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Sensory perception and lubrication properties of milk: Influence of fat content

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ABSTRACT

The sensory perception of homogenized milk with a fat content between 0.06 and 8% was correlated with its friction coefficient and viscosity. Above a threshold of 1% fat, there was a strong decrease in friction coefficient at low speeds, which is associated with shear-induced coalescence. Creamy perception was perceived only for products with the friction coefficient below 0.25 for silicone rubber at entrainment speeds lower than 200 mm s^{-1}. Under those conditions, a linear correlation between perceived creaminess and friction was obtained at a fat content above 1%. The increased creaminess and thus decreased friction was attributed to the coalescence of fat globules on the surface of the tongue and rubber disc, respectively. At higher speeds, fused fat droplets were broken into smaller droplets (reversing coalescence) due to the high shear, thereby eliminating the correlation.

1. Introduction

An important sensory attribute of dairy products is the perceived creaminess or fat film formation, which is associated with the amount of fat present (Van Aken, Vingerhoeds, & De Wijk, 2011). Creamy or fatty perception was previously related to the apparent viscosity of the product (Akhtar, Stenzel, Murray, & Dickinson, 2005; Kokini, 1987; Moore, Langley, Wilde, Fillery-Travis, & Mela, 1998). Fat content and viscosity are directly related in dispersion rheology. However, the sensory properties of emulsions cannot be fully described by bulk rheology. Therefore, the ability to relate another physical property, e.g., the friction coefficient, of a product to its sensory perception (e.g., fat) is of key importance for the food industry, and it was highlighted by De Wijk and Prinz (2005); Giasson, Israelachvili, and Yoshizawa (1997); Lee, Heuberger, Rousset, and Spencer (2004); Malone, Appelqvist, and Norton (2003); and Ranc, Servais, Chauvy, Debaud, and Mischler (2006). Previous studies showed that creaminess is a complex attribute composed of different factors such as viscosity, taste, aroma, smoothness and thickness sensation (Cutler, Morris, & Taylor, 1983; Kokini & Cussler, 1983; Richardson & Booth, 1993). The aim of studying the physical parameters (e.g., viscosity or friction) is to simplify the process of product development and to provide a measure for programming sensory perception based on modification of tribological properties of a food product. During consumption, food is rubbed between the tongue and the palate, forming a thin film on the oral mucosa that experiences friction forces. The relationship between thin film properties and food texture was investigated for chocolate (Luengo, Tsuchiya, Heuberger, & Israelachvili, 1997) and mayonnaise (Giasson et al., 1997). These studies confirmed that tribology, thin film morphology and wetting properties of the samples provide relevant insight that could not be obtained from bulk rheology alone. It was concluded that sensory texture of the samples correlated better with thin film tribological properties than with bulk characteristics (Giasson et al., 1997; Luengo et al., 1997).

In a tribological measurement, the friction coefficient is measured as a function of speed. Three different lubrication regimes (boundary, mixed and hydrodynamic) can be distinguished (Bongaerts, Fourtouni, & Stokes, 2007; Cassin, Heinrich, & Spikes, 2001; De Vicente, Stokes, & Spikes, 2005). As a result, a so-called Stribeck curve is obtained. The boundary regime shows high and constant friction at low speeds, where surfaces are in contact and the friction coefficient is affected by the properties of the rotating surfaces and a thin boundary film. As the entrainment speed increases, the surfaces become partially separated. Therefore, the friction coefficient decreases and depends partly on both the bulk rheological behaviour of the lubricant and the properties of the surfaces. At high speeds, in the hydrodynamic regime, surfaces are separated and lubrication is mostly governed by bulk rheological properties of the lubricant.

There is a large volume of research on the tribology of emulsions in relation to rolling and cutting of metals, in which emulsions are used. Usually these measurements are done in and relevant to the

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boundary regime. Kumar, Daniel, and Biswas (2010) recently argued that it is entrapment of small and marginally stable oil droplets that provides the lubrication. However, in practice, rather crude emulsions are used for cutting metal. Usually lubrication is attributed to an adsorbed layer of oil in the contact area.

The lubrication properties of the oil in water emulsions in a steel-soft elastomer contact were studied by De Vicente, Spikes, and Stokes (2006), who showed that oil droplets enter the contact zone and dominate the lubrication properties when the viscosity of the dispersed oil phase is at least four times larger than the viscosity of the continuous phase. Otherwise, the dispersion medium dominates the friction properties.

Recently, an extensive study was reported relating the creamy and fatty perception of emulsions to their lubrication properties (Dresselhuis et al., 2007; Dresselhuis, Cohen Stuart, Van Aken, Schipper, & De Hoog, 2008a; Dresselhuis, De Hoog, Cohen Stuart, Vingerhoeds, & Van Aken, 2008b). Dresselhuis et al. (2008b) demonstrated that emulsions with higher efficiency towards coalescence showed lower friction and were perceived as more creamy/fatty. The friction force was measured experimentally in a custom-made tribometer, for emulsions with 10–40% fat content (De Hoog, Prinz, Huntjens, Dresselhuis, & Van Aken, 2006). That work showed that in the range studied, of high fat content, no correlation between measured friction force and oil remaining at the mucosal surfaces existed.

A number of studies investigated the relationship between sensory attributes and composition or physical characteristics of the product. De Wijk, Prinz, and Janssen (2006a) suggested a three dimensional sensory space for dairy custard desserts. The principal axis, ranging roughness to creaminess, correlated with fat content; the melting to thick axis correlated with the starch content and the viscosity of the food. A further study showed that the creamy attribute could be associated with bulk and surface properties of the oral food bolus (De Wijk, Terpstra, Janssen & Prinz, 2006b). All these studies were made for custard with fat contents varying between 0 and 15%.

