Microstructure Engineering of Emulsion-based systems for the Control of Satiation, Satiety, Hedonic Acceptability and Sensory Quality

by

Aaron Mitchell Lett

A thesis submitted to
University of Birmingham
for the degree of
DOCTOR OF PHILOSOPHY

School of Chemical Engineering
University of Birmingham

March 2016

Abstract

In response to the global obesogenic food environment, one current food reformulation approach to produce inherently “healthier” food products is to modify the consumer’s consumption behaviour by increasing the satiating power of foods. However, reformulation to promote satiety is often detrimental to sensory quality and thus negatively impacts on the products commercial success.

The motivation of this thesis stems from recent research highlighting sensory characteristics experienced during eating significantly impact upon food consumption behaviour, and that microstructural reformulation approaches have been shown to alter perception of sensory characteristics and hedonics in model and applied emulsion food systems. This thesis project took a unique multidisciplinary approach, combining understanding of food engineering, sensory science, nutrition and psychology, with the projects overall aim being to investigate how emulsion-based food products can be made more satiating whilst maintaining or improving the products sensory and hedonic qualities.

To achieve these aims model oil-in-water emulsion systems were designed. Oil droplet size (0.1 µm – 50µm) was the main microstructural variable investigated, as it is an integral part of an emulsions microstructure. From this, the influence of oil droplet size upon physical properties (viscosity and friction coefficient), sensory perception, hedonics and expected and actual food intake behaviour was investigated and inter-related where suitable.

The main finding of the work highlighted oil droplet size significantly affects hedonics, food intake behaviour expectations, actual food intake behaviour and the perception of numerous sensory attributes. It was shown the perception of Creaminess was a strong hedonic indicator, but interestingly it was also shown to significantly induce greater expectations of satiety and satiation. Structurally, Creaminess significantly increased with decreasing oil droplet size.

Through a preload study using the Sussex ingestion pattern monitor, expectations were shown to be reflected in actual food intake behaviour, with smaller droplets resulting in a significant 12% reduction of food intake.

Results from studies assessing the sensory properties of emulsions, strongly suggested that the mechanism in which oil droplet size modified Creaminess was through altered texture and mouthfeel. Instrumental characterisation of the emulsion samples indicated that this was a result of a combined influence of viscosity and droplet behaviour during oral processing, the sensory interpretation of these two physical variables being Thickness and Smoothness, reflecting Kokini's Creaminess predication equation.

Throughout this thesis the potential of emulsion structure on synergistically increasing both the satiety and hedonics of an emulsion based foods was realised and the understanding of emulsion structure on sensory properties, food intake and physical behaviour was furthered.

Acknowledgements

I would like to express my gratitude to my academic supervisors, Professor Ian Norton, Professor Martin Yeomans and especially Dr Jennifer Norton for firstly providing this unique opportunity, in addition to their support, guidance and supervision throughout. Thank you to members of both the Microstructure group (University of Birmingham) and Ingestive Behaviour Group (University of Sussex) for your support and companionship. Furthermore, thank you to both the Lett family and McEwan Clan for all your love.

Most of all I thank my Mum and Dad for always believing in me, the most fitting way I can repay you is too make you proud.

Dedicated to my parents

“Thanks and Praise”

Table of Contents

List of Figures

List of Tables

Nomenclature

τ_{max}	Shear stress
°C	Celsius
μ	Friction coefficient
μ_c	Continuous phase viscosity
μ_d	Dispersed phase viscosity
μm	Micrometre
A_r	Contact area
Ca	Capillary number
$D_{4,3}$	Volume mean diameter
E^*	Composite young's modulus
F_F	Frictional force
F_t	Point of tangential friction force assessment
g	Gram
Kg	Kilogram
L	Litre
M	Metre
ml	Millilitre
mm/s or U	Entrainment speed
mPas	Millipascal second
N	Newton
Pas	Pascal second

R	Droplet radius
r	Pearson correlation
R^2	Coefficient of determination
s^{-1}	Shear rate
W	Ball load
wt. %	Weight percentage
wt.%	Weight percentage
λ_μ	Viscosity ratio
σ	Interfacial tension

ANOVA	Hydrodynamic radius
BMI	Body mass index
CCK	Cholecystokinin
DEBQ	The Dutch Eating Behavior Questionnaire
GDP	Gross Domestic Product
GLP-1	Glucagon-like peptide-1
Kcal	Kilocalorie
NaCas	Sodium caseinate
O/W	Oil-in-water (emulsion)
O1/W/O2	Oil-in-water-in-oil (emulsion)
PCA	Principal component analysis
PDMS	Polydimethyl siloxane
PP	Pancreatic polypeptide
PYY	Peptide YY

RPM	Revolutions per minute
SEM	Standard error of the mean
SIPM	Sussex Ingestion Pattern Monitor
SRR	Slide roll ratio
VAS	Visual analogue scale
W/O	Water-in-oil (emulsion)
W1/O/W2	Water-in-oil-in-water (emulsion)
WHO	World health organisation
WPI	Whey protein isolate

Chapter 1. Introduction

1.1. Background

The increasing prevalence of overweight and obese populations globally is substantial. In 2014, 39% of the world's adult population were classified as overweight, with 13% classified as obese. This indicates that the worldwide prevalence of obesity has more than doubled between 1980 and 2014 (WHO, 2015). In parallel, global rates of obesity related non-communicable disease has risen, causing a significant global health and economic burden, with obesity being attributed to 8% of the overall burden of disease (Pomerleau *et al.,* 2003) and costing 0.09% - 0.61% of a country's GDP in direct and indirect costs (Muller-Riemenschneider *et al*., 2008).

Energy expenditure can be achieved through several methods, but there is only one mode of energy intake: food consumption. The failure to identify a defect in the metabolic control of energy expenditure, and the contrary observation of higher levels of energy expenditure in obese subjects (Jequier, 1984) has led to a focus on food intake to explain the aetiology of obesity. The current obesogenic food environment has been held primarily responsible, created by the increasing availability of cheap high calorie foods (Drewnowski and Specter, 2004). As the food industry dictates the global food environment, the food industry is being increasingly encouraged to contribute to the alleviation of the obesity burden through product reformulation and redesign, and to develop the next generation of foods (Norton, Moore and Fryer, 2007).

One approach is increasing the satiating power of foods and beverages, reducing the amount an individual consumes and increasing the time duration until the next meal is consumed; thus over a set time period a lower energy intake is achieved (Blundell, 2010; van Kleef *et al.*, 2012).

Sensory characteristics experienced during eating have been shown to impact upon food consumption behaviour (de Graaf, 2012). Such sensory characteristics have been predominantly texture related, with even subtle differences altering eating behaviour significantly (de Graaf and Kok, 2010; McCrickerd *et al.*, 2012; Yeomans and Chambers, 2011; Zijlstra *et al.*, 2009a; Zijlstra *et al.*, 2009b). This indicates that the degree to which certain sensory characteristics are perceived during eating changes the food or beverages capacity to generate satiety expectations. Sensory attributes which promote lower food consumption are referred to as *satiety-relevant sensory cues* (Hogenkamp *et al.*, 2011; Mattes and Rothacker, 2001; McCrickerd *et al.*, 2012; Yeomans and Chambers, 2011; Zijlstra *et al.*, 2009b). Identifying satiety-relevant sensory cues and engineering foods to promote them should increase a foods satiating power.

Nonetheless, disadvantages of using satiety-relevant sensory cues as a reformulation approach have been highlighted. Firstly, as learned sensory cues are associated with a given caloric value and satiety expectation, producing low-energy dense foods with sensory characteristics indicative of a greater energy content, which is not delivered on ingestion, typically results in compensatory intake (Yeomans and Chambers, 2011). Secondly, palatability has been shown to be inversely correlated to satiating power (Drewnowski, 1998; de Graaf, de Jong and Lambers, 1999; Holt *et al.*, 1995), a commercial disadvantage when we consider that hedonic appeal is a driver in consumer purchasing habits (Dhar and Wertenbroch, 2000). If hedonic properties can be maintained or even enhanced, an effective design approach would be to increase the satiating functionality of foods independent of energy content. Typically, energy dense foods associated with nutrients, such as fat, have a strong hedonic appeal (Prentice and Jebb, 2003). Additionally, fat is considered to be the least satiating macronutrient (Blundell, Green, and Burley, 1994; Blundell and Macdiarmid, 1997; Blundell and Tremblay, 1995). As fat within numerous food systems is structured in the form of an emulsion, emulsions are a suitable food model for reformulation or redesign to enhance satiety.

Microstructural reformulation and redesign approaches have been shown to alter sensory characteristics and hedonics in model and applied emulsion food systems (Akhtar *et al.*, 2005; Akhtar, Murray and Dickinson, 2006; de Wijk and Prinz, 2005; Kilcast and Clegg, 2002; Mela, Langley and Martin, 1994; Moore *et al.*, 1998; van Aken, Vingerhoeds and de Wijk, 2011; Vingerhoeds *et al.*, 2008). Subsequently, through manipulation of microstructural properties, the capability to change the satiating capacity of emulsion based products could be realised, through identifying and altering the perception intensity of satiety-relevant sensory cues. Additionally, understanding the physical construction of such sensory attributes, through relating quantitative instrumental measures of food structure with perception (Meullenet *et al.*, 1998; Szczesniak 1987), adds tremendous value for future product development. As such approaches, when fully understood, effectively permit the ability to "engineer" satiety, via satiety-relevant sensory cues, from the laboratory, through predicting consumer responses to newly developed food structures from instrumental data.

1.2. Overall thesis aims and themes

Taking a unique multidisciplinary approach, combining understanding of food engineering, sensory science, nutrition and psychology, the aim of this thesis is to advance the understanding of microstructural engineering approaches to produce emulsion-based food products, which have maintained or improved sensory and hedonic qualities, but modify food intake behaviour. Specifically, oil droplet size will be investigated in regards to its effect on emulsion physical, sensory, hedonic and satiating properties.

To achieve these aims model oil-in-water emulsion systems, with oil droplet sizes ranging from 0.1 µm – 50µm were investigated in regards to physical properties (viscosity and friction coefficient), sensory perception, hedonics and expected and actual food intake behaviour. The analysis of these results will be discussed and inter-related, where suitable, to focus on two main topics:

- Inter-relation between structure, physical properties and sensory outcomes.
- Inter-relation between structure, sensory outcomes and food intake behaviour.

1.3. Thesis layout

This manuscript is composed of seven chapters: an introduction; a literature review; four results chapters; and a conclusions and future work chapter.

- Chapter 1 is an introduction outlining background information and the rationale for the work.
- Chapter 2 reviews the relevant literature of the topics of interest in this thesis. Emulsion systems, sensory perception and its relationship with instrumental measurements and emulsion microstructure and the concept of satiety, satiety-relevant sensory cues and previous emulsion microstructure approaches to enhance satiety are discussed.
- Chapter 3 is the first results chapter, which is concerned with the physical and structural characterisation of the model emulsion systems that are used within chapter 3, 5 and 6 in the systems relation to sensory perception.
- Chapter 4 is the second results chapter, investigating the contribution of viscosity and friction in sensory perception through the use of novel hydrocolloid controlled model emulsion systems designed to either a iso-viscous or iso-friction criterion.
- Chapter 5 is the third results chapter, which investigates microstructural properties (oil droplet size and flavour), in relation to sensory characteristics, hedonics and the resulting expected food intake behaviours, of the model emulsions.
- Chapter 6 is the final results chapter, which looks at emulsion oil droplet size in relation to actual food intake behaviour via a preload paradigm study.
- Chapter 7 summarises the conclusions of the thesis and provides recommendations for future work.

1.4. Publications and conferences

Results and discussions obtained throughout this study have been published as follows:

Publications:

- Lett, A.M. and Norton, J.E. (In Press). Engineering Satiety. In: Tepper, B.J. and Yeomans, M.R. ed. *Flavor, Satiety and Food Intake*. Chichester: Wiley
- Lett, A.M., Norton, J.E., Yeomans, M.R. and Norton, I.T. (In preparation). Iso-viscous and Iso-friction oil-in-water emulsions: The contribution of viscosity and friction in sensory perception. *Food Hydrocolloids*
- Lett, A.M., Norton, J.E. and Yeomans, M.R. (2016). Emulsion oil droplet size significantly affects satiety: A pre-ingestive approach**.** .*Appetite, 96*, 18-24
- Lett, A.M., Yeomans, M.R., Norton, I.T. and Norton, J.E. (2016). Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour. *Food Quality and Preference, 47*, 148-155

Oral presentations (speaker underlined):

- <u>Lett, A.M.</u>, Norton, J.E., Yeomans, M.R. and Norton, I.T. Synergistic optimisation of sensory quality, preference and consumption behaviours in model emulsion based food systems through processing routes. *Advances in Food Processing – Challenges for the Future*, Campinas, São Paulo, 2014.
- Lett, A.M., Norton, I.T. and <u>Norton, J.E.</u> Relationship between tribology and sensory properties of oil in water food emulsions varying in droplet size. *2nd International Conference on BioTribology*, Toronto, 2014.

