mass of liquid held above the plane liquid surface. Moreover as the separation of the probe from the plane surface increase, the shape of the meniscus changes as the probe rises from the surface [5].

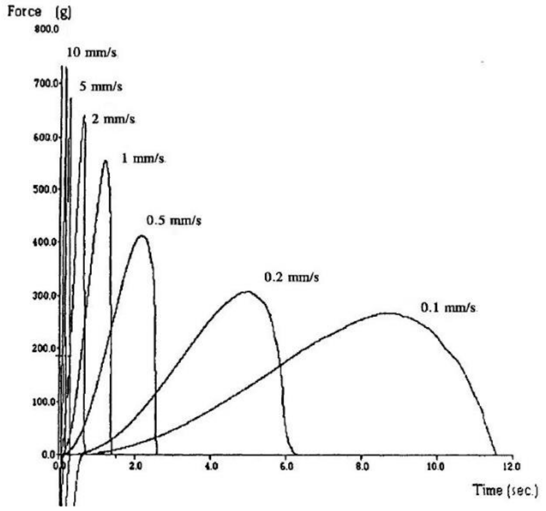

Figure 1. typical force time curves of stickiness at different withdrawal speed (adapted from [4]).

The aim of this work was to better understand the forces and factors involved in the instrumental measurement of stickiness. We aimed to quantify the forces involved in the stickiness of syrups with the hope that lessons learned might be extended to other materials.

2. Materials and Methods

2.1. Sample Materials

Several sticky proprietary syrups were used for tests Golden Syrup (Tate & Lyle, London, UK), Black Treacle (Tate & Lyle, London, UK) and clear honey (Rowse, UK). Syrups were poured into plastic petri dishes, clamped on to the base of the texture analyser and tested as below.

2.2. Texture Analyser and Probes

Stable Micro Systems (Godalming, UK) manufactured a series of three bespoke acrylic probes. One had a single head, but the other two were multi headed with three and six heads all milled out of an acrylic block. The contact surfaces of all the probes were on the same plane.

The geometries of the three probes are given in Table 1.

Table 1. Dimensions of the multi-headed probes.

Number of Heads	Diameter of Heads (mm)	Total Perimeter (mm)	Total Contact Area (mm^2)
1	35.0	110	962
3	20.0	190	962
6	14.0	269	962

A TA.HD texture analyser (Stable Micro Systems, Godalming, UK) was used with a 5kg load cell. The TestMaker application (Stable Micro Systems, Godalming, UK), was used to write a sequence whereby the probe was brought into contact with the liquid surface, remember that position then push 0.3 mm into the material followed by a 10 s pause to try to allow the liquids to achieve good contact. The probe was then pulled back to the remembered position and there then followed a further 2 min pause. The probe was then withdrawn from the surface at a defined speed until detachment was achieved. Photographic images of the probe liquid contact were taken with an iPhone 7 plus.

3. Results and Discussion

The spread of the peaks in Figure 1 is resolved by transposing the horizontal time-axis to distance. Figure 2 has the same experimental design as undertaken by Hoseney and Smewing [4], with a range of separation speeds which almost span the capability of texture analyser used. In Figure 2 we have drawn the curves as negative peaks, however throughout this discussion we will refer to the peak as the maximum force (as referring to a minimum force makes little sense).

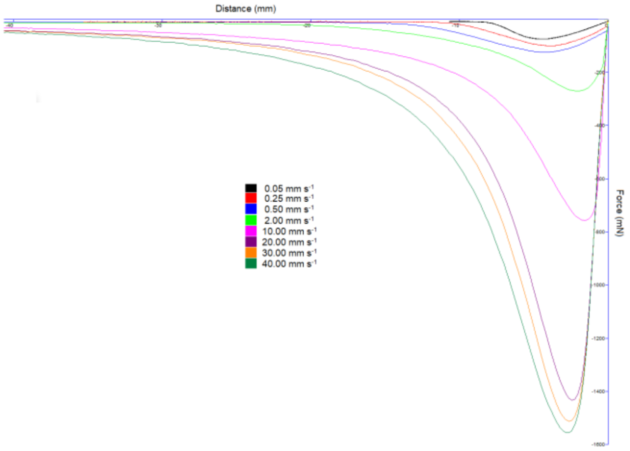

Figure 2. Typical force-distance curves depicting stickiness at different withdrawal speeds for a single headed probe.

