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An alternative theory to explain the effects of coalescing oil drops on *mouthfeel*

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In this letter, we demonstrate that through non trivial rheological effects occurring in narrow gaps, it is possible to explain how coalescent oil drops contribute to the specific *mouthfeel* of such unstable emulsions. This theoretical framework allows to get away from the commonly referred to *lubrication* argumentation and offers the advantage of being simple enough in terms of computation to be tested both numerically and sensorially using oils of different viscosities. We show that this interpretation allows to account for around two orders of magnitudes of apparent viscosity increase in such emulsions, when the coalescence is catastrophic (total). Such phenomena are predicted to have profound effects on the damping of the mechanical signal perceived by mechanoreceptors in the oral cavity and thus on *mouthfeel* perception.

Introduction

It has been frequently reported that the *mouthfeel* of liquids and soft solids can be explained by a combination of shear and a squeeze flows[1] and the mechanical stresses produced in those flows which are sensed by human mechanoreceptors[2]. Key sensory attributes considered are *thickness* and *smoothness* (which can both be related to different degrees of friction between tongue and palate). Simply put, a creamy liquid food product is expected to have a high viscosity and a low friction coefficient, in addition to the appropriate aroma and taste chemical compounds compatible with the targeted product sensory expectations. While viscosity is measured with a rheometer, friction can be measured with a tribometer, with measurements and interpretation often relying on Amontons' and Coulomb's laws of friction[3]. Such a lubrication analysis has been approached over the past decade using commercial[4,5] and custom made[6–8] tribometers and also using a theoretical approximations from slider or ball bearings[9] or elastohydrodynamic (EHD) contacts[10,11].

One main physical attribute that has been reported to decrease the friction coefficient translating into a modified *mouthfeel* of food dispersions is their ability to coalescence in mouth[12]. In numerous articles, the idea that an oil film would form on the tongue and / or palate and that this film would *lubricate* the tongue / palate contact *i.e.* decrease friction coefficient has been proposed[12–14]. It appears that emulsions which are less stable in mouth, as those stabilized with melting fat crystals or amylase sensitive starch are *thicker, creamier or fattier* than those stabilized for structure robustness[6,15]. Although commercially available tribometers seem to be able to differentiate emulsions based on their fat content and correlate with sensory measurements[4], we discuss in this article whether this is the only explanation. Indeed tribology experiments seem to provide insight in differentiating emulsions within the very low range of volume fractions ($\phi = 0 - 10\%$)[4], but fail to further differentiate emulsions between $\phi = 10 - 50\%$, in spite of large sensory *thickness* differences and small *instrumental viscosity* differences[?]. In particular we show that coalescence of oil droplets in water emulsion may increase the shear stress (and thus the apparent sensory viscosity or *thickness*) perceived by mechanoreceptors on tongue and palate rather than decreasing it. A decrease would however be necessary to reduce apparent friction, provided that the normal stress is kept constant (the usual condition in tribological experiments).

In this article we propose an alternative theory to the one commonly proposed in the literature and try to formulate a consistent fluid mechanic explanation for mouthfeel modification of coalesced emulsions. We consider not a decrease in friction but to an increase in the apparent viscosity as it could be perceived by mechanoreceptors in the tongue and the palate.

Is oil coalescence really leading to a increase in lubrication in mouth ?

Some apparent contradictions may be the consequence of confusion between lubrication effects on different perception

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mechanisms: the total stress perceived by a set of mechanoreceptors (probably similar to that present in the glabrous skin[16]) is distinct from the individual variations of stress perceived by single receptors. The latter mechanism is thought to be strongly related to perceived *roughness* (the opposite of *smoothness*) of liquids and semi-solids[17]. A reduction of the total average stress (e.g. by a decrease of viscosity) may coincide with an increase of the size of individual fluctuations sensed by receptors (due to reduced damping of fluctuations). Standard tribometers measure the total stress on solid bodies (e.g. a pin, sphere or disk), *i.e.* an averaged property which provides no information on local variations. In addition, the locations and physiology of human mechanoreceptors[18] in the oral cavity do not permit them to decompose stresses or forces into different directions (normal vs. tangential). They therefore have no simple, obvious mechanism to evaluate a coefficient of friction as done in a tribometer. One should be careful when trying to obtain simple relationships between standard tribometry results and perceptions of *thickness* and *smoothness*, even when leaving aside higher-level interference of other sensorial (taste, aroma) and psychological (bias, prior expectation) factors.