- Mills, T.M. and Lett, A.M. Lubrication behaviour of food-grade emulsions as a function of droplet size and emulsifier type. *2nd International Conference on BioTribology*, Toronto, 2014.

Poster presentations (presenter underlined):

- Lett, A.M., Norton, J.E., Norton, I.T. and Yeomans, M.R. Enhancing sensory characteristics, hedonics and expected food intake behaviour through emulsion design. *6th European Conference on Sensory and Consumer Research – EuroSense 2014: A Sense of Life,* Copenhagen, 2014.

- Lett, A.M., Norton, J.E., Yeomans, M.R. and Norton, I.T. Microstructural and physical properties of food emulsions in relation to sensory perception. *Food Structure and Functionality Forum Symposium - from Molecules to Functionality,* Amsterdam, 2014.

Chapter 2. Literature Review

Discussions contained within this chapter have contributed to the publication:

Lett, A.M. and Norton, J.E. (In Press). Engineering Satiety. In: Tepper, B.J. and Yeomans, M.R. ed. *Flavor, Satiety and Food Intake*. Chichester: Wiley

Purpose of Chapter

The aim of this chapter is to present a comprehensive review of the relevant literature for this thesis, which spans several disciplines. Firstly, an overview of emulsion structure, behaviour and their application in foods is presented.

Then, previous literature concerning emulsion structuring approaches to manipulate sensory characteristics is discussed including an overview of oral processing phenomena, sensory-instrumental relationships and food texture.

Finally, an overview of satiety and its theories, explaining our current understanding is given, with a specific focus on sensory mediated satiety and previous emulsion structuring approaches for satiety. Opportunities will then be highlighted through the inter-relation of the discussed sections.

2.1. Emulsions

2.1.1. Emulsion Structure

Ostwald (1910) first categorised emulsions, highlighting their structure to be constructed of two phases, a dispersed phase within a continuous phase. These two phases are immiscible, so to ensure the phases can be incorporated together within an application, the dispersed phases exists in the form of droplets dispersed within the continuous phase (McClements, 2005; McClements, 2007). The two immiscible phases within food products are typically oil and water. Emulsions would therefore be classified as an oil-in-water emulsion (O/W), in which the dispersed phase is oil and the continuous phase is water, or conversely a water-in-oil emulsion (W/O), whereby the dispersed phase is water and the continuous phase is oil. Additionally, advances in emulsion science has led to understanding in which emulsions with greater structural complexity (i.e. beyond a two phase system can be produced). An example of this is, multiple emulsions in which droplets of one phase are embedded within droplets of a second phase, which itself is dispersed within a continuous phase (Chung and McClements, 2014). The two main types of multiple emulsions being water-in-oil-in-water (W1/O/W2) and oil-in-water-in-oil (O1/W/O2) emulsions. This thesis focusses on oil-in-water emulsion systems.

Emulsion type can be further classified in regards to the size of the dispersed phase droplets, to either a macro, mini or nano emulsion. Macroemulsions possess a droplet size of approximately >1 μm, with miniemulsion (also known as sub-micron) droplets ranging from 0.1 - 1 μm and nanoemulsion droplets ranging from 0.01 – 0.1 μm (Chung and McClements, 2014; Tadros *et al.*, 2004). The majority of food emulsions are found as macro or miniemulsion, nanoemulsion use is currently limited in its applications due to complications with their production and a lack of understanding concerning their physiological effects on health (Tadros *et al.*, 2004).

The emulsified state of droplet within a continuous medium is achieved through the use of an emulsifier. An emulsifier is a surface-active molecule that absorbs at the oil-water interface during emulsification (see section 2.1.1.1.), achieving stability between the two immiscible phases through reducing interfacial tension and forming a mechanically cohesive barrier (McClements, 2005; McClements, 2009; Walstra, 1993). Emulsifiers are amphiphilic, having hydrophilic and lipophilic groups, so can act within and unite both phases to produce the most entropically stable state possible (Gunstone, Harwood and Dijkstra, 2007). The type and stability of the emulsion produced is dependent on both the emulsifier used and its concentration. There is a wide range of emulsifiers available. Within foods these commonly include edible surfactants (McClements, 2005), proteins (Dickinson, 1999) or solid particles (Pickering particles) (Pickering, 1907). A common example of emulsifier behaviour in an oil-in-water food emulsion is milk, where the naturally present casein and whey protein stabilises the milk fat-water interface (Dickinson, 2001).

2.1.1.1. Emulsion formation

Under quiescent conditions, the most thermodynamically stable state for oil and water to exist is in separate phases, in which oil is “layered” over the water, given that oil has a lower density. This phase separated state minimises the interfacial contact area between the two phases, minimising free energy (Pichot, 2010). In order to form an emulsion, energy is required to disrupt the two phases and disperse one within the other in the presence of an emulsifier, in a process called emulsification. The emulsions formation and droplet size, is dependent on emulsifier absorption to the interface (concentration and type), droplet stability (see section 2.1.2) and droplet break up. The disruption of oil into smaller volumes to form droplets (droplet break up), is achieved typically by mechanical agitation using processing equipment, which promotes hydrodynamic flow conditions which apply an external disruptive force greater than that of the interfacial force, also known as Laplace pressure, the pressure differential between the inside and the outside of an emulsion droplet maintaining the droplets characteristic spherical shape (McClements, 2005; Walstra, 1993). This results in oil droplets being formed and dispersed within the aqueous continuous medium. Droplet size will continue to decrease if further disruptive forces are greater than that of the interfacial force, or will increase if insufficient emulsifier is present to stabilise the droplet interface (which will increase in surface area with decreasing droplet size) leading to behaviours promoting droplet growth (see section 2.1.2).

Common processing equipment for emulsification includes high shear mixers and high pressure homogenisers. High shear mixing involves a mechanical mixing head which rotates at high speed (1-10000 rpm) whilst submerged within the fluid mixing medium; duration, mixing head geometrics and speed (RPM) are user decided processing variables (Zhang, Xu and Li, 2012). High pressure homogenisation for emulsification involves passing a pre-emulsion, formed using equipment such as a high shear mixer, within a gap in a homogenisation valve. The gap is a localised area in which high turbulent flow and shear exists which promotes droplet break up (Floury *et al.*, 2004; Lee *et al.*,2014). High pressure homogenisation achieves droplet dispersions with greater uniformity and smaller size than high shear mixing, dependent on processing variables used (operating pressure and number of passes).

2.1.2. Emulsion behaviour

The term “emulsion stability” refers to the ability of an emulsions resistance to change, in its physicochemical properties (McClements, 2005; McClements, 2007).

The inherently thermodynamically unstable nature of emulsions, and the droplets state of constant motion (due to gravitational forces and Brownian motion), promotes a dynamic system in which droplet collisions, and interactions between the systems constitutes promotes instability, leading to a thermodynamic propensity for phase separation to achieve the most entropically stable state possible. Complete breakdown and phase separation is therefore inevitable over time (McClements, 2005; McClements, 2007; Taylor, 1998). A suitable example of this breakdown behaviour is the rapid phase separation observed in salad dressings once agitated and then left under quiescent conditions, with consumers instructed to redisperse the system through “shaking” before use. An instable emulsion system maybe the result of a number of common physicochemical mechanisms (see Fig. 2.1), and discussed below:

2.1.2.1. Gravitational separation

Gravitational separation is a density dependent instability behaviour in which stabilised oil droplets either move upwards, due to their lower density than the continuous phase, in a process called “Creaming” or move downwards, due to their higher density than the continuous phase, in a process called “Sedimentation” (McClements, 2007). The rate of gravitational separation in either behaviour is proportional to the difference in density and can be predicated by Stokes law (McClements, 1998). Droplets within gravitational separated emulsions can be redispersed with mild agitation, providing coalescence or strong aggregation has not occurred (McClements, 1998). Gravitational separation promotes further droplet instability behaviours, such as flocculation and coalescence, as it encourages closer contact of droplets (McClements, 2007).

2.1.2.2. Flocculation

Flocculation is a type of droplet aggregation, in which droplets associate to form “flocs”, whilst maintaining their individual droplet integrity (McClements, 2007). Flocculation behaviour can classified as either bridging flocculation or depletion flocculation (Dickinson, 2010). *Bridging flocculation* occurs when a direct link or "bridge" between neighbouring droplets is formed. It is result of insufficient emulsifier coverage on the droplet interface (Dickinson, 1989; Dickinson, 2010). *Depletion flocculation* occurs between droplets when the continuous phase contains excess non-absorbed entities, such as emulsifier, which promotes an attractive interaction between droplets (Dickinson, 1989; Dickinson, 2010). Unlike bridging flocculation, depletion flocculation is weak and reversible (Somasundaran, 2006).

2.1.2.3. Coalescence

Coalescence is a thermodynamically driven droplet destabilisation process in which two or more emulsion droplets merge together producing one larger emulsion droplet (McClements, 2007). Coalescence occurs as it promotes the emulsions movement towards a more thermodynamically stable state, decreasing the contact surface area between the two immiscible phases (McClements, 2005). Coalescence additionally promotes other destabilisation behaviours such as gravitational separation.

2.1.2.4. Ostwald ripening

Ostwald ripening is a process which sees larger droplet size increase at the expense of smaller droplets. This behaviour occurs due to the diffusive mass transport of dispersed phase, between droplets, through the continuous phase driven by the Laplace pressure differential between droplets (McClements, 2007; Taylor, 1998).

2.1.2.5. Phase inversion

Phase inversion is a process in which the continuous phase and the dispersed phase, swap their phase categorisation i.e. an oil-in-water emulsion becomes a water-in-oil emulsion, or a water-in-oil emulsion becomes an oil-in-water emulsion (McClements, 2007). Phase inversion can be an essential step in the production of some emulsion based foods, such as butter or margarine (McClements, 1998). However, in a formed emulsion product it is an undesirable behaviour triggered by compositional or environmental condition changes, such as dispersed phase volume fraction, emulsifier type or concentration, temperature, solvent presence and mechanical agitation. Phase inversion is complex, involving aspects of droplet flocculation and coalescence, and can only be achieved in certain emulsion formulations, which do not degrade to a phase separated state (Binks, 1998; Cosgrove, 2010; McClements, 1998).

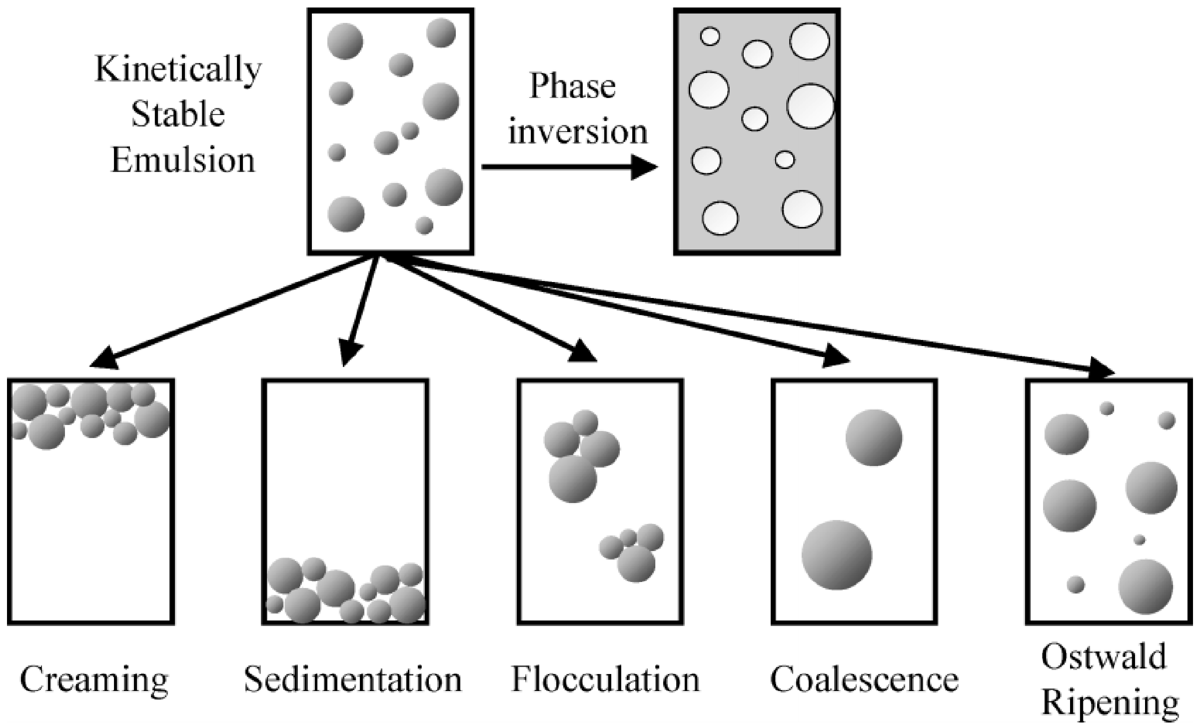


Fig. 2.1. Schematic diagram of common emulsion instability behaviours. Image reproduced from McClements, 2007.