It is a common experience that skim milk is perceived as watery and not creamy. However, leaving a small percentage of fat in the milk improves taste considerably. The influence of fat on the sensory properties, viscosity and colour of low fat milk (0–2% fat) was studied by Phillips, McGiff, Barbano, and Lawless (1995), who showed that the mouth-feel attributes increased with fat content. However, the unambiguous interpretation of the results was argued over because of a difference in visual appearance. Similar results were obtained by Pangborn, Bos, and Stern (1985) who showed that once the panellists were not able to observe the milk visually, they were not capable to discriminate the differences between 0 and 3.5% fat in milk.

In this work, a tribometer and a rheometer were used to characterise homogenised and pasteurised milk with varying fat content. Sensory evaluations of the samples were made using quantitative descriptive analysis (QDA), and data were correlated with the physical parameters. Confocal scanning laser microscopy (CSLM) was employed to detect fat coalescence and its influence on friction measurements. As far as we know, this is a first study at (very) low fat content that relates sensory attributes to the measured friction coefficient between two soft surfaces. The importance of coalescence is considered as well.

2. Materials and methods

2.1. Sample preparation

Homogenised and pasteurised skimmed milk with fat content of 0.06% (w/w) and full fat milk with 8.68% (w/w) of fat was standardised to the same protein content (3.3%, w/w) and supplied by FrieslandCampina Innovation (Wageningen, The Netherlands). The full fat milk was obtained by mixing cream (40% fat) with skim milk (0.06% fat). The milk was heated to 65 °C, and homogenised at 95 bar (first stage) and at 10 bar (second stage). Finally, it was heated to 73 °C with 15 s holding time, and cooled to 6 °C. Skimmed and full fat milk were mixed in order to obtain samples containing 0.15, 0.3, 0.5, 0.7, 1.0, 2.0, 3.0, 4.0, and 6.5% (w/w) fat. The samples were stored at 5 °C, and used within two days. Freshly prepared samples were used in each experiment. The volume–surface average or Sauter diameter ($d_{3,2}$) of the fat globules in homogenised milk was determined using a Mastersizer 2000 (Malvern Instruments Ltd., Malvern, UK) and found to be 0.27 μm.

2.2. Rheological and tribological measurements

The viscosity of the samples was determined at 20 °C using an AR2000 rheometer (TA Instruments, Leatherhead, UK) with double concentric cylinder geometry. Flow curves were obtained by measuring the apparent viscosity as a function of increasing shear rate. The experiments consisted of two steps: a conditioning step where the system was temperature equilibrated, followed by actual measurement with increasing shear rate from 0.1 to 1000 s^{-1}. Each point was measured at a fixed shear rate with a duration time of 12–18 s.

A mini traction machine (MTM; PCS Instrument Ltd., London, UK) with compliant rotating surfaces was used to measure friction coefficient as a function of the entrainment speed. Formerly, the setup consisted of a steel ball and disc, which led to high contact pressure. In this work the equipment design was modified to a steel cylinder with an attached neoprene O-ring. In addition, a 3 mm thick disc (made of silicone, neoprene or Teflon; Eriks Company, Arnhem, The Netherlands) was fixed on top of the steel disc. These adjustments led to a decrease of the contact pressure (from GPa to kPa) between the two rotating surfaces. A more detailed description of this method and the properties of the rubbers were reported by Chojnicka, Visschers, and de Kruif (2008). The measurements consisted of five steps, with the first one as conditioning step where the system was temperature equilibrated. The other steps were performed with load (W) of 5 N, slide-to-roll ratio of 50% and speed varying from 500 mm s^{-1} to 5 mm s^{-1}. Each measurement was taken at 20 °C with a new surface cleaned with ethanol and reverse osmosis water and then dried with air. All measurements were performed at least three times and averaged (each presented point corresponds to a mean value). The standard deviation in all the measurements was below $\sigma = \pm 0.02$. The temperature of 20 °C was chosen for rheological and tribological measurements to be in line with the temperature at which the panellist evaluates the samples.

2.3. Confocal laser scanning microscopy

Special samples were prepared in order to observe a possible coalescence of the fat on the surface of the discs in the tribometer. A mixture of fluorescein 5-isothiocyanate and Nile Red (FITC/NR) was used to stain the protein and oil phases of the milk, respectively. A 55 mL aliquot of sample was mixed with 1 mL of FITC/NR prior to the tribological experiment. After the MTM measurements, the visualisation of the surfaces used was done with a LEICA TCS SP by confocal laser scanning microscope (CLSM) equipped with an inverted microscope (model Leica DM IRBE) and Ar, DPSS and HeNe or Ar/Kr visible light lasers (Leica Microsystems GmbH., Mannheim, Germany). The objective lenses of 5 × magnification and 20 × magnification (HC PL APO 20×/0.70 CS) were used. Excitation was performed at 488 nm and 520 nm for FITC and Nile Red, respectively. Reported digital images for silicone and Teflon disc were recorded at a penetration depth of 10 μm. Due to large asperities in the neoprene rubber a projection (scan) of 30 μm inside the track was performed. Images were obtained in 1024×1024 pixel resolution.