Proposed theory relating oil drop coalescence with enhanced textural perception

We consider that an emulsion with dispersed phase volume fraction ϕ of viscosity η_e is sheared between tongue and palate, which are modelled as two infinite smooth parallel plates, separated by a distance h (of the order of magnitude of 1 mm, that is very large compared to the size of the oil drops, $r \leq 30\mu$m). Upon coalescence in mouth (e.g. shear, heat or enzymatically induced), an oil film with viscosity η_δ would then form by buoyancy along the surface of the palate. The thickness of this film is denoted δ and by definition of the volume fraction ϕ, $0 < \delta/h \leq \phi$. We consider that the emulsion is sheared due to the relative tangential motion with velocity V of the tongue compared to the palate. At the interface between the two domains (oil layer and emulsion), the velocity of the fluids is denoted V_i (see Figure 1).

If we consider the continuity of stress in this biphasic fluid, the wall shear stress τ that will be induced by the fluid flow will be constant throughout h and mathematically formulated as equation 1 which can be rearranged in terms of V_i in equation 2.

$$\tau = \frac{V_i \cdot \eta_\delta}{\delta} = \frac{(V - V_i) \cdot \eta_e}{h - \delta} \qquad (1)$$

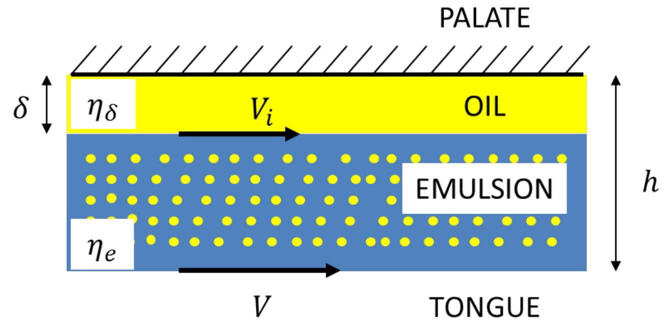


Fig. 1 Physical schematic representation of the phenomena occuring in mouth during texture evaluation after partial oil coalescence of O/W emulsion.

$$V_i = V \cdot \frac{\frac{\delta}{h}}{\frac{\delta}{h} + \frac{\eta_\delta}{\eta_e} - \frac{\delta}{h} \cdot \frac{\eta_\delta}{\eta_e}} \qquad (2)$$

To simplify this expression we define a dimensionless characteristic length $\Delta = \delta/h$ and dimensionless viscosity ratio $\Psi = \eta_\delta/\eta_e$ and thus equation 2 simplifies as equation 3.

$$V_i = V \cdot \frac{\Delta}{\Delta + \Psi - \Delta \cdot \Psi} \qquad (3)$$

We can now combine equations 1 and 3 to obtain the general expression for the shear stress perceived at the walls bounding the space which is given by equation 4.

$$\tau = \frac{V \cdot \frac{\Delta}{\Delta + \Psi - \Delta \cdot \Psi} \cdot \eta_\delta}{\delta} \qquad (4)$$

We can easily see that at the limit where the emulsion is not coalescing ($\delta \to 0$) then we obtain an indeterminate form ($V_i \to 0$) and we then use the second expression for τ in equation 1 which leads to $\lim_{\delta \to 0} \tau = V \cdot \eta_e/h$, the simple expression for the stress in such a Couette flow. We are now able to describe the relative increase in perceived stress under such conditions as a result of the coalescence of emulsions in the mouth. We derive in equation 5 that the dimensionless increase in stress Λ is independent of the velocity V and thus of the shear rate experienced by the fluid in the mouth. This is rather convenient since typical values of the shear rate experienced by the fluid in the mouth are difficult to establish.

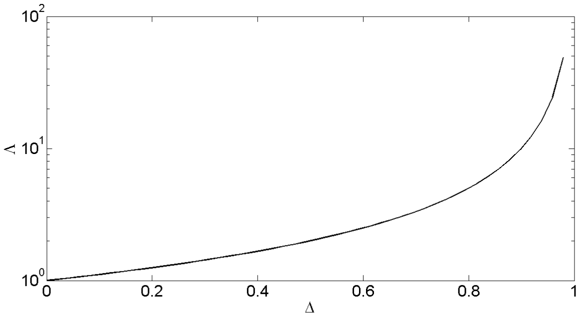


Fig. 2 Evolution of the relative stress increase Λ depending on the relative thickness of the coalesced oil layer Δ when we consider the usual numerical case where $\Delta \ll \Psi$.