2.1.3. Emulsion applications in common foods

Numerous food products contain the two immiscible phases, oil and water, as part of their overall microstructure. A vast majority of these products are or contain the water and oil phases as an emulsion, or have been emulsified during a certain state of their production (Chung and McClements, 2014). Structuring these immiscible constitutes in the form of an emulsion allows the product to be produced to the desired quality of the manufacturer, as the emulsion structure will largely determine the products functional attributes, such as stability and sensory qualities (see section 2.2).

Common examples of oil-in-water emulsion based foods include: milk, milkshakes, cream, cream liqueurs, smoothies, deserts, ice cream, condiments such as mayonnaise, dressings, sauces such as Hollandaise or Béarnaise, soups, fresh cheese, batters and yoghurts. Common examples of water-in-oil emulsion based foods include butter and margarine. This list highlights the diversity of emulsions in common food applications. This thesis will focus on liquid model oil-in-water emulsion systems.

2.2. Sensory

Understanding sensory sensations arising from the consumption of foods and beverages requires an appreciation of the influences, principles and mechanisms involved in sensory perception. A multitude of textural, flavour, auditory and visual contributions combine in perception (Chen, 2009). However, this thesis will predominantly focus on food texture. Concentrating on texture, this approach itself requires understanding of both human physiology and food structure and its breakdown during oral processing.

2.2.1. Oral processing and perception

2.2.1.1. Oral cavity

The oral cavity can be defined as the void space between the lips and the velum (see Fig. 2.2) (Chen, 2009). The oral cavity hosts the processing and manipulation of food pre-deglutition and is a key stage in food assessment and an essential first step in food digestion. The universal characterisation of the oral cavity is problematic due to its complexity, influenced by an individual's gender, ethnicity, age and health status (Chen, 2009). However, the average "full" mouthful, (i.e. oral volume size), for adult males is approximately 30g and females 25g, as shown using water (Medicis and Hiiemae, 1998). This average "full" mouthful will of course vary with the particular food consumed, dependent on its structure (Chen, 2009; Medicis and Hiiemae, 1998). In reality humans consume mouthfuls below the actual capacity the oral cavity can accommodate (Vincent and Lillford, 1991).

2.2.1.2. Tongue

At the floor of the oral cavity sits the tongue (see Fig. 2.2). The tongue is a dynamic and integral muscle for food manipulation, management, processing, discrimination and swallowing (Chen, 2009; Heath, 2002).

The tongues movement and shape is muscular controlled and its surface consists of stratified squamous epithelium, with numerous papillae, in which the majority of taste buds are embedded within (Chen, 2009; Dresselhuis *et al.*, 2008; Guinard and Mazzucchelli, 1996; van Aken, Vingerhoeds and de Hoog, 2007). Receptors also are embedded within the tongue relaying information concerning oral processes; of interest in the assessment of food texture is mechanoreceptors. Mechanoreceptors are sensory receptors which response to mechanical pressure or distortion and are also present within the masticatory muscles and tendons, the periodontal membrane surrounding the roots of teeth and embed with the palate and gums (Guinard and Mazzucchelli, 1996). As such, mechanoreceptors play a primary role in the assessment of food breakdown and texture throughout oral processing.

2.2.1.3. Saliva

Human saliva is a complex biological fluid consisting of approximately 98% water and 2% organic and inorganic substances, such as electrolytes, mucus, antibacterial compounds, lingual digestive enzymes and proteins such as mucin, and has an average pH of approximately 6.8 (Jenkins, 1978; Levine et al., 1987; Sarkar, Goh and Singh, 2009).

Numerous factors affect the amount and the composition of secreted human saliva. However the main stimuli for increased salivation is food (Vingerhoeds *et al.*, 2005). 90% of all saliva secretions originate from three major salivary glands located within the oral cavity (Humphrey and Williamson, 2001) and the quantity produced ranges from 0.3 - 7 ml saliva per minute (Edgar, 1990) with about 0.5–1.5 L of saliva secreted per day (Humphrey & Williamson, 2001; Vingerhoeds *et al.*, 2005).

In food oral processing, Saliva facilitates initial food digestion, oral and bolus lubrication and protection, food dilution and enhancement of taste (Chen, 2009; Humphrey and Williamson, 2001). Saliva also interacts with food components, enabling structural formation or breakdown, and thus changes the sensorial, and particularly the textural, properties of foods (van Aken, Vingerhoeds and de Hoog, 2007; Vingerhoeds *et al.,* 2009).

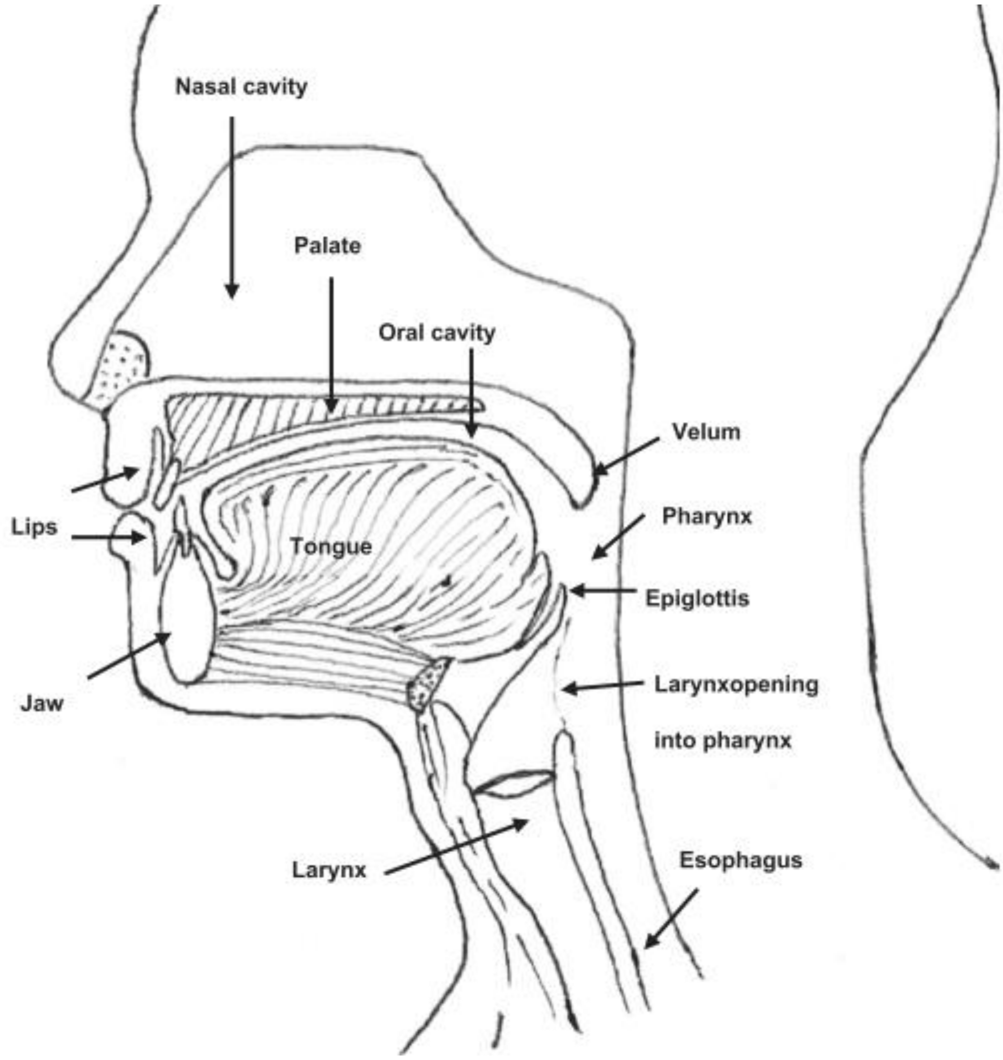


Fig. 2.2. An anatomic diagram of the oral organ. Image reproduced from Chen, 2009.

2.2.1.4. Oral processing

Oral processing is a dynamic and complex process controlled by both a physiological process mediated by the central nervous system and a physical process responding to the mechanical and geometrical properties of the food (Chen, 2009). The process facilities the breakdown of food structure via mechanical (teeth, palate, tongue) and chemical (saliva) mechanisms, to achieve a food bolus which is safe to swallow. Oral processing can be partitioned into three key phases (Guinard and Mazzucchelli, 1996) and will be discussed in relation to liquid products. An understanding of oral processing is needed as it is associated with this process is the sensory perception of food (Chen, 2014).

The first process is *food introduction and processing*, in which the liquid food is mixed with saliva, via the physical movement of oral constitutes, such as the tongue. Within this stage the liquid will be subjected to shear and will remain largely as a bulk entity (Stokes, Boehm and Baier, 2013). Secondly, *oral propulsion* occurs, in which the tongue moves towards the palate inducing flow and squeezing the fluid against the palate. This readies the bolus for swallowing. Within this stage the liquid will be subjected to high shear and will be manipulated against the palate, creating thin-films (Stokes, Boehm and Baier, 2013). These first two phases are particularly important in regards to sensory perception of the foods texture, as textural sensations are derived primarily during the progressive change of the foods structure (Malone, Appelqvist and Norton, 2003). The third and final stage is *swallowing*, in which the oral cavity is cleared and the fluid is expelled down the oesophagus.

For liquids, oral processing is a relatively quick process compared to that of semi-solids and solids, with oral residence time lasting approximately two seconds (Ertekin and Aydogdu, 2003). Its short oral residence time is primarily a result of its structure demanding less mechanical breakdown. Conveniently, this allows clearer definition between oral processing phases, making understanding of fluid oral processing simpler.

2.2.1.5. Texture perception

Food texture is a multi-parameter attribute, which can be defined as ‘the sensory manifestation of the food structure and the manner in which this structure reacts to applied force” (Jowitt, 1974; Szczesniak, 1963). Although this thesis will focus on oral interpretations of food texture, visual, auditory, olfactory and touch are also important senses in textural perception.

Food texture is typically described by a term, or defined as an attribute or a number of attributes. For example Thick, Smooth, Hard, Creamy are all common textural terms to describe sensory aspects of a product. This ability for consumers to discriminate a specific textural attribute and describe it, highlights texture as an integral part of a food products sensory profile. Texture can be used as a sensory indicator of a specific foods quality, and is used by consumers to determine whether a food should be discriminated or appreciated, and to what degree (Scott and Downey, 2007). For example, the texture of “Hard” stale bread, “Soft” potato crisps or a “Flat” soda beverage may be an indication that the food product has passed a level of quality, which the consumer deems no longer enjoyable or safe. Conversely, “Hard” sugar candy, “Soft” set-desserts or “Flat” non-carbonated beverages, are attributes consumers desire in these products.

Receptors throughout the oral cavity detect the wide range of sensations which can occur throughout eating, and provide the consumer with textural information concerning the product. Although there are a number of oral receptor types, Mechanoreceptors responding to mechanical pressure and distortion are primarily involved in the assessment of a foods physical texture (Guinard and Mazzucchelli, 1996). Physical texture is comprised of physical properties such as viscosity and friction, which are determined by the foods structure (Chung and McClements, 2013; Stokes, Boehm and Baier, 2013).