2.4. Quantitative descriptive analysis (QDA)

An expert panel (consisting of 10 persons) evaluated the sensory properties of the milk with different fat content. Due to the limited number of samples (nine) that could be tested by assessors in the QDA sensory panel, only milk with up to 4% fat content was used. The panel was familiarised and trained to score on seventeen attributes common in the assessment of milk (see Table 1). These attributes belong to four different categories, i.e., smell/taste, mouth-feel, mouth/after-feel and after-taste/feel sensation. The samples were presented to each assessor at 14 ± 2 °C, in two different groups due to the large differences in the fat content. The five lowest fat contents (from 0.06 to 0.7%, w/w) were presented in the first group, whereas the second group consisted of the samples with the highest fat percentages (from 1 to 4%, w/w). The reason for the grouping was to prevent cross-over effects (of especially fat) as much as possible. Each sample was served in a small plastic cup that was covered with aluminium foil to avoid the influence of light and to ensure that the head space, for the smell evaluation, was optimal. Moreover, the panellists were not able to see the samples prior to testing. Within each group the samples were presented unmarked to assessors and in a randomised sequence. Assessors assigned a score on a line scale from 0 to 100 for each attribute. Since samples were typically kept in the mouth for few seconds during the sensory evaluation, we expect that the apparent temperature of the tasted product warmed up to about 20 ± 2 °C.

2.5. Statistical analysis

Analysis of variance (ANOVA) with least square difference (LSD) tests was done using STATISTICA data analysis software (version 7, StatSoft Inc., Tulsa, OK, USA). The level of significance (α) was set on 0.05. Statistical significance of differences between mean values was analysed by using the multiple comparison Tukey test. In a separate analysis, sensory attributes were averaged across judges and replicates, and analysed using principal components analysis (PCA) (Piggott & Sharman, 1986) using FIZZ software (Biosystemes, Dijon, France).

Table 1
Definitions of attributes generated by quantitative descriptive sensory analysis.

Attribute	Definition
Smell/taste (S/T)	
Creamy	Smell and taste associated with (whipped) cream
Flat/Watery	Lack of milk smell and taste
Sour	Taste of lactic acid
Sweet	Taste of milk sugar
Musty (muff)	Smell and taste associated with a cellar/old cupboard/cabinet
Metallic	Smell and taste associated with (blood) metal
Cardboard	Smell and taste associated with wet paper/cardboard
Mouth-feel (MF)	
Creamy	Mouth-feel associated with (whipped) cream
Powdery	The sensation of small particles like flour
Soft/velvet	A soft sensation in the mouth
Watery	The sensation of water in the mouth (no body)
Sticky	A sticky sensation on palate and between the teeth
Mouth/after-feel (AF)	
Rough	A rough/coarse sensation in the mouth/on the tongue (after swallowing)
Fat film	The sensation of a fat-like layer in the mouth (after swallowing)
After-taste/feel (AT)	
Slimy	A slimy sensation in the throat after swallowing
Dry	A dry sensation on the tongue/mouth after swallowing

3. Results

3.1. Friction coefficient

We used three different surfaces, with the aim to find which data correlated best with sensory results. The three surfaces were silicone, neoprene and Teflon, and differed in hardness and surface roughness. The tribological data are presented in the form of the Stribeck curve where friction coefficient is shown as a function of speed (maximum 500 mm s^{-1}).

3.1.1. Silicone

Fig. 1a shows the friction coefficient for all samples. For the lowest fat content, we observe boundary lubrication below a speed of about 10 mm s^{-1}. The mixed regime follows at higher speeds. Interestingly, all emulsions with a fat content below 1% exhibited similar behaviour. If the fat content was further increased, there was an abrupt decrease in friction, which became constant at a fat content of about 6.5%. As the friction coefficient decreased with increasing fat content, the boundary regime extended as well to higher speeds. This behaviour is most likely related to coalescence of fat droplets and subsequent adhesion of fat onto the surfaces. The higher the fat content, the thicker or the more extended the boundary film becomes. Once sufficient film thickness is established, the bulk fluid entrainment is suppressed. Initially, for emulsions with a low fat content, the friction may be governed by the bulk fluid, and possibly lubrication is dominated by proteins at all entrainment speeds. On the other hand, for high fat content, the fat forms the boundary film and dominates the lubrication, allowing the entrainment of the bulk emulsion to be suppressed and causing an extended boundary regime with very low friction coefficient. High speed, however, most likely leads to breaking the adhered fat layer and emulsifying the fat. The friction increases then as a result of entrainment of bulk fluid and influence of proteins that provide poorer lubrication than fat.

3.1.2. Neoprene

The Stribeck curves for neoprene rubber (Fig. 1b) were very similar to those of silicone (Fig. 1a), with the exception of milk with 0.06% fat. The latter sample had a friction coefficient about 0.1 units higher than other low fat samples, and was comparable to that of water. This suggests that for neoprene there are two thresholds. The first one requires a minimum fat content to show any change in lubrication characteristics. The second threshold is for fat content of ~2%. Below this concentration, Stribeck curves are practically undistinguishable. Above that value, an increasing fat content causes a decrease of the friction coefficient.

The friction coefficient was for all samples higher in the case of neoprene rubber than for silicone (with asperities of around 10 μm). This is probably a consequence of the larger surface asperities of the neoprene rubber (with asperities of around 30 μm), as was discussed previously by Chojnicka et al. (2008).