$$\frac{\tau}{\lim_{\delta\to 0}\tau} = \frac{V\cdot\frac{\Delta}{\Delta+\Psi-\Delta\cdot\Psi}\cdot\eta_\delta}{\delta}\cdot\frac{h}{V\cdot\eta_e} \quad (5)$$

$$\Lambda = \frac{\Psi}{\Delta+\Psi-\Delta\cdot\Psi} \quad (6)$$

A simplification of this last equation is to consider that if $\Delta \ll \Psi$ (in a typical food oil in water emulsion, $0 < \Delta < 1$ and $\Psi \sim 10^2$) leads to the simplification 7, which allows to account for around two orders of magnitudes of apparent viscosity increase in such emulsions, when the coalescence is catastrophic.

$$\Lambda \approx \frac{1}{1-\Delta} \quad (7)$$

In view of this approximation shown in Figure 2, one may argue that this effect can only occur for sufficiently concentrated emulsions ($\Lambda = 2$ when $\Delta = 0.5$), since we assume that $0 < \Delta \leq \phi$. One should however consider that for any squeeze flow of the system shown in Figure 1, less of the oil film than of the emulsion will be expelled from the gap, thus increasing Δ. Any low-gap measurements as in tribology could lead to such a situation and thus that our proposed theory should also be qualitatively valid for dilute emulsions if they are concentrated by squeezing between tongue and palate preceding the shearing action used for texture evaluation[1]. As long as the flow remains quasi-static and the fluids Newtonian, the analyses of film thinning and shearing motion can be decoupled and our analysis remains valid. For more information on this kind of film thinning dynamics, we refer to Burbidge *et al.*[19].

This theory was derived for a total coalescence event leading to the formation of an oil film, but at least qualitatively, any coalescence leading to drops large enough to cream significantly in the time scale of the residence time of the emulsion in the mouth would lead to similar results. Indeed upon creaming, a high droplet volume fraction (and thus high viscosity) layer will be formed, thus leading to the same apparent viscosity increase for the mechanoreceptors due to a lower effective gap available for shearing.

Classical tribology can still bring understanding to texture perception

Although the theory that we propose is an alternative to the lubrication or tribology argument to explain effects of coalescence on texture perception, we still believe that tribology can bring insight to the understanding such effects.Indeed the effects we consider in this letter are still hydrodynamical in nature and would not of course describe in any way the left part of a Stribeck curve, which are dealing with *dry* contact mechanics, heavily dependent on surface properties. Its added value lies for example in the ability to design surfaces matching those of the human mouth (topology, elasticity, hydrophobicity). This is not addressed by standard rheological characterization which simply aims to describe the properties of the fluid present in the gap[3]. In this sense tribology may help to bridge the gap between classical rheology and sensory experiences by introducing surface dependence and making interfacial phenomena explicit in the lubrication theory[13].

Conclusions

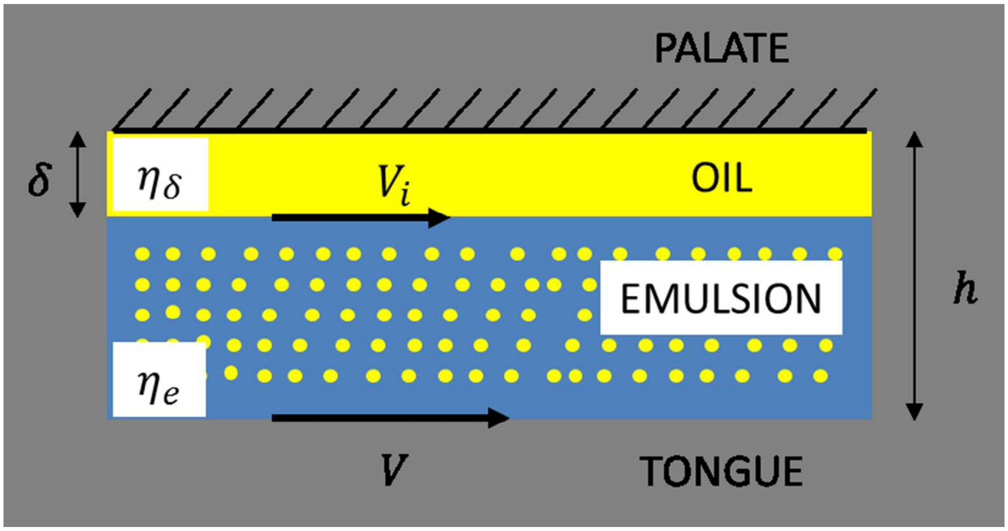
When effects of oil droplet coalescence on in-mouth texture perception of food emulsions are discussed, some careful distinctions must be made. Firstly, effects on viscosity (which usually lead to changes for the sensory attribute *thickness*) and lubrication (which usually lead to changes in the sensory attribute *smoothness*) both contribute to the perceived *body*, *richness* or *creaminess* of such products. However, effects of an oil layer on viscosity and lubrication are not necessarily the same. When speaking about *friction* and *lubrication* in the tongue-palate system, one should be clear if stress fluctuations (as for example originating from suspended solids) or a reduction of the overall shear stress is considered: a higher viscosity may be more effective in reducing stress fluctuations, but also increase the total shear stress (which would lead to higher friction as measured with classical tribometry methods). Secondly, when considering the effect of a layer of coalesced oil droplets on the overall viscosity of an emulsion, we need to distinguish between cases where the viscosity of the bulk emulsion is higher or lower than that of the oil layer itself.