2.2.2. Texture-instrumental relationships

Characterising and understanding food texture through instrumental measurement techniques is a highly desired approach in food development research. These techniques not only act as tools of quality control, but to some extent allow for predictions of consumer responses, by relating quantitative measurements of food structure with perception (Meullenet *et al.,* 1998; Szczesniak 1987). Despite the multi-dimensional influences which form sensory attributes and the highly complex process of food oral processing, instrumental approaches have been developed which provide insight into oral behaviour, food structure breakdown and the physical make-up of sensory attributes (Stokes, Boehm and Baier, 2013). A result of the shears, stresses and forces exerted during the dynamic process of oral processing being somewhat replicated (Stokes, Boehm and Baier, 2013; van Aken, Vingerhoeds and de Hoog, 2007).

Common instrumental approaches within fluid foods have typically considered rheological and tribological characterisation, in order to understand the foods bulk and surface properties (Stokes, Boehm and Baier, 2013; van Aken, Vingerhoeds and de Hoog, 2007). These approaches have shown success in understanding the relationship between physical instrumental measures and sensory attribute perception. This is because they characterise the physical state of the fluid food as a bulk entity and as a thin-film (Fig. 2.3.), and during the oral processing of fluids there is a transition from bulk property dominance to thin-film property dominance (see section 2.2.1.4). Nevertheless, a limitation of using these two techniques is that the transition from the bulk to the surface property dominate region is not and cannot be measure, however in actual oral processing this transition does occur. This thesis will focus on rheology and tribology techniques for the assessment of food texture and its relationship with sensory perception.

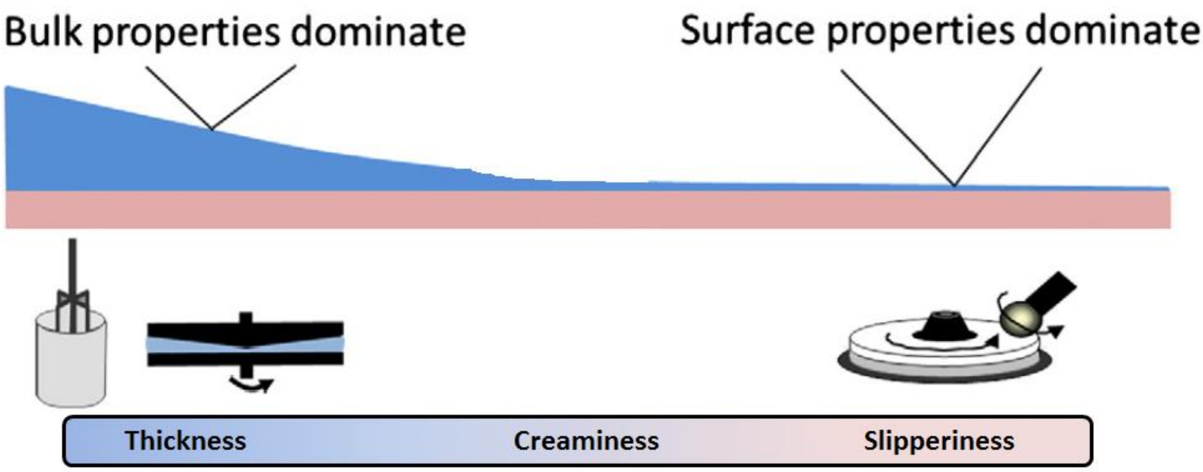


Fig. 2.3. Fluid food transition from rheology-dominated (bulk properties) to tribology-dominated (surface properties) region, throughout oral processing, further indicating possible techniques for their measurement and where certain sensory attributes may lie in relation. Image adapted from Stokes, Boehm and Baier, 2013.

2.2.2.1. Oral rheology and sensory perception

Rheology is the study of the deformation and flow of matter. Oral rheology is concerned with the flow and movement of food around the oral cavity, and in a textural context, the mechano-sensations arising from the foods resistance to shear imposed by oral entities such as the tongue (Chen and Engelen, 2012). Sensory-instrumental relationship research centred around rheological properties has become one of the most extensively investigated physical properties in relation to textural perception of liquid foods.

The simplest model used to describe oral fluid dynamics is shear flow. Using this approach, specific sensory attributes have associated with viscosity at single shear rates representative of the physical force conditions experienced within the mouth. The earliest work of this nature was carried out by Wood (1968). By using hydrocolloid thickened soups, Wood (1968) indicated 50 s^{-1} is representative of shear rates experienced in the mouth and showed perceived Thickness correlated most strongly with viscosity at this shear rate. Since Woods

(1968) pioneering work, further studies have additionally shown strong correlations between viscosity and perceptions of Thickness, with the relationship being one of the most extensively investigated in fluid foods (Akhtar, Murray and Dickinson, 2006; Cutler, Morris and Taylor, 1983; Kokini, Kadane and Cussler, 1977; Morris and Taylor, 1982; Morris, Richardson and Taylor, 1984; Shama and Sherman, 1973). Although, 50 s^{-1} has been, and still is, used extensively as a convenient value to represent the shear rate experienced during oral processing of non-Newtonian liquid foods, a number of these extensional studies highlighted that this value could be misleading, as oral processing exerts a range of shears upon a food, and therefore correlations between thickness and viscosity, at a given shear rate, could differ dependent on the food's rheology. For example, Sharma and Sherman (1973) evaluated a number of fluid and semi-fluid foods. For a limited number of foods, results agreed with Wood (1968). However, shear rates between 10-1000 s^{-1} were experienced when orally evaluating thickness, with the shear rate which best correlated with the food increasing with decreasing viscosity of the fluid. Nevertheless, viscosity at 50 s^{-1} is still thought to be a reasonable predictor of liquid foods at an elementary level (Chen and Engelen, 2012).

As well as Thickness perception, viscosity has also been indicated to be influential in perception of other common textural attributes within fluid foods, such as Slipperiness and Creaminess (Kokini and Cussler, 1984; Kokini, 1987).

Slipperiness has been shown to correlate with the reciprocal of the sum of the viscous force and friction force (Kokini, 1987) and Creaminess has been shown to be largely depend on the perception of both independent attributes Thickness and Smoothness (Kokini, Kadane and Cussler, 1977); Thickness is a sensory interpretation of viscosity and Smoothness has been shown to be correlated with the reciprocal of the friction force between oral surfaces, i.e. tongue and the palate (Kokini, 1987). These conclusions of Creaminess are reflected in Kokini's Creaminess prediction equation (Kokini and Cussler, 1983) (See equation 2.1).

$$Creaminess = Thickness^{0.54} X\ Smoothness^{0.84}$$

Eq. 2.1

However, unlike Thickness perception, given the definitions, it is clear that both these textural attributes are not thought to be fully characterised by just viscosity. Early work first concluding that viscosity did not satisfactorily explain the physical make-up of these attributes led to studies investigating surface properties (Chen and Stokes, 2012). This work highlighted that the understanding of such attributes improved when surface related properties were considered, as these new approaches provided insight into material properties that bulk rheology measures could not deduce (Kokini and Cussler, 1984; Kokini, 1987; Kokini, Kadane and Cussler, 1977; Malone, Appelqvist and Norton, 2003). As such, the following section will describe our current understanding of tribology and its relationship with sensory perception.

2.2.2.2. Oral tribology and sensory perception

Tribology is the science of adhesion, friction and lubrication of interacting surfaces in relative motion. In an oral processing context, the “interacting surfaces in relative motion” are oral constitutes, such as the compression of the tongue and palate against one another. As we know oral processing moves from bulk property to surface property dominance (see Fig. 2.3), the behaviour of thin films within the oral cavity, generated by interacting oral surfaces, are therefore clearly of interest in relation to sensory perception. The influence of the surface properties of these thin-films, have long been recognised as an important factor in sensory perception, due to the lack of complete understanding when only considering bulk rheology (Hutchings and Lillford, 1988; Kokini, Kadane and Cussler, 1977). However until recently, ‘oral tribology’ and surface properties, have been largely disregarded, with only more recent work considering their influence (Chen and Engelen, 2012; Chen and Stokes, 2012).

The main physical property investigated via tribology in relation to sensory perception is friction. Categorising the friction of a fluid food (lubricant) involves measuring the lubricants friction coefficient over a range of entrainment speeds. Monitoring friction coefficient across entrainment speed generates a Stribeck curve. An idealistic Stribeck curve will map the lubricate under three distinct regimes of lubrication (Boundary, Mixed and Hydrodynamic) (See Fig. 2.4). The regimes will depend on the extent of lubricate entrainment and the hydrodynamic lubricate pressure between the two surfaces, both of which will increase with increasing entrainment speed. These are boundary, mixed and hydrodynamic lubrication, and will be discussed in turn.

Boundary lubrication – This is observed at low entrainment speeds, when the hydrodynamics cannot support the applied load. This results in high friction as both surfaces are in contact (Stokes, Boehm and Baier, 2013).

Mixed lubrication – With increasing entrainment speeds, hydrodynamics become sufficient to begin parting the tribo-surfaces. Within this regime partial contact is observed, lowering friction relative to the boundary lubrication regime. As entrainment speed increases, surface contact further reduces until full separation occurs where friction coefficient reaches a minimum and the hydrodynamic lubrication regime begins (Spikes, 1997).

Hydrodynamic lubrication – This is characterised by an increase in friction coefficient with entrainment speed, determined by drag caused by the lubricant, a result of its viscosity. Within this regime hydrodynamics are sufficient to cause complete separation of tribo-surfaces.

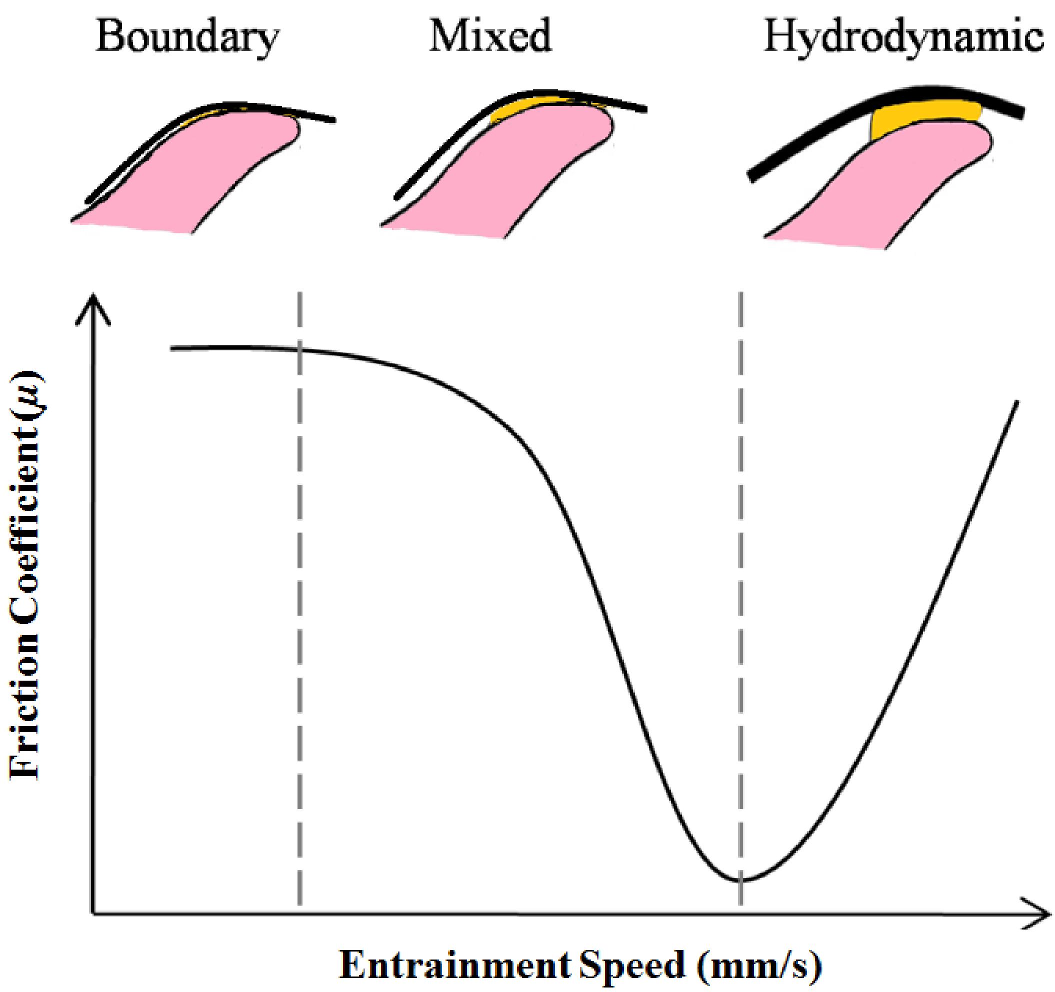


Fig. 2.4. Schematic of a Stribeck curve showing the three principle regimes of lubrication: boundary, mixed and hydrodynamic, where the surfaces are in full contact, partial separation or full separation, respectively. Surfaces are depicted as the tongue and palate.