3.1.3. Teflon

The Stribeck curves of Teflon (Fig. 1c) are very different from the other two materials. The Teflon surface was very smooth and much harder (with Young's modulus of around 30 MPa) than silicone (Young's modulus of 10 MPa) or neoprene (Young's modulus of

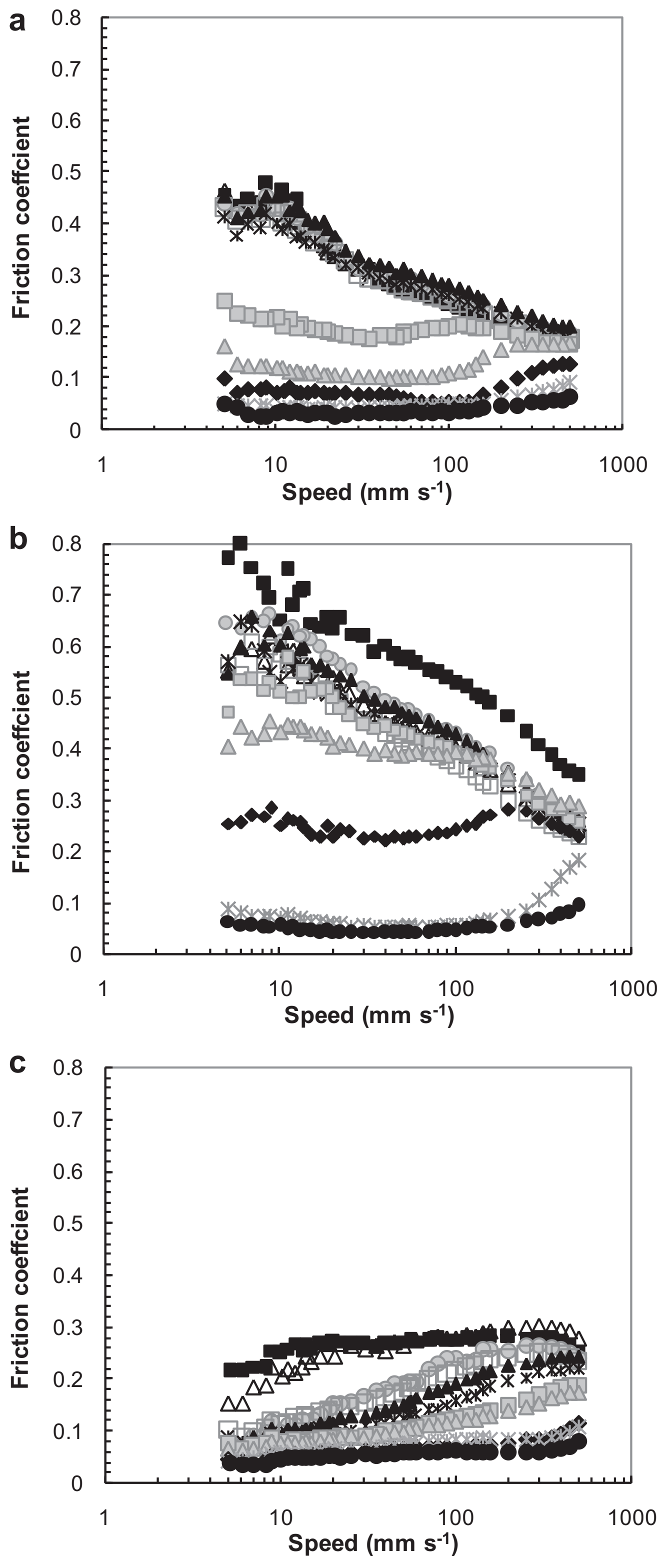

Fig. 1. Friction coefficient measured at load $W = 5$ N with (a) silicone, (b) neoprene or (c) Teflon for milk samples with different fat content. The symbols correspond to: ■ 0.06; △ 0.15; ● 0.3; □ 0.5; ▲ 0.7; ✱ 1; ■ 2; ▲ 3; ◆ 4; ✱ 6.5; ● 8.7%.

11 MPa) (unpublished results). This resulted in significantly lower friction in the low speed range. The boundary regime extended to very high speeds, and the onset of the mixed regime was observed only for speeds ≥300 mm s^{-1}. Moreover, all milk samples showed unique characteristics and could easily be distinguished based on their frictional properties. An increasing fat content caused a decrease of friction even for very low concentrations of fat, unlike for the two other rubbers, where a certain threshold was needed in order to further decrease the friction coefficient.

3.2. Viscosity

Fig. 2 shows the viscosity as a function of shear rate. Two orders of magnitude change in fat content translated to only a factor of two in viscosity. Moreover, the flow curves practically overlap for the milk samples with fat content below 1% (a marginal increase of the viscosity was observed between 0.06 and 1% fat). The fact that the dispersion is higher in viscosity and also shear thinning indicates that there are attractive interactions between the fat globules, probably caused by depletion forces induced by the casein micelles (Ten Grotenhuis, Tuinier, & de Kruif, 2003). Whatever the precise origin, the changes in viscosity are only fractional, whereas changes in friction coefficient are an order of magnitude. Therefore, the friction coefficient appears not to be directly determined by dispersion viscosity.

3.3. Confocal laser scanning microscopy (CLSM)

CLSM images, presented in Fig. 3, revealed that fat droplets coalesced at the surface, forming larger structures surrounded by bulk solution. In the case of silicone, the surface was well covered by an adhered fat layer. The track formed during the tribological experiment is easily visible, as fat seems to be concentrated more towards the centre of this track (light grey), while protein is visible at the edges (dark grey).

For neoprene, the fat seemed to form a structure as a result of adhesion and coalescence of fat droplets within large asperities. The surface, however, was not covered with a uniform layer. The distinction between protein and fat components was more visible than for silicone.