By plotting the distance of separation, as opposed to time, on the horizontal axis we observe that the distance to the negative peak is roughly the same regardless of the speed of the probe. This is consistent with Hoseney and Smewing [4] though not at all obvious from Figure 1. In contrast the distance that the probe moves from the liquids plane surface to the maximum negative peak, is both visibly similar and intuitively the same for all probe speeds in Figure 2. In Figure 1 the areas under curves were greater for the slower speeds with the units of this quantity being Force-time. In Figure 2 the units are force-distance and the areas under the curves are greatest for the higher speeds. Moreover, if researchers assign stickiness to the area under the curve then the derived units of force-distance are energy or work (J).

Figure 3 shows a typical curve for the separation of a probe from the surface of a sticky liquid (at a relatively low speed). When separation is about 1.5 mm, the liquid is still in good contact with the edge of the probe, though the surface is developing a curvature (inset Figure 3a). Before the peak is reached, the force starts to lessen (inset Figure 3b) as the curve starts to flatten, this this is followed by a narrowing of the column of liquid joining the probe to the body of liquid below. The curvature of the glucose syrup as the probe pulls away exhibits concave necking (inset Figure 3d)—holding firm to the probe perimeter and narrowing within the liquid itself (this is shown diagrammatically in Figure 4d). In contrast the inset image with a broken line (inset Figure 3c) is for golden syrup at the peak and while the column of liquid thins, it does not hold fast to the perimeter of the probe but forms a narrowing cylinder attached to the base of the probe (this is shown diagrammatically in Figure 4b). After the peak is past, the force continues to reduce as the thickness of column of liquid progressively thins (e.g., inset Figure 3e).

Stickiness has been attributed to the interaction of liquid viscosity and interfacial properties between the probe material and the liquid [2]. Figure 3 shows a linear region at the start of probe withdrawal during which time the perimeter is fully in contact with the liquid. The inset images in Figure 3 show that the curvature of the liquid observed during these early stages of probe withdrawal. The forces acting downward on the probe are due to the surface tension, but also the mass of the liquid below the probe and within the truncated annular region beyond the cylindrical perimeter of the probe (Figure 4a). During the linear part of the force-distance curve, during the stages of probe withdrawal from the surface, the curvature of the liquid adhering to the perimeter of the probe is a complex relationship which changes as the probe progressively moves up [5].

In an attempt to compensate for the effects of curvature we have developed a practical (non-theoretical) solution which utilizes multi-headed probes with a common surface area in contact with the liquid. The dimensions of these probes are shown in Table 1. Despite having the same contact area in touch with the liquid, if we sequentially undertake the stickiness test outlined above with each of these probes, we produce three different curves which do not superimpose on each other (Figure 5). While the curves in Figure 5 are all of a similar basic shape, curve features such as: distance to reach the maximum negative peak; force at the maximum negative peak; area under the curve; gradient of the linear portion of the curve, are all slightly different. However, if we plot these features against total probe perimeter we unsurprisingly obtain good straight lines.

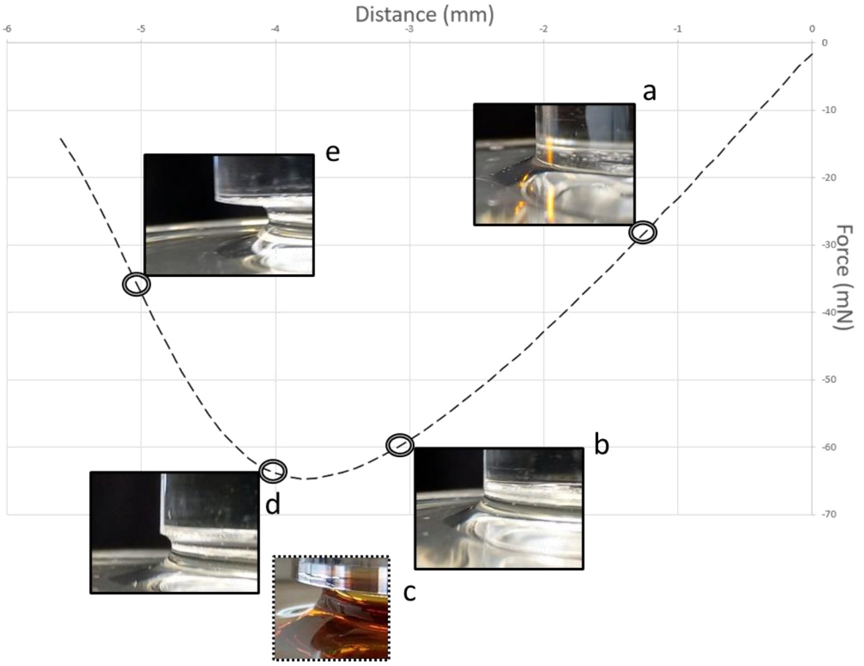

Figure 3. Force-Distance curve for glucose syrup with images of probe contact surface. the inset images from with a broken border is Golden syrup at its force-distance peak.