Soft tribology techniques are deployed in studies of ‘oral tribology’ in which typically one surface has a low young’s modulus and is deformable under the contact pressure, as to mimic the deformability of oral surfaces such as the tongue. Some novel studies have actually looked to use human oral analogue surfaces, such as pigs tongue (Dresselhuis *et al.*, 2008). However, due to issues with the availability, data reproducibility, animal-animal variation and tissue degradation of tongue surfaces, artificial and well defined soft- tribometer surfaces such as PDMS (polydimethyl siloxane) or silicone elastomer are typically used.

Tribometer configuration can vary widely. The work within this thesis was carried out on a mini traction machine (MTM) using silicone elastomer. As such, the tribometer configuration consisted of a rotating silicone elastomer disc (diameter: 46mm, thickness: 4mm) mounted in a sample pot, on which a rotating ¾ polished steel ball connected to a shaft was lowered at a 45° angle on to the disc, creating a contact point and the site of friction assessment between the two surfaces, which was measured by a laterally placed force transducer attached to the ball shaft (see Fig. 2.5). Friction coefficient was assessed across 20 averaged entrainment speeds, logarithmically set between 1 to 1000 mm/s, using a 50% slide-roll ratio, between the ball and the disc, to mimic the complex sliding and rolling friction associated with oral processing conditions. Plotted against entrainment speed, Stribeck curves are obtained via measuring friction coefficient in 6 consecutive sweeps, alternating between ascending and descending sweeps. Such a set up in the assessment of liquid food systems has shown promise in mimicking thin film behaviour associated with oral processing (Mills, 2011), as such it provides a suitable instrumental tool to assess frictional properties of fluid foods in relation to sensory perception.

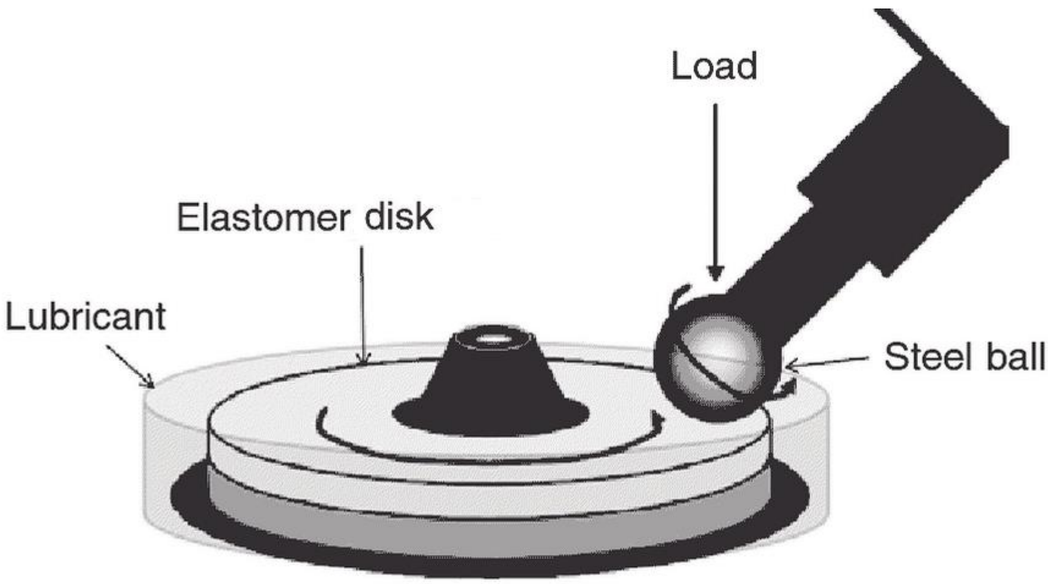


Fig. 2.5. MTM ball on disc tribometer configuration schematic. Image adapted from Chen and Engelen, 2012.

As previously mentioned, surface properties in relation to sensory perception have long been disregard until recently. However, there is a small, but growing, body of evidence from the literature in relation to friction and texture perception. Kokini's work (1977, 1983, 1987) is considered to be pioneering in food tribology research and led to the design of Kokini's model of oral lubrication (Kokini, 1987) (See Fig. 2.6). This model quantified physical measures of friction and viscosity in fluid and semi-solids foods, in relation to the perception of sensory attributes Smoothness and Slipperiness. "Smoothness" correlated with the reciprocal of the friction force between oral surfaces and "Slipperiness" with the reciprocal of the sum of the viscous force and the friction force. Creaminess was shown to be a combined effect of the perception of Thickness and Smoothness (see equation 2.1). To this notion Creaminess is therefore "indirectly" related to friction, through the perception of Smoothness, and thus can be partly explained by tribology.

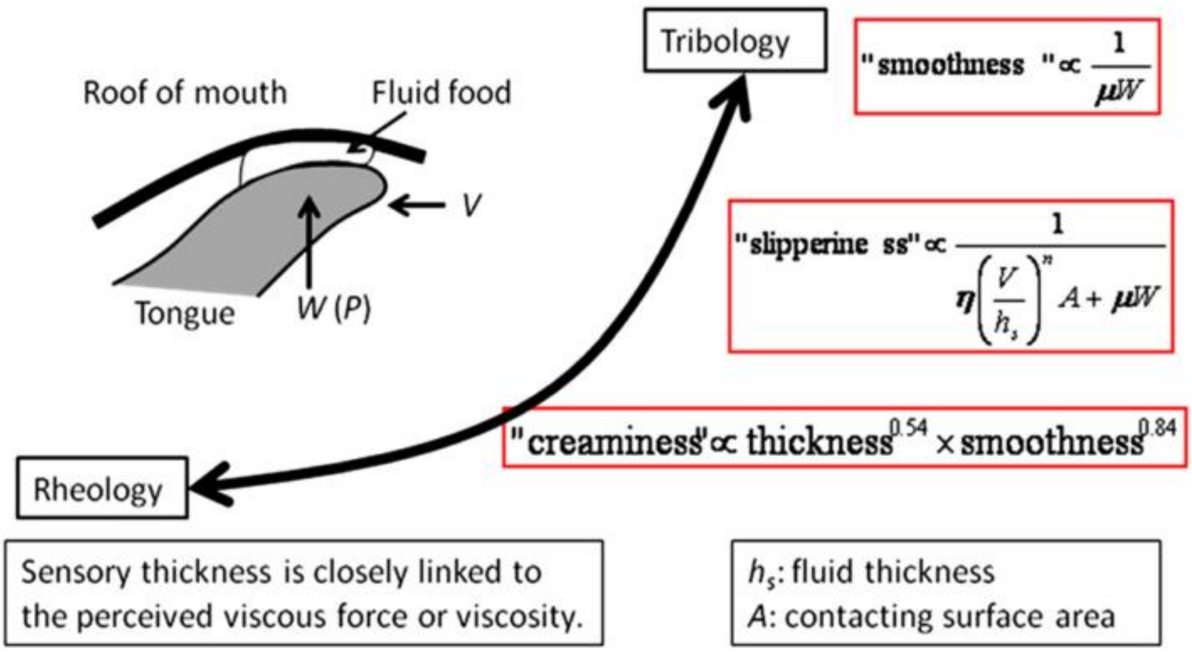


Fig. 2.6. Kokini's model of oral lubrication. The given sensory properties show the varying contribution of oral lubrication and food rheology, highlighting a transition between the two regimes, where η is the viscosity, V is the speed of tongue movement, W is the surface load, μ is the friction coefficient. Image reproduced from Chen and Stokes, 2012.

As a result of Kokini's findings, within the past 15 years emulsion based products have received increasing attention, due to the association between fat and textural attributes, such as Creaminess (Chen and Engelen, 2012; de Wijk, Rasing, and Wilkinson, 2003; Frost and Janhoj, 2007).

de Wijk and Prinz, (2005) showed that increasing friction coefficient resulted in increased sensations of Roughness, Prickling, Dryness and Mealiness, and decreasing friction coefficient increased sensations of Creaminess, Fattiness, Stickiness, Slipperiness and Smoothness. These results were shown in mayonnaise and custards. Similar results have also been shown using milk (Chojnicka-Paszun, de Jongh and de Kruif, 2012), yoghurt (Sonne *et al.,* 2014), thickened creams (Selway and Stokes, 2013) and white sauces (de Wijk and Prinz, 2007). Within these studies the emulsion structure had been changed in order to vary friction coefficient. Greater fat content (de Wijk and Prinz, 2007; Chojnicka-Paszun, de Jongh and de Kruif, 2012; Selway and Stokes, 2013) and smaller particle/droplet size (de Wijk and Prinz, 2005; Sonne *et al.,* 2014), were shown to lower friction coefficient and increase sensations of attributes such as Creaminess.

Although work investigating friction-sensory relationships is limited, to date the majority of studies within emulsion based products have focused on fat content, with only one study investigating a limited range of oil droplet sizes (de Wijk and Prinz, 2005). To identify such relationships these studies have correlated sensory scores with friction coefficient at certain entrainment speeds. Although simplistic, this approach has clearly identified frictional influences in perception of textural attributes, with certain attributes being further related to perception in particular regimes of lubrication. For example, using hydrocolloid thickened sunflower oil-in-water emulsions, Malone, Appelqvist and Norton (2003), showed that the perception of Slipperiness is strongly correlated with friction coefficient within the mixed regime of lubrication. However, limitations of directly correlating friction coefficient of a product with its mean sensory score, is the remarkably different physical and chemical conditions the product experiences in the mouth, compared to the tribometer. This can result in no, or misleading, correlations if the initial structure changes orally, such as through saliva-induced structural formation or instability mechanisms within emulsion systems (van Aken, Vingerhoeds and de Hoog, 2007) (See section 2.2.3).

2.2.3. Emulsion microstructure and sensory properties

Microstructural engineering of food emulsions has emerged as an integral part of emulsion food development for numerous consumer responses. A microstructure-function relationship, which is of considerable interest to food manufacturers, is the relationship between emulsion microstructure and the product's resulting sensory characteristics. This is due to the products sensory profile is fundamental to the consumers enjoyment, and therefore largely its commercial success. Furthermore, the products sensory profile also has additional implications, such as its role in pre-ingestive satiety (see section 2.3.2). Listed below are some of the common approaches used to manipulate O/W emulsion microstructure to change textural sensory properties.

2.2.3.1. Oil droplet size

The size of the oil droplets present within O/W emulsions are known to play a critical role in determining the emulsions physical properties. As a result, they are of interest as a structural property which could change textural perception. Despite little research being carried out with droplet size as a dependent variable, conflicting results have been shown. Some studies have concluded that oil droplet size has no significant effect on textural perceptions (Akhtar *et al.*, 2005; de Wijk and Prinz, 2005; Mela, Langley and Martin, 1994; Vingerhoeds *et al.*, 2008). However, other studies have reported that oil droplet size significantly affects perception, particularly Creaminess (Kilcast and Clegg, 2002; Lett *et al.,* 2016). These conflicting findings are a result of the larger oil droplet size range used in Lett's study (2016) (See Table 2.1) and the fat type used by Kilcast and Clegg (2002), in which a semi-solid fat was used that aggregated on cooling, increasing viscosity through floc formation.

2.2.3.2. Emulsifier

The physical and chemical conditions of the mouth can change the behaviour of oil droplets within the emulsion, which has implications on the emulsions physical properties, and therefore sensory properties. Oil droplets in the presence of saliva, under *in vivo* and *in vitro* conditions, have shown that initially stable emulsions, can flocculate or coalesce. Saliva-induced flocculation occurs almost instantaneously within weakly negatively charged, neutral and positively charged emulsions, a result of the emulsifier used (Vingerhoeds et al., 2005). Weakly negatively charged and neutral emulsions demonstrate depletion flocculation, which has been hypothesised to be a result of mucins and salivary complexes present, promoting attractive interactions between droplets. Positively charged emulsions, however, demonstrate bridging flocculation, induced by electrostatic attractions between salivary proteins and emulsion droplets (Sarkar, Goh and Singh, 2009; Silletti et al., 2007). As a result, saliva-induced flocculation behaviour can impact on the emulsions sensory profile, through the structures changing physical properties. Particularly of note, is floc formation increasing viscosity (Silletti et al., 2007). However, this has not necessarily increased perception of attributes associated with viscosity (see section 2.2.2.1); Depletion flocculate emulsions do, having been shown to produce Creamy and Fatty sensations, however bridging flocculated emulsions do not, producing Rough and Dry sensations, due to the retention of firm insoluble aggregates and droplets on the tongues surface, which are thought to override sensations produced with the floc induced increase in viscosity (Vingerhoeds et al., 2009). Shear induced coalescence of oil droplets, has also been shown to occur during oral processing.