Teflon shows a different picture, with structures formed by the coalescing fat. The lipophobic surface prevents adhesion, and thus large droplets have a more confined shape. This results in sharper boundaries between the components.

The coalescence of fat is clearly visible for all discs, and thus verifies the lubrication hypothesis.

3.4. Sensory analysis

The results of the sensory analysis of milk samples containing 0.06–4% fat are shown in Fig. 4 for a creamy attribute. There was no systematic trend for the low fat samples (below 1%). The creamy score is low and ‘noisy’. Differences in sensory attributes were perceived only above 1%, suggesting that there was a critical fat content of ∼1%, below which the fat has no impact on the perceived creaminess.

Several sensory attributes showed a good correlation for fat content above 1%, while others showed no relation at all (data not shown). It should be noted that two groups of attributes can be distinguished, i.e., describing taste and describing texture perception. In the case of taste, the creamy attribute increased with increasing fat content. Similarly, creamy texture showed the same trend, although at a lower score. A similar score as for creamy texture is obtained for the soft/velvet attribute, which increased with increasing fat content. The watery attribute is found in both taste and texture categories and it scored almost exactly the same in both categories.

Interestingly, there was no correlation between (low) stickiness and fat content. This is in agreement with previous findings, where

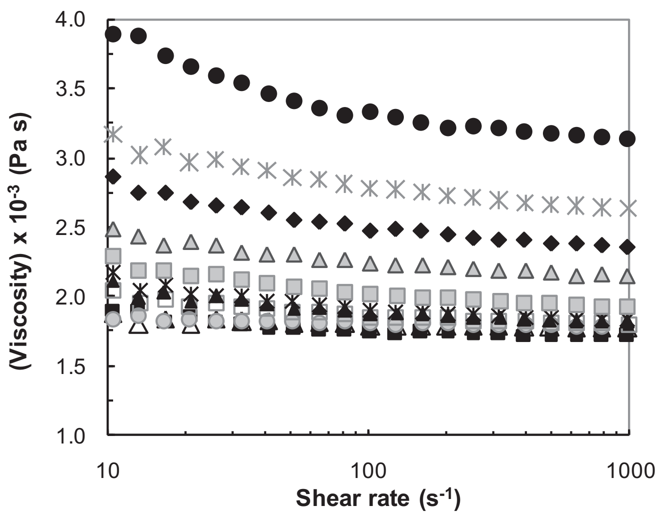

Fig. 2. Viscosity as a function of the shear rate for milk samples containing different fat content. The symbols correspond to: ■ 0.06; △ 0.15; ◯ 0.3; □ 0.5; ▲ 0.7; ✱ 1; ▩ 2; △ 3; ◆ 4; ✱ 6.5; ● 8.7%.

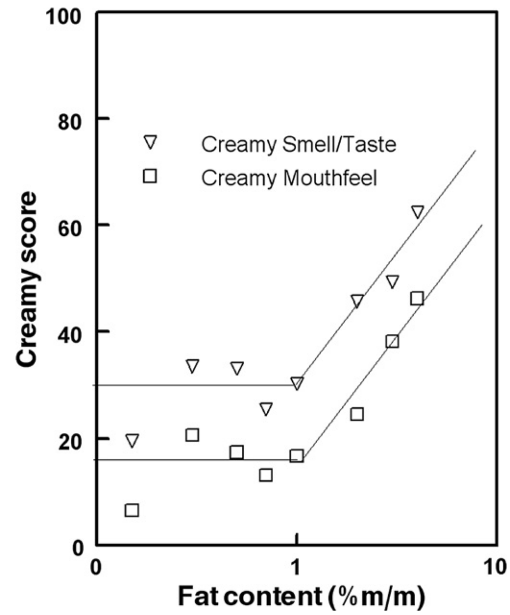

Fig. 4. Creamy smell/taste (▽) and creamy mouth-feel (□) attributes as a function of fat content of the milk.

low perception of stickiness was associated with high creamy perception for custard products (Kootstra, Holthuysen, & Mojet, 2007). On the other hand, the fat film attribute shows an increasing trend with an increasing concentration of fat.

4. Discussion

4.1. Coalescence of the emulsion droplets on the discs

As shown in Fig. 3, after the tribological measurements, the coalescence of the emulsion droplets occurred on the rubber surfaces, e.g., in the case of silicone rubber, the fat droplets size reached up to even 50 μm. This coalescence most probably originated from the adhesion and subsequent spreading of the oil on the surface in the contact zone (De Hoog et al., 2006; Dresselhuis et al., 2007, Dresselhuis et al., 2008b; Van Aken, Vingerhoeds, & De Hoog, 2007). This is called surface-induced coalescence, and this process might be related to the affinity of the fat globules to the surface, which leads to a cascade. Once the process has been initiated, more and more fat is deposited on the disc in the contact zone.