Furthermore, if we extrapolate such lines to zero, we effectively obtain the force we would obtain from a zero-perimeter probe. Of course such a probe does not exist, yet the force exerted on such a virtual probe would be solely due to mass of the liquid in a cylinder below a 962 mm^2 probe.

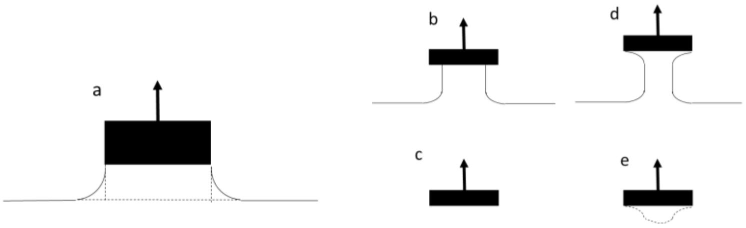

Figure 4. Schematic view of stickiness test of liquids. (**a**) starting state; (**b**) early stages of pull-linear portion of curve; (**c**) narrowing liquid column-loss of adhesion; (**d**) adhesive detachment; (**e**) narrowing liquid column-necking; (**f**) cohesive failure.

Our zero perimeter virtual probe overcomes problems with the unpredictable meniscus, however we can see from Figure 3 that strictly speaking the known contact area (962 mm^2) is only valid during the linear part of the curve and this in reality excludes the use parameters such as the peak force or the area under the curve. However, we have continued to make use of the parameters of peak force and the area under the curve as measures of stickiness albeit with our zero-perimeter curve. At least the zero-perimeter probe excludes effect of the curved meniscus and as such better defines the system.

If the purpose of a stickiness test is to determine a defined material property, then the dimensions of the geometry are important. That is to say, in an ideal world we would be able to express the stickiness per m^2. Yet once the curve begins to deviate from a linear force-distance behaviour, the sample geometry has changed. Ideally, we should obtain our measure of stickiness from the linear portion of the curve otherwise we are measuring a property of ill-defined dimensions. Certainly, during the linear part of the force-distance curve there is neither separation of the liquid from the perimeter nor necking of the sample, thus the instrumental readings obtained will relate to the contact area of the probe. Moreover, if we consider that property our zero-perimeter virtual probe we can overcome changes in the meniscus. On this basis, using a property of this linear region such as its gradient would perhaps give us a better measure of stickiness and one which can be related to the probe geometry. Appropriately the units of this gradient are force per unit distance (Nm^{-1}), in other words a tension—which is possibly a better match for stickiness than either the peak force or work used to separate the probe. At low test speeds the gradient of the linear portion of the force-distance curve is straight forward to measure, however as is apparent from Figure 2, at higher speeds and especially with the more viscous liquids, the curve is almost vertical. It is as if the liquid cannot flow or yield, with the stress exerted by the texture analyser being unable to be dissipated thereby rapidly rising. While in Figure 2 these high-speed curves appear to superimpose upon each other, if the horizontal axis is enlarged we can discriminate between them.

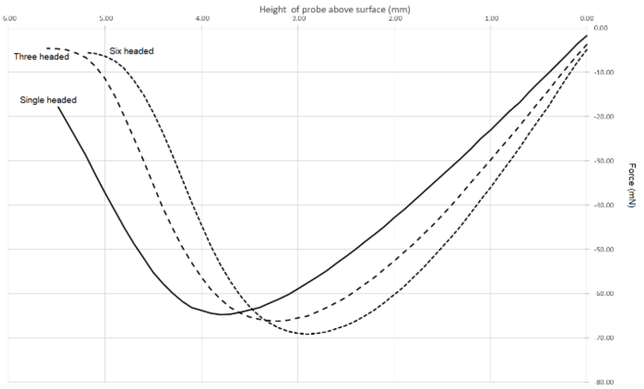

Figure 5. Force-Distance curves of for Glucose syrup with constant area (varying perimeter) probes. Probe withdrawal speed is 0.01 mm.s$^{-1.}$

Figures 1 and 2 emphasise the importance of probe withdrawal speed on the curve obtained. Making use of our multi-headed probes we have been able to estimate the various parts of the curves of our zero-perimeter virtual probe, being withdrawn from different sticky liquids at a range of speeds. Reminding ourselves that the purpose of these tests is to measure stickiness as a characteristic property of the material, we have attempted to examine the relationship between the initial gradient, the maximum negative peak force and the area under the curve of the zero perimeter virtual probe as functions of withdrawal speed.