Low stability protein-stabilised oil-in-water emulsions were shown to have a higher tendency towards coalescence under shear produced by tribometer surfaces and oral surfaces. This was shown to lead to a decrease in friction coefficient and an increase in sensations of Creaminess and Fattiness (Dresselhuis et al., 2008).

2.2.3.3. Fat content

Increasing oil volume fraction, therefore oil droplet volume fraction, has shown to increase the perception of attributes such as Richness, Creaminess, Smoothness, Thickness and Fattiness (Chojnicka-Paszun, de Jongh and de Kruif, 2012; Chung and McClements, 2014; de Wijk and Prinz, 2005; de Wijk and Prinz, 2007; Kilcast and Clegg, 2002; Mela, Langley and Martin, 1994; van Aken, Vingerhoeds and de Wijk, 2011; Vingerhoeds *et al.*, 2008). A relatable example of these findings would be the sensory difference between skimmed and full-fat milk, with the sensory differences being partly attributed to the higher fat product having greater viscosity and lower friction coefficient (Chojnicka-Paszun, de Jongh and de Kruif, 2012; Chung and McClements, 2014).

2.2.3.4. Hydrocolloids

Increasing the complexity of O/W emulsions, by the addition of certain hydrocolloids is extremely common practice to produce desired functionalities. The use of biopolymers, particularly those which thicken or gel, enhance viscosity, and therefore viscosity related textural perceptions such as Thickness (Akhtar *et al.,* 2005; Akhtar, Murray, and Dickinson, 2006; Tarrega and Costell, 2006). The addition of hydrocolloids is therefore a common approach to maintain physical properties, such as viscosity, when removing fat from emulsion based products.

Table 2.1. Summary of studies examining the effect of microstructural properties on sensory perception in liquid oil-in-water emulsions.

Structural variable		Food model	Reference
Oil droplet size	0.5-2 µm	Model O/W emulsions	Akhtar *et al.*, 2005
	2-6 µm	Mayonnaise	de Wijk and Prinz, 2005
	3-9 µm	Artificial creams	Kilcast and Clegg, 2002
	0.2-50 µm	Model O/W emulsions	Lett *et al.*, 2016
	0.5-2 µm	Model O/W emulsions	Mela, Langley and Martin, 1994
	1-8 µm	Model O/W emulsions	Vingerhoeds *et al.*, 2008
Emulsifier	NaCas, WPI, β-lactoglobulin, β-casein and Tween 20	Model O/W emulsions	Vingerhoeds *et al.*, 2005
	Whey protein in different pH emulsions	Model O/W emulsions	Vingerhoeds *et al.*, 2008
	Whey protein (depletion floc) and lysozyme (Bridging floc)	Model O/W emulsions	Vingerhoeds *et al.*, 2009
	Lactoferrin and β-lactoglobulin	Model O/W emulsions	Sarkar, Goh and Singh, 2009
	NaCas/WPI, WPI and octenylsuccinate starch	Model O/W emulsions	Dresselhuis *et al.*, 2008
Fat Content	0.06-8%	Milk	Chojnicka-Paszun, de Jongh and de Kruif, 2012
	0-15%	Vanilla custard desserts	de Wijk and Prinz, 2005
	0-72%	Custard desserts, Mayonnaise, White sauces	de Wijk and Prinz, 2007
	10-20%	Artificial creams	Kilcast and Clegg, 2002
	0-50%	Model O/W emulsions	Mela, Langley and Martin, 1994
	2-20%	Model O/W emulsions	van Aken, Vingerhoeds and de Wijk, 2011
	5-40%	Model O/W emulsions	Vingerhoeds *et al.*, 2008
Hydrocolloid	0.03-1% low-methoxyl pectin and xanthan gum	Model O/W emulsions	Akhtar *et al.*, 2005
	10-25% Maltodextrin	Model O/W emulsions	Akhtar, Murray, and Dickinson, 2006

2.3. Satiety

The global obesogenic food environment and its implications on human health and wellbeing, has pressured food manufactures to contribute to the alleviation of the global obesity burden through "healthier" product reformulation. A pragmatic approach is to make numerous small changes to the food environment to promote a healthier diet (Chambers, McCrickerd and Yeomans, 2015). Enhancing the impact that foods have on satiety is one reasonable approach, as the satiating capacity of food largely dictates our food intake, by reducing sensations of hunger and increasing and prolonging fullness (Chambers, McCrickerd and Yeomans, 2015; Hetherington *et al.,* 2013). In recent years, this has been reflected in western market trends, with an increase in the sale of enhanced satiety food products (Bilman *et al.,* 2012).

2.3.1. Concept of satiety

The terms Satiety and Satiation form part of the appetite control system and are used to describe the processes inhibiting food intake. Satiation can be defined as within meal termination of eating as a result of inhibition of hunger and appetite during a meal. Satiety can be defined as inhibition of hunger and appetite in the post meal phase. So where satiation will govern how much is eaten at a meal, satiety will determine when the next meal will be consumed.

A conceptual framework that details the processes that influence satiety and satiation is proposed in the "Satiety Cascade" (Blundell, 1999). The original satiety cascade has since been modified to highlight the mediating psychological and physiological processes involved within each of the four phases of the cascade (See Fig. 2.7). The four phases are sensory, cognitive, post-ingestive and post-absorptive, with each phase proceeding the next, over time. As such, pinpointing the phase responsible for the satiating effect can be notoriously difficult due to the considerable overlap between phases in its development (Livingstone *et al.,* 2000).

Therefore, the main phase involved in generating satiety is often assumed based on the methodology used in the collection of satiety data.

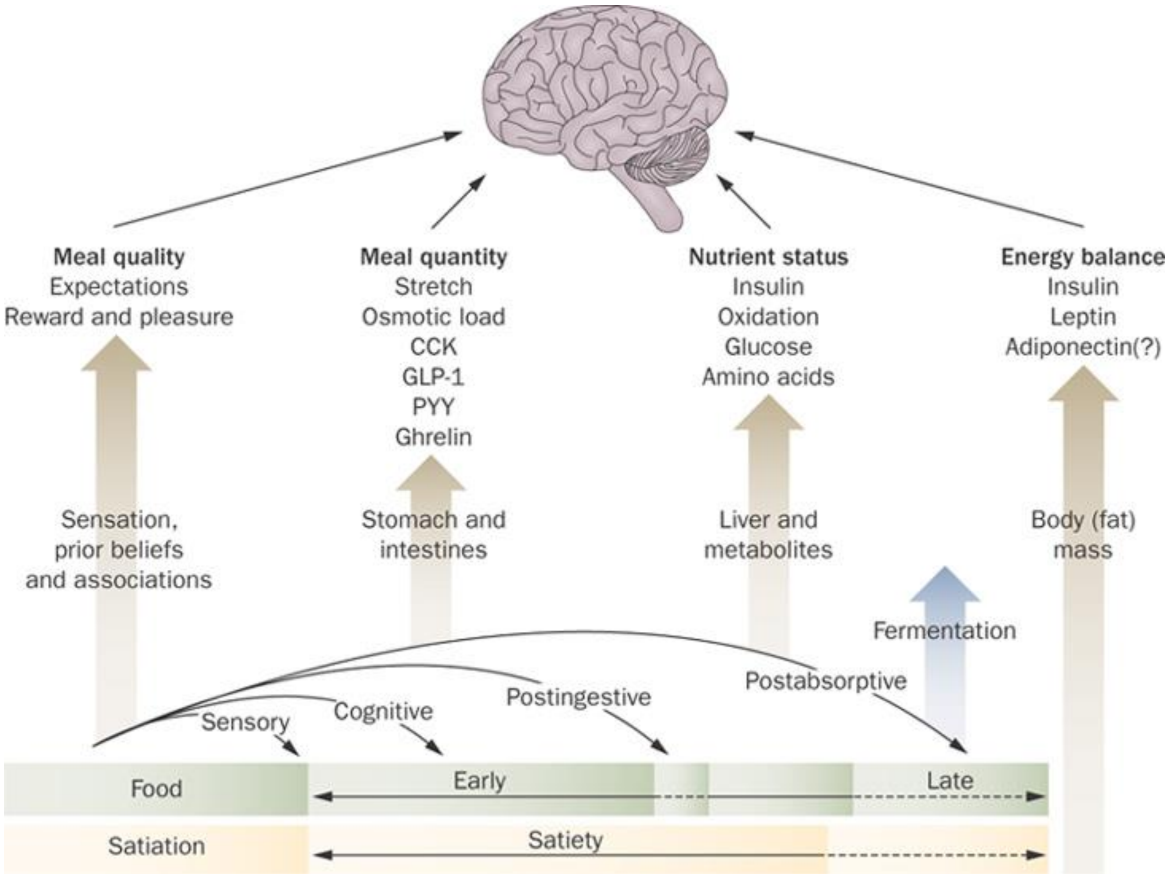


Fig. 2.7. Satiety cascade showing the relationship between satiation and satiety, and some mediating psychological and physiological processes (Blundell, 2010).

2.3.1.1. Sensory

The first phase of the satiety cascade, and one concerned largely with early or short-term satiety, is the foods sensory effects. The notion of sensory factors inducing satiation is known as "sensory-specific satiety". Sensory-specific satiety can be defined as the decrease in the appetite for a particular food as it is eaten in a meal, without a decrease in the appetite for different foods, as a result of the sensory characteristics of the food being ingested (Rolls, 2007). A common example of this would be the desire to consume a sweet desert after stating you are "full" from the consumption of a savoury main meal.

2.3.1.2. Cognitive

The second phase of the satiety cascade and largely involved in early and short-term satiety is the foods cognitive effects. Cognitive influences on satiety are based on a person's previous experiences or preconceptions of the food. Previous experiences create associations between the characteristics of the food and its later post-ingestive effects. Thus, being presented with a similar stimuli as experienced previously, leads the consumer to generate expectations on the foods satiety capacity, which influences food intake (Brunstrom, Shakeshaft and Scott-Samuel, 2008). Common food properties which are influential in cognitive driven satiety are factors such as sensory characteristics, portion size, mass, expected energy/macronutrient, messaging about the healthiness of the food and food consumption occasion type e.g. snack or meal (Brunstrom, Collingwood, and Rogers, 2010; Brunstrom, Shakeshaft and Scott-Samuel, 2008; Crum *et al.*, 2011; Hogenkamp *et al.*, 2011; McCrickerd, Lensing and Yeomans, 2015; Piqueras-Fiszman and Spence, 2012).

Of interest for this thesis, beyond early and short-term satiety, sensory characteristics through cognitive effects, have shown to be important in later satiety (de Graaf, 2012). With studies bypassing pre-ingestive phases, such as sensory factors, showing weaker satiety responses than studies considering sensory and cognitive influences (Cecil *et al*, 1998; Cecil, Francis and Read, 1998; Lavin *et al.*, 2002) (See section 2.3.2).

2.3.1.3. Post-ingestive

The third phase of the satiety cascade and predominantly associated with mid-term satiety, is the post-ingestive effects. Following a meal, various 'post-ingestive' neural and hormonal signals originating from the gastrointestinal tract inform the brain that food is being processed (See Fig. 2.7) (Bellisle, 2008). Such signals are typically afferent signals reflecting the food quantity and nutrient composition and current digestion processing state, for example the level of gastric distension or gastric emptying rate. Hormonal signals may also begin to reflect a satiated state with raised post-prandial concentrations and secretions of CCK, GLP-1 and PYY. Post-ingestive satiety is particularly influenced by factors such as the food macronutrient content, which exerts different ‘satiating potencies’ in the order of protein > carbohydrate > fat (Blundell and Macdiarmid, 1997), and the food physical state (i.e. liquid or solid) which has implications on physiological digestion transport mechanisms such as gastric emptying rate.