In the tribometer setup, where strong shear is applied, the emulsions in the contact zone are subjected to this shear in a very small gap. This might facilitate the surface-induced coalescence, and thus can be called shear-induced-surface coalescence. Moreover, deposition of the fat globules might be facilitated by shear-induced coalescence in the bulk. However, the latter process is less probable, as homogenised milk is known as a very stable emulsion. Surface-induced coalescence for a number of emulsions was observed, while no shear-induced coalescence in the bulk was observed (Dresselhuis et al., 2007). Similarly, deposition and coalescence of the oil in the contact zone was observed (Dresselhuis et al., 2008b) between two shearing surfaces, whereas no increase of the droplet size in the bulk of the emulsion was noted. In the current study, the CLSM images of the oil for the neoprene disc (Fig. 3b) seemed to form a structure, which is likely caused by the deposition of the emulsion droplets between large asperities inside the contact zone. It can be noticed that fat is deposited only in the contact zone, where shear occurred. The adhesion to the surface is very likely supported by large surface roughness (especially large cracks formed in the disc) that can physically entrap the emulsion. Similar observations were made by De Hoog et al. (2006), where oil patches were visible only in the contact zone, after shearing of oil in water emulsions between a rubber ball and oscillating glass. Dresselhuis et al. (2008a) showed that surface-induced coalescence of the emulsions leads to a decrease in oral friction measured in-vivo (human tongue) and ex-vivo (pig's tongue). This is in line with our study, even though different tribological methods and conditions were applied (e.g. rotation instead of oscillation, different surfaces, speed, and load). Therefore, in the oral

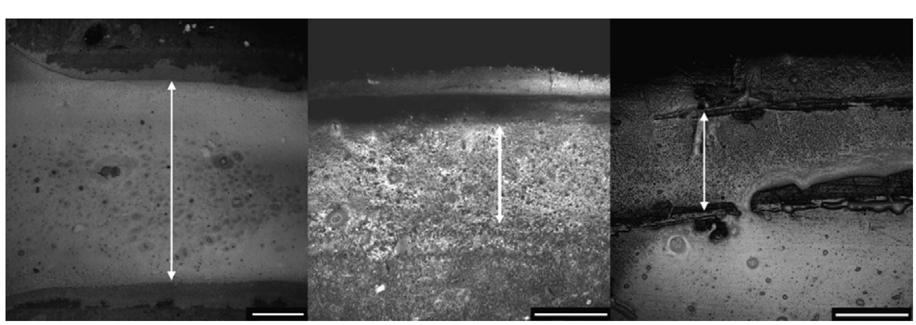

Fig. 3. Confocal laser scanning microscopy images of three different rubbers (left image, silicone; middle image, neoprene; right image, Teflon) after processing 3.5% milk in a Mini Traction Machine. Scale bar corresponds to 250 μm for silicone and 750 μm for neoprene and Teflon.

Table 2
The correlation coefficient between sensory attributes and the friction coefficient and viscosity (for 0.06–4% fat content).[a,b]

Attribute[c]	Viscosity (Pa s)	Friction coefficient		
		Silicone	Neoprene	Teflon[d]
Creamy (S/T)	0.93	−0.95	−0.93	−0.74
Creamy (MF)	0.95	−0.94	−0.92	−0.67
Soft/velvet (MF)	0.95	−0.96	−0.93	−0.71
Watery (MF)	−0.89	0.94	0.87	**0.64**
Fat film (AF)	0.89	−0.88	−0.90	−0.78
Slimy (AT)	0.81	−0.77	−0.80	−0.84

[a] The viscosity and friction coefficient of emulsions with different fat content were taken at high shear rate and entrainment speed of 10 mm s^{-1} (with $W = 5$ N), respectively.
[b] The significance level of the correlation coefficient (r) for studied population size ($n = 11$) and $\alpha = 0.05$ is 0.666.
[c] S/T, smell/taste; MF, mouth-feel; AF, mouth/after-feel; AT, after-taste/feel.
[d] In bold the watery attribute for Teflon disc is shown as the only one below the significance level.

environment, the adhesion and coalescence of oil droplets between the papillae will lower the friction.

In the case of Teflon (Fig. 3c), fat seems to adopt large structures outside the track. Most likely the droplets were ejected from the contact zone where they underwent coalescence. As the surface is lipophobic, fat cannot adhere to it.

4.2. Correlation between data obtained by different methods

Testing oral perception is commonly performed by trained assessors who assign relative scores to certain attributes. Other methods like tribology provide more absolute values of parameters that are not easily transformable to attributes of oral perception. Here, we aim to find correlations between oral perception data on *creamy*, *watery*, *soft*, *fat film*, and *slimy* obtained from the QDA panel and the experimental physical data.

4.2.1. Correlation between viscosity data and QDA data

Although the range of viscosity of the emulsions studied in this work is narrow, the viscosity increases systematically with an increasing fat content. The initial increase, however, is very small (Fig. 2). Below 1% fat, no correlation was observed between sensory attributes and viscosity. The correlation coefficient for milk samples with varying fat content between 0.06 and 4% for presented attributes is well above significance level (Table 2). Although there is quite some correlation between the data, we believe that this due to the presence of fat rather than due to viscosity. Increasing viscosity by adding more casein micelles or adding a small polymer does not change sensory attributes in a similar way (Stokes, Macakova, Chojnicka-Paszun, de Kruif, & de Jongh, 2011).

4.2.2. Correlation between friction data and sensory data

Friction data were obtained for a wide range of entrainment speeds. Therefore a specific speed needed to be chosen in order to determine the correlation coefficient. In this work, four different speeds were selected, from both the boundary and the mixed regime, to evaluate correlations: 10, 50, 150, and 400 mm s^{-1} (data not shown). The highest correlations were expected at the lowest speeds, as they correspond to typical velocities in the oral environment (Malone et al., 2003; Van Aken et al., 2011). Moreover, the friction depends on the type of the surface (rubber) used in the tribometer setup. The correlation analysis allows the determination of the most suitable oral surface analogue among the three investigated materials.