The simple analysis presented in this article shows that for a simple shear flow in a gap of constant height, the effective viscosity enhancement produced by a layer of oil, relative to the viscosity of the emulsion, can be predicted by a simple relation containing only the thickness of the oil layer relative to the gap height and the viscosity ratio as parameters.

Since our objective in this short communication was primarily to point out some non-intuitive effects of oil droplet coalescence on in-mouth perception, we have not considered more complex contributions, e.g. from non-constant gap thickness, shear rate dependence or transient effects. Future theoretical and experimental work on flow behavior of stable and unstable emulsions in model gap systems, as well as simple well though sensory experiments should further elucidate such effects.

References

1 J. L. Kokini and E. L. Cussler, *Journal of Food Science*, 1983, **48**, 1221–1225.
2 A. S. Burbidge, in *Food Oral Processing*, ed. J. Chen and L. Engelen, Wiley, 2012, ch. Design of Food Structure for Enhanced Oral Experience, pp. 357–379.
3 J. Stokes, M. Boehm and S. Baier, *Current Opinion in Colloid and Interface Science*, 2013, **18**, 349–359.
4 M. Malone, I. Appelqvist and I. Norton, *Food Hydrocolloids*, 2003, **17**, 763–773.
5 J. Stokes, L. MacAkova, A. Chojnicka-Paszun, C. De Kruif and H. De Jongh, *Langmuir*, 2011, **27**, 3474–3484.
6 D. Dresselhuis, E. de Hoog, M. Cohen Stuart and G. van Aken, *Food Hydrocolloids*, 2008, **22**, 323–335.
7 D. Dresselhuis, H. Klok, M. Stuart, R. De Vries, G. Van Aken and E. De Hoog, *Food Biophysics*, 2007, **2**, 158–171.
8 S. Goh, P. Versluis, I. Appelqvist and L. Bialek, *Food Research International*, 2010, **43**, 183–186.
9 J. Engmann and A. S. Burbidge, *Food and Function*, 2013, **4**, 443–447.
10 J. Skotheim and L. Mahadevan, *Physics of Fluids*, 2005, **17**, 1–23.
11 G. Van Aken, *Soft Matter*, 2010, **6**, 826–834.
12 D. Dresselhuis, E. de Hoog, M. Cohen Stuart, M. Vingerhoeds and G. van Aken, *Food Hydrocolloids*, 2008, **22**, 1170–1183.
13 E. De Hoog, J. Prinz, L. Huntjens, D. Dresselhuis and G. Van Aken, *Journal of Food Science*, 2006, **71**, E337–E341.
14 D. Dresselhuis, G. Van Aken, E. De Hoog and M. Cohen Stuart, *Soft Matter*, 2008, **4**, 1079–1085.
15 J. Benjamins, M. Vingerhoeds, F. Zoet, E. de Hoog and G. van Aken, *Food Hydrocolloids*, 2009, **23**, 102–115.
16 E. A. Lumpkin, K. L. Marshall and A. M. Nelson, *The Journal of Cell Biology*, 2010, **191**, 237–248.
17 L. Engelen, R. A. de Wijk, A. van der Bilt, J. F. Prinz, A. M. Janssen and F. Bosman, *Physiology & Behavior*, 2005, **86**, 111 – 117.
18
19 A. S. Burbidge and C. Servais, *Journal of Non-Newtonian Fluid Mechanics*, 2004, **124**, 115 – 127.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