2.3.1.4. Post-absorptive

The final phase of the satiety cascade, which is associated with long-term satiety, is the post-absorptive stage. Post-absorptive satiety is associated with signals of food digestion, such as metabolite concentrations (insulin, glucose, amino acids, free fatty acids) in peripheral blood and oxidative metabolism of nutrients in liver and other metabolically active tissues (van Keelf *et al.*, 2012). Post-absorptive signals maintain satiety until the energy delivered by the previous meal has been utilised and/or stored. At the end of this phase, hunger signals return to acquire further energy intake, and the beginning of a new ingestive episode will start upon consumption.

Although there is considerable overlap between phases in satiety development (Livingstone *et al.*, 2000), the benefit of understanding satiety through such a conceptual framework, is that potentially each phase can be used as a target to engineer foods in order to maximise satiety. This thesis will concentrate on microstructural engineering of emulsion based food for pre-ingestive effects on satiety.

2.3.2. Early satiety

Even before the arrival of food in the gut, cognitive and sensory signals stimulated by cephalic digestion stimulators, such as the sight and smell of the food, as well as the products sensory properties during oral processing are influencing satiety. This is due to these early pre-ingestive satiety signals integrate with post-ingestive and post-absorptive signals to determine overall satiety capacity of a food, by influencing physiologically readiness for effective digestion, absorption and metabolism, through mechanisms such as endocrine response and gastric/intestinal secretions and motility. Cassady, Considine and Mattes (2012) demonstrated the importance of this relationship between pre-ingestive sensory and cognitive information and physiological satiety responses, as when participants believed a beverage preload would gel in their stomach causing a feeling of "Fullness", even though it did not, ratings of hunger were lower, gastric emptying was slower, insulin and GLP-1 release increased, ghrelin decreased and subsequent *ad libitum* food intake was lower. This was also reflected in the subjective comments made by participants after the consumption of the preloads. Additionally, studies designed to bypass pre-ingestive signals have demonstrated weaker satiety responses than studies also considering sensory and cognitive influences (Cecil *et al*, 1998; Cecil, Francis and Read, 1998; Lavin *et al.*, 2002).

An example of the communication between pre-ingestive signals and physiological readiness for digestion would be salivating at the thought, sight or smell of food (Wooley and Wooley, 1973). Further cephalic phase digestion responses such as gastric acid secretion (Feldman and Richardson, 1986) and insulin release (Rodin, 1985; Smeets, Erkner and de Graff, 2010) also occur upon thoughts about food. In the case of the foods sensory characteristics, upon the food entering the oral cavity, the food provides oro-sensory stimulation. This is the pre-ingestive signal which alters physiological readiness through activating vagus nerve signalling (Laughton and Powley, 1987; Sakata *et al.*, 2003), which initiates numerous digestion responses such as secretions of gastric acid, pancreatic and duodenal digestive enzymes, insulin, glucagon, PP and ghrelin, in addition to increased gastric motor activity (Smeets, Erkner and de Graff, 2010).

These responses clearly highlight the role of pre-ingestive stimuli on satiety. Although this thesis will focus on oro-sensory stimulation, both these examples (visual and oro-sensory) highlight a conditioned response to food-related stimuli. This conditioning is a result of associative learning between certain sensory characteristics and the subsequent sensation of satiety post-meal, which is collated and relayed back through the peripheral nervous system and brain centres and mediated further by longer term metabolic signals. This associative learning has been demonstrated by numerous studies in which increased satiety, reduced food intake and therefore earlier satiation were shown upon repeated exposure to a caloric initially novel food, in which the satiating behaviour achieved was a result of participants associating the initially novel sensory characteristics with ingestive effects post-consumption after the initial exposure (Birch and Deysher, 1985; Booth, Mather and Fuller, 1982; Booth and McAleavey, 1976; Hogenkamp *et al.,* 2012; Le Magnen, 1982; Yeomans *et al.*, 2009; Yeomans *et al.,* 2014; Yeomans, Weinberg and James, 2005). This evidence highlights the importance of pre-ingestive signals in subsequent satiety response through interaction with subsequent satiety mechanisms (anticipatory physiological regulation interactions).

To this conclusion, in oro-sensory stimulated satiety, the degree of vagal nerve stimulation will therefore depend on the cognitive associations with the food sensory profile. Textural attributes are particularly of note in oro-sensory stimulation (de Graaf, 2012) due to texture is one sensory characteristic that reliably predicts energy and nutrient content (Drewnowski, 1990). As such if a certain amount of energy is expected to be consumed physiological readiness for digestion will be adjusted accordingly.

2.3.3. Oro-sensory targets for pre-ingestive mediated satiety

Numerous studies have documented approaches in which satiety can be mediated through sensory properties (Davis and Smith, 2009; de Graaf, 2012; Dhillon *et al.*, 2015; Lavin *et al.*, 2002; McCrickerd *et al.*, 2012; McCrickerd, Chambers and Yeomans, 2014; Weijzen, Smeets, and de Graaf, 2009; Wijlens *et al.*, 2012; Zijlstra *et al.*, 2009). The benefit of this approach is clearly highlighted by studies demonstrating that bypassing oro-sensory stimulation through intra-gastric or intra-duodenal infusion, results in weaker satiety responses than studies considering oro-sensory influences (Cecil *et al*, 1998; Cecil, Francis and Read, 1998; Lavin *et al.*, 2002). The sensory experience is also reflective of "real-world" eating behaviour, in which food digestion begins with oral processing, and thus oro-sensory exposure.

Foods and beverages with a higher degree of structure, which require greater oral processing to be determined suitable for swallowing, have longer oral residence time, therefore longer oro-sensory exposure. As such greater sensory stimulation occurs, which increases satiety (Hogenkamp *et al.*, 2010). Zijlstra *et al.*, (2009) highlights this behaviour through the comparison of a chocolate flavoured milk-based liquid and a chocolate flavoured milk-based semi-solid product matched on palatability, energy density and macronutrient composition, in which fullness was significantly higher and subsequent intake significantly lower after consuming the semi-solid. Similar relationships have also been shown by other authors (Hogenkamp *et al.*, 2011). In a beverage context, greater viscosity has been shown to exert an independent inverse effect on hunger and subsequent intake, thus increasing satiety (Mattes and Rothacker, 2001; McCrickerd *et al.*, 2012; Perrigue *et al.*, 2010; Zhu, Hsu and Hollis, 2013).

Due to the correlations between a systems viscosity and perceptions of Thickness (See section 2.2.2.1), work such as this has highlighted that within beverages, Thickness is a satiety-relevant sensory cue. Of course, due to the multi-faceted nature of satiety, factors independent of just oro-sensory effects cannot be ignored, which has resulted in debate concerning the mechanism in which viscosity predominantly mediates satiety. For example, in such studies in which food is orally consumed, lower viscosity foods theoretically could reach the duodenum quicker during early rapid-emptying phase of digestion. As such different viscosities would elicit different responses that then influence satiety (Bateman, 1982). However, pre-ingestion viscosity is not always reflective of oral viscosity due to dilution from secretions, which has led some researchers to conclude that viscosity enhances satiety predominantly via oro-sensory rather than post-ingestive effects, and therefore Thickness, a sensory interpretation of viscosity, is the very definition of a satiety-relevant sensory cue (Hoad *et al.*, 2004).

One theory as to why Thickness is such a common satiety-relevant sensory cue is that more energy dense breast milk has higher viscosity (Picciano, 1998). As such from birth we learn to associate higher viscosity (Thicker) foods with greater energy and greater energy post-ingestive consequences (Davidson and Swithers, 2004). Supplementary to this, more viscous beverages generally tend to have a higher energy density, for example milk compared with water. Therefore, through every stage of our life time we have been conditioned to relate greater viscosity with greater energy content.

The sensory experience of low-viscosity high energy beverages may therefore not be reflective of the beverages actual energy content, which could lead to greater intake. Such a result was shown by Yeomans and Chambers (2011) in which high-energy (279 kcal) beverages were more satiating when they possessed sensory characteristics reflective of their energy content (Thick and Creamy) than sensory characteristics reflective of a less energy dense beverage (Thinner and less Creamy). Importantly this work highlights that enhancing the sensory characteristics of energy dense beverages to reflect its real energy content, may be a suitable route to increase the beverages satiating power.

Creaminess has also been shown to be modulated by viscosity and sensations of Thickness (see section 2.2.2.1), however only a number of recent studies have considered Creaminess in satiety (Bertenshaw, Lluch, Yeomans, 2012; Lett et al., 2016; McCrickerd *et al.*, 2012; McCrickerd, Chambers and Yeomans, 2014; Yeomans and Chambers, 2011). Yeomans and Chambers (2011) initially highlighted Creaminess potential as a Satiety-relevant sensory cue, as increasing Creaminess increased satiety of high energy beverages. Further findings demonstrated that matching carbohydrate preloads for perceived Thickness and Creaminess, to that of protein preloads resulted in very similar satiety responses. Protein is considered a more satiating macronutrient than carbohydrate; as such, this work highlights that sensory stimuli is critical in satiety and Creaminess can be an effective sensory attribute to increase the satiety impact of beverages (Bertenshaw, Lluch, Yeomans, 2012).

Within systems of a similar macronutrient composition, McCrickerd and co-authors (2012) showed that subtle increases in Thickness and Creaminess increased satiety expectations, by incrementally increasing tara gum concentration in fruit yoghurt drinks. Although the work focused on Creamy flavour rather than texture, due to the multi-influenced nature of Creaminess, it was concluded that even subtle increases Creaminess could increase satiety. The expectations were later shown to be reflected in actual eating behaviour (McCrickerd, Chambers and Yeomans, 2014). Lett *et al.*, (2015) showed similar results, however by taking a novel microstructural engineering approach, energy content and macronutrient composition were held constant. Through decreasing oil droplet size within O/W emulsions, Creaminess was shown to be significantly increased, which in turn significantly increased expectations of satiety and satiation. Lett, Norton and Yeomans (2016) later showed that these expectation were reflected in food intake behaviour, with significantly lower food intake after the consumption of an O/W emulsion containing smaller oil droplets.

Creaminess has also been shown to be a satiety-relevant sensory cue in real foods systems. A recent study by McCrickerd, Lensing and Yeomans (2015), explored the relationship between commercially available product's anticipated sensory characteristics and their expected impact on feelings of satiety and satiation. 40 widely available food and beverage products (varying in physical characteristics, packaging, serving size and total energy content) were evaluated. Creaminess positively correlated with total energy content, and products anticipated to be Creamier were expected to be the more filling and hunger suppressing. This conclusion shows similarity with Thickness, in which Thicker products are presumed to be more energy dense, a relationship also shown with Creaminess due to the association be perceived Creaminess and fat content (Chojnicka-Paszun, de Jongh, and de Kruif, 2012; de Wijk, Rasing, and Wilkinson, 2003; Frost and Janhoj, 2007; Richardson-Harman *et al.,* 2000).

Findings highlighting Thickness and Creaminess as oro-sensory targets for pre-ingestive mediated satiety in products with high energy density, certainly presents interesting opportunities in emulsion based systems, given emulsions are typically found within energy dense foods (see section 2.1.3). However a challenge in food reformulation for satiety is that often palatability decreases as satiety increases (Holt *et al.,* 1995). Conversely Creaminess as well as being shown to be a satiety-relevant sensory cue, has consistently shown to be strongly linked with product hedonics (Elmore *et al.,* 1999; Frost and Janhoj, 2007; Kilcast and Clegg, 2002; Richardson-Harman *et al.,* 2000). Therefore Creaminess could potentially increase satiety as well as increasing or maintain the foods hedonic properties, a result which was shown in Lett *et al.,* (2015) study.