Fig. 5 shows the friction coefficient as a function of fat content measured at different experimental conditions for silicone and neoprene discs. Friction was nearly independent of fat content up to a critical value of about 1% fat. Interestingly, this value was the same for silicone rubber and neoprene rubber. A decrease in friction coefficient with an increasing fat content means that fat supports boundary film formation and provides better lubrication conditions. The data for neoprene rubber are shifted to higher values with respect to the silicone data, which results from significantly larger surface roughness.

Teflon (data not shown) showed almost no dependence of the friction coefficient on the fat content at low speeds. This is most likely due to the lipophobic nature of this surface. The boundary fat film cannot be formed on this surface and thus the friction coefficient remains constant regardless of the fat content in the lubricant. We therefore did not present further data on Teflon. We concluded that Teflon is not suitable for the purpose of this investigation as is not a good candidate for an oral surface analogue. At higher speeds (see Fig. 1), silicone and neoprene rubbers show a much weaker dependence of the friction on the fat content, as they are in the

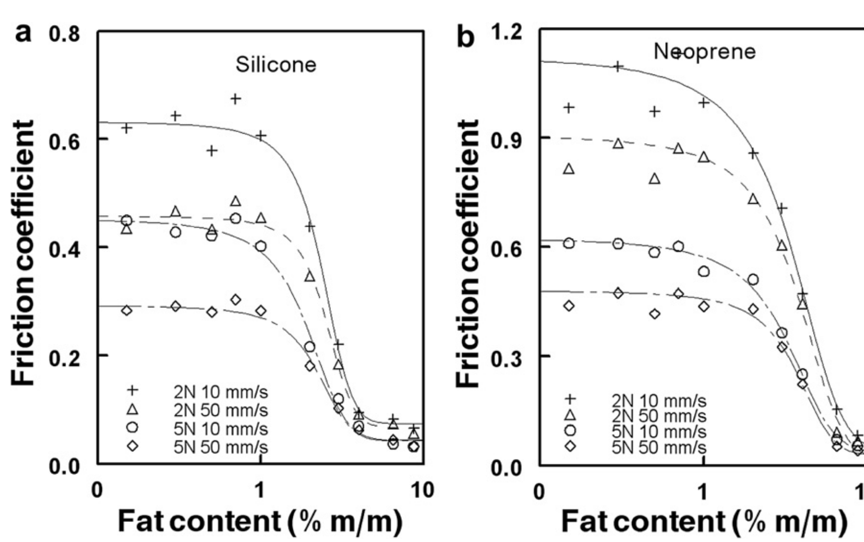

Fig. 5. Friction coefficient as a function of fat content for (a) silicone and (b) neoprene. Symbols correspond to different experimental conditions: +, load (W) of 2 N and speed (U) of 10 mm s^{-1}; Δ, $W = 2$ N, $U = 50$ mm s^{-1}; O, $W = 5$ N, $U = 10$ mm s^{-1}; ◇, $W = 5$ N, $U = 50$ mm s^{-1}. Lines are fits to a sigmoidal function.

mixed regime; in those conditions the boundary film is of lower importance for the lubrication.

Fig. 6 presents creaminess as a function of friction coefficient at an entrainment speed of 10 mm s^{-1} and load of 5 N. The data show a linear relationship over a large dynamic range.

Thus it seems that the friction coefficient is a good parameter for creamy attributes determined for taste and texture perception. These are the representative attributes among all others tested by the panellist. Almost all attributes show negative correlation, except for the watery attribute.

We observed that a creamy sensation (and fat film formation) implicates efficient lubrication in the boundary regime. Previous studies (De Wijk & Prinz, 2005, 2006; Malone et al., 2003) showed that creaminess was associated with an increase of the fat content in the product, which resulted in decreased friction and thus better lubrication properties of the product. Similarly to creamy attributes and fat film, a soft or slimy feeling could not occur when the lubrication was inefficient and the friction coefficient was high. In such a case the perception is expected to be rough. The watery attribute, however, implies liquid lubricant that cannot adhere to the surface and thus cannot form a boundary film.

The correlation coefficients for relationships presented in Fig. 6 are summarised in Table 2. Data for silicone and neoprene were well above a significance level of 0.666 (for the studied population of $n = 11$ and $\alpha = 0.05$). This shows that a very good correlation was established between the sensory attributes and the friction coefficient. Besides, fat film and slimy attributes show a higher correlation coefficient for silicone rubber, thus suggesting that this rubber may represent the oral environment better. In addition, neoprene rubber undergoes a wearing process that changes the surface properties. Significant correlation coefficients were obtained for these rubbers in almost all cases, despite significant differences in material and surface properties of tested rubbers. This suggests that the friction data can be well related to the oral perception regardless of the two rubbers applied in the setup.

Interestingly, the choice of the speed at which the friction coefficient is selected for the correlation is of great importance. Oral processing occurs at speeds which range from about 10 to 50 mm s^{-1} (John Prinz, personal communication). In this work, a broader range of speed was applied in the tribometer (varying from 500 mm s^{-1} to 5 mm s^{-1}) in order to obtain a general knowledge of how the milk samples behaved at different lubrication regimes. Regardless of the disc used in the tribometer, correlation coefficients decreased with increasing entrainment speed.