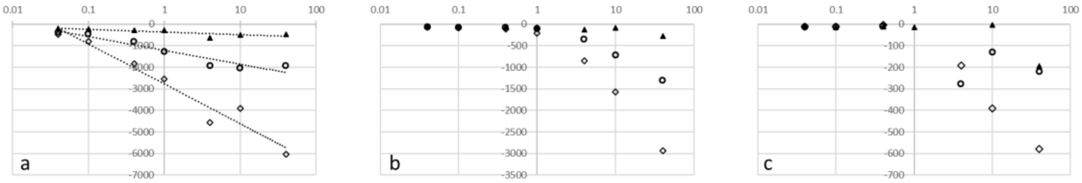

Figure 6. Influence of probe withdrawal velocity on parameters of zero perimeter virtual probe curves (**a**) Area under curve (**b**) peak force (**c**) Initial gradient **O** Golden syrup, ◇ black treacle, ▲ Honey.

To cope with the wide range of withdrawal speeds employed to collect the data in Figure 6, we have plotted the withdrawal speed on a log axis. Figures 6b and 6c appear to show a discontinuity, remaining relatively low at slow speeds and then increasing logarithmically after some critical value which is specific for each liquid.

Kilcast and Roberts [6] introduced the idea of adhesive and cohesive failure based on whether a sticky liquid leaves a residue on the probe or separates cleanly when a probe is pulled out of a sticky material. We concur with Noren, Scanlon and Arntfield [7] who observed that the cohesive/adhesive failure behaviour actually depends on the speed with which the test is undertaken. We speculate that as the probe is withdrawn from the liquid surface, at low speeds the liquid is able to flow back to the liquid bulk–allowing it to cleanly separate from the surface of the probe, progressing from Figure 4a,b and finishing at Figure 4c—i.e., adhesive failure. In contrast once we exceed the critical speed which marks the discontinuity in Figure 6b and 6c, the separating probe moves faster than allows the liquid to flow back to the bulk with the result that the probe liquid behaviour progresses from Figure 4a,d ending with Figure 4e—i.e., cohesive failure.

Earlier we comment on the almost vertical force-distance curves of the high speed tests, the idea that the material cannot dissipate the stress through flow or relaxation is consistent with stresses building within the material until that material cannot support further stretching and undergoes catastrophic failure. If the separation of the probe from the surface of the liquid is faster than allows the liquid to flow, we get cohesive failure. In this study we are dealing with viscous syrups. Many foods are glassy materials, which are often considered to be super cooled liquids. Such materials have immensely high viscosities which would be difficult to flow in the time frame of the test protocol outline here. Thus, we would expect glassy materials to undergo cohesive failure. Some other foods are viscoelastic (e.g., doughs) and in such situations we expect the failure to depend on the predominance of viscous and elastic elements present. In the case of viscoelastic materials both the ease of flow and the elastic limits of the material will dictate whether the texture analyser imposed force can be adequately dissipated in the time frame of the test or whether we will end with cohesive failure.

In our daily life we experience the phenomenon of stickiness in the liquids we interact with and perhaps the approach we should take from a physical testing point of view is to employ separation speeds akin to those employed in the manual manipulation of materials with our fingers and jaw motion. Shama and Sherman [8] used a similar approach in recommending shear rates with which to evaluate the viscosity of liquids if they are to match our human experience.

4. Conclusions

Clearly data collected to measure stickiness of liquids has a huge dependence on the speed and geometry of the test being undertaken. We might even go so far as to say the results are artefacts of the test method employed.

Our zero-perimeter virtual probe overcomes problems with the unpredictable meniscus. The probe is only in full contact with the probe during the linear region of the force distance curve and as such, perhaps the gradient, would give us a better measure of stickiness and one which can be related to the probe geometry. In contrast, while the peak force and the area under the curve are not able to relate the force to the geometry of the probe, the zero perimeter virtual probe does reduce some of the variability in results arising from the curved meniscus.

Author Contributions: S.M.K. and A.J.R. designed the experiments, analysed the data and wrote the manuscript together. A.J.R. conceived and designed the multiheaded probes. S.M.K. undertook the data collection. All authors have read and agreed to the published version of the manuscript.

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