2.3.4. Emulsion structuring approaches to satiety

With the growing realisation that designing low fat foods to achieve palatability and satiety may be unachievable, as lower fat foods are typically less palatable and low energy dense foods, which taste satiating, can promote appetite through rebound hunger mechanisms (Chambers, McCrickerd and Yeomans, 2015). Increasing the satiating functionality of the fat in fat based food to reduce intake, is a novel alternative (Himaya *et al.*, 1997). Due to the simple, yet dynamic, structure of emulsions, the opportunities to mediate satiety in emulsions are quite clear, given the myriad of physicochemical properties achievable through structural design and formulation approaches. As such, emulsions are being designed for functions within the human body. The design approach must account for the environmental conditions which will be experienced throughout the gastrointestinal tract to ensure the desired function is achieved *in vivo* (see Fig 2.8). For example, previous discussions highlighted emulsion structuring approaches to provide desired sensory characteristics under the physical and chemical environment of the oral cavity (see section 2.2.3). Despite the benefits and opportunities to mediate satiety via pre-ingestive routes (see section 2.3.2 and 2.3.3), thus far structuring emulsions to positively affect satiety has only concentrated on gastric and/or intestinal emulsion structuring to mediate satiety via post-ingestive and post-absorptive mechanisms. Approaches have considered two forms of gastric structuring for satiety, 1) intra-gastric layering, which involves structuring emulsions so gravitational separation (see section 2.1.2.1) occurs, so the lipid layer sits at a desired position within the stomach, and 2) controlling lipolysis, to control the rate of lipid digestion and uptake to trigger post-ingestive satiety signals (see Fig. 2.7). Within the intestine, the "ileal brake" mechanism has been an extensively proposed and investigated target, due to the potent effect of fat detection at the ileum (distal large intestine) on satiety (Maljaars *et al.*, 2008).

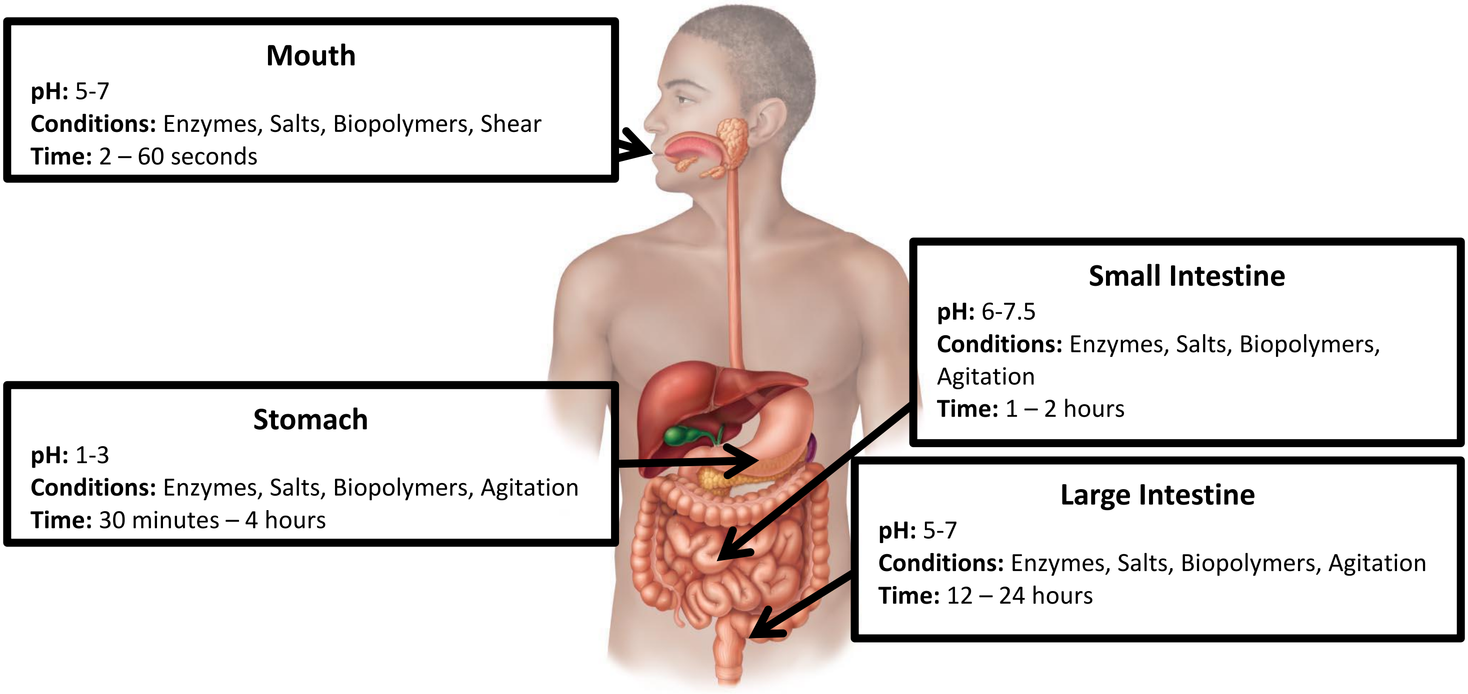


Fig. 2.8. Schematic diagram of the physiochemical environment at each stage of the human gastrointestinal tract.

2.3.4.1. Intra-gastric layering

Intra-gastric layering as an approach to enhancing satiety uses the thermodynamically unstable nature of emulsions to its advantage, by designing emulsions which will phase separate under gastric conditions. As such, gravitational separation can be utilised to ensure that the lipid layer sits at a desired position within the stomach to manipulate physiological responses for appetite related outcomes.

A large body of work by Marciani and co-workers (2006, 2007, 2009) showed that under the pH conditions of the stomach (see Fig. 2.8), acid-unstable O/W emulsions phase separate, leading to a lipid layer on top of the aqueous gastric contents. As a consequence of such a layered structure, the energy dense lipid layer is situated away from the pylorus (stomach exit valve), delaying lipid entrance into the small intestine resulting in relatively fast gastric emptying, reduced gastric distention and slower onset of satiation to that of acid-stable emulsions. Differences in the physiological effects related to satiety, between acid stable and unstable emulsions, are due to the quicker entrance of fatty acids into the duodenum (proximal small intestine), resulting in greater CCK secretion, slowing gastric emptying by suppressing the muscular contractions of the antrum and enhancing the contractions of the pylorus (Golding *et al.,* 2011; Keogh *et al.,* 2011). Similar results to Marciani and co-workers (2006, 2007, 2009) were shown by Foltz *et al.,* (2009) who instead of structuring the emulsion to be acid-unstable, positioned a lipid layer on top of the aqueous gastric contents via a naso-gastric tube.

The stability of an emulsion under gastric conditions is largely dependent on the emulsifier used to stabilise the water and oil interface, Marciani's work, for example, used the surfactants Tween 60 (acid-stable) and Span 80 (acid-unstable) to produce to two O/W emulsion types whose lipid component either remained as dispersed individual emulsion droplets (stable) or creamed (unstable). van Aken and co-workers (2011) have carried out similar work investigating protein-stabilised O/W emulsions, but under *in vitro* gastric conditions. As within Marciani's work the reference surfactant (Tween 80) remained stable with slight coalescence, WPI and NaCas stabilised emulsions, however, both creamed, with droplets flocculating before coalescence occurred. Full fat milk also underwent the same conditions; flocculation occurred, but interestingly arrested stirring resulted in sedimentation of the fat layer. Although this hasn't been studied *in vivo* the possible physiological implication of this finding is that the lipid layer would enter the small intestine before the remaining aqueous gastric contents. As such, a significantly quicker onset and extension of satiation would be expected.

In all these studies discussed, oil droplet size has been kept constant across test samples. Although predominantly controlled by emulsifier type, the effect of oil droplet size on intra-gastric layering and its rate on formation is yet to be shown. However, there is evidence that oil emulsion droplet size effects satiety, however this is through modulation of lipolysis rate.

2.3.4.2. Lipolysis

Lipolysis, the hydrolysis of triglyceride lipids by lipase, produces the absorbable lipid constitutes fatty acids and monoglycerides. As triglycerides are insoluble in an aqueous medium and lipases are water-soluble, lipolysis occurs at the oil/water interface, with the lipid in the form of an emulsion. As such, lipid surface area, and properties of the oil/water interface, can affect the rate of lipolysis, which can have an impact on fatty acid sensing, and therefore physiological mechanisms related to satiety (Armand *et al.*, 1999).

Although emulsion surface area is the key physicochemical parameter affecting the rate of lipolysis, the interfacial properties of the oil/water interface are also important, as the nature of the droplet interface will determine the stability behaviour of emulsion droplets upon exposure to the environment of the stomach (Golding *et al.*, 2011; Singh, Ye and Horne, 2009). *In vitro* studies have shown that in the absence of bile salts, interfaces composed of biopolymers, phospholipids or small molecule surfactants (e.g. polysorbates), are resistant to lipolysis due to the interface not possessing a significant number of areas for lipase access (Wickham *et al.*, 1998). However, under *in vivo* conditions, co-lipase and bile salts are present, which aid in the displacement of molecules from the interface, increasing lipolysis rate (Singh, Ye and Horne, 2009). As such, it has been concluded in cases where bile salt absorption to the interface can be inhibited, lipolysis can be inhibited (Vinarov et al., 2012).

One example of a substance effective in such retardation of bile salt action is polysaccharides. Polysaccharide presence within emulsion systems has been shown to bind bile salts, reducing bile salt availability for lipolysis (Dongowski, 1997; Koseki *et al.*, 1987), and also absorb to the emulsion interface forming a multi-layer interface, increasing interfacial thickness, and thus reducing lipase access to lipid (Ogawa, Decker, McClements, 2003).

Lipid surface area in an emulsion is governed by the oil droplet size. Armand and co-workers (1999), intragastrically gave subjects either a coarse (10 µm) or a fine (0.7 µm) O/W emulsion. Results showed that although droplet size increased in the stomach, the rate of lipolysis was greater for the fine emulsion. The authors, observed that gastric emptying rate was reduced, a result of greater surface area for lipolysis, providing greater and quicker release of fatty acids initiating numerous satiety mechanisms. Ledeboer and co-workers (1999), showed similar results, but provided evidence that reducing emulsion droplet size significantly increased secretion of CCK. Other studies have also shown that smaller oil droplets increase lipolysis, due to the greater interfacial area available for digestive lipase binding and its positive implications on satiety, satiety physiological mechanisms and/or satiety biomarkers both; *in vitro* (Armand *et al.*, 1992; Peters *et al.*, 2014) and *in vivo* (Borel *et al*, 1994; Maljaars *et al.*, 2012; Peters *et al.*, 2014; Seimon *et al.*, 2009). As shown by Armand and co-workers (1999) and Borel and co-workers (2001), It is important to note, that differences in satiety physiological mechanisms and biomarkers are not a result of a greater concentration or quicker uptake of fatty acids in the bloodstream as a result of droplet size, as plasma triglyceride area-under-the-curve values between coarse and fine emulsions have showed no difference. This indicates that the differences in satiety physiological mechanisms and biomarkers are a result of fatty acid sensing by the small intestinal mucosa, leading to secretion of CCK and PYY and/or direct activation of vagal neural afferent signals (Maljaars, Peters and Mascalee, 2007).

2.4. Summary of literature review

The literature review has been presented in the context of the two main areas of research covered in this thesis:

- Inter-relation between structure, physical properties and sensory outcomes.
- Inter-relation between structure, sensory outcomes and food intake behaviour.

Given the extensive review of the available literature relevant to this thesis, there is a clear ability to exploit emulsion microstructural approaches, in engineering, desired sensory characteristics or satiety outcomes; however, emulsion microstructural approaches are yet to consider combing both. Inevitably, with the growing consumer trend of satiety enhanced products, maintenance of this growth requires innovative approaches to produce satiating yet desirable foods.

As modulating emulsion microstructure can change sensory characteristics and sensory characteristics change satiety via pre-ingestive satiety signals, a clear opportunity is presented to engineer microstructure for satiety via a pre-ingestive approach, especially if attributes, such as Creaminess, which are satiety-relevant sensory cues and have strong hedonic appeal, can be manipulated. Additionally, in the interest of understanding for future development, it is important to understand the physical properties (viscosity and friction coefficient) which construct such sensory attributes.

Chapter 3. Influence of oil droplet size on the tribological properties of oil-in-water emulsion systems and the relation to sensory perception

3.1. Abstract

A fundamental component of an emulsion's microstructure is its droplet size, which can affect sensory perception. Previous research has only considered a relatively narrow range of droplet sizes. This paper examines oil droplet sizes from 0.2 - 50 µm ($d_{4,3}$) and the effect on surface (tribological) properties and the perception of sensory attributes, discussing the relationship between this physical property and sensory perception. Bulk (rheological) properties were also considered but samples were perceptually iso-viscous. Increasing oil droplet size resulted in a negative linear relationship with friction coefficient. Distinct lubrication mechanisms as a function of droplet size were reported. Oil droplet size affected ratings of sensory attributes, especially Smoothness, Slipperiness and Creaminess. Strong correlations between sensory attributes and friction coefficient were shown, such as Oiliness and Slipperiness perception. However, correlation strength between friction coefficient and sensory perception differed dependent on the droplet size range considered highlighting why previously investigated droplet size ranges resulted in only minor differences in the