4.3. Transition point

An interesting observation follows from evaluation of Fig. 1. For silicone and neoprene rubbers, friction curves at high speeds are similar for different fat content below certain threshold. In the case of silicone discs, samples containing up to 3% fat show very similar friction curves above 300 mm s^{-1}. In fact, Stribeck curves for 2% and 3% fat overlap above the entrainment speed of 350 mm s^{-1}. Higher concentrations, however, can be easily distinguished as their friction coefficients decrease with increasing fat content. For neoprene rubbers, this effect was observed above 200 mm s^{-1} and up to 4% fat. This indicates similar lubrication characteristics of emulsions with fat content below some threshold at high speeds (mixed regime in this case). This might result from the entrainment of the bulk emulsion into the contact zone between two surfaces that have been already partly separated. High speed and shear disturb the coalescent fat film and mix the fat droplets back into the emulsion. Therefore, the lubrication is governed by the bulk emulsion in all cases resulting in a similar friction coefficient.

Most likely the differences in threshold concentration of fat between silicone and neoprene rubbers are related to their differences in surface roughness. The rougher neoprene requires higher fat content to provide good lubrication, as larger asperities result in a more violent environment and thus easier distortion of the fat film. Consequently, for the neoprene rubber more fat is needed to discriminate the differences between low and high fat emulsions. For silicone rubber the threshold concentration is 4%, whereas for the rougher neoprene rubber this threshold is observed at 6.5% fat. This effect might be due to either i) the viscosity of the sample that is high enough to make a difference at these higher speeds, or ii) coalescence of the fat on the surfaces that is reversed at high speed and shear environment. Another explanation for the difference in threshold between silicone and neoprene might be their different hydrophobicity. As was shown by Chojnicka et al. (2008), the neoprene rubber showed a more hydrophilic character than the silicone rubber. Therefore, the latter rubber has a more favourable surface for adhesion of the fat droplets, which would result in lower friction and lower fat threshold than neoprene. Most probably both the effect of the surface roughness and the hydrophobicity of the

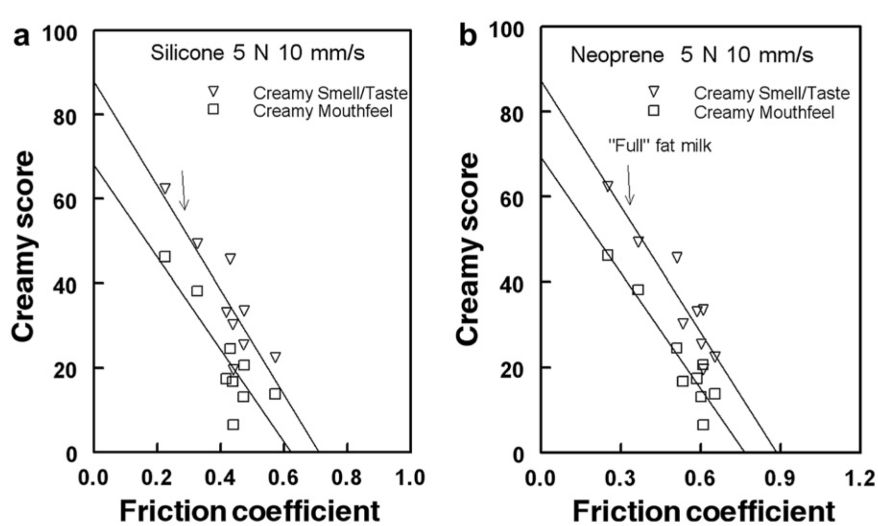

Fig. 6. Creamy score as a function of friction coefficient of the milk measured at 5 N and 10 mm s^{-1} for (a) silicone and (b) neoprene discs. Symbols correspond to a creamy smell/taste (▽) and creamy mouth-feel (□). The solid line is the fitted power-law function.

rubbers influenced the behaviour of the studied emulsions, although the latter parameter probably is less important, as the differences between hydrophobicity of rubbers are minor.

5. Conclusions

The sensory analysis of milk samples with different fat content showed that fat content below 1% cannot be distinguished, and thus has little influence on the texture and/or taste of food products. Above this threshold concentration, a significant correlation is found between a number of attributes (e.g., creamy) and the friction coefficient. The best correlations were obtained for low speeds that corresponded to the boundary lubrication regime and were comparable with the speed in the mouth. Although significant correlation was also established for the friction coefficient taken at higher speeds, material properties and surface characteristics seemed to have a stronger influence on the outcome of the analysis under those conditions.

Coalescence of fat droplets on the rubber discs occurred at low entrainment speed. This was attributed to the shear-surface-induced coalescence on the disc that had a significant effect on the lubrication properties of the emulsion enriched milk. This effect was observed above a fat content of 1% and 2% for silicone and neoprene, respectively, where a gradual decrease of the friction coefficients with fat content took place. At high speeds, however, the coalescence was reversed (fused fat droplets were broken apart), resulting in an increased friction.

Emulsions with fat content up to 3% for silicone and 4% for neoprene had similar lubrication properties at high speed. In spite of these high speeds, it would be interesting to investigate further whether these similarities can be translated to food developments.

A Teflon surface showed very different behaviour from other discs. This suggested that distinct characteristics of this material, in particular its lipophobic nature and very smooth surface, disqualify Teflon as a potential industrial analogue of oral surfaces. However, its properties may be of great interest in other areas, where oral surfaces are not considered.

Finally, the data presented suggest that creaminess is perceived if the friction coefficient at low speed (below $100\ \mathrm{mm\ s^{-1}}$) is below a threshold value (for silicone friction coefficient threshold ≤ 0.25). The creamy perception, as well as lubrication properties of the milk, increased gradually with fat content, above 1% fat for silicone rubber. This indicated a good correlation between creamy attributes and measured friction coefficient, a result that validates the use of tribology as an analytical technique to better programme specific sensory products in product development and reformulation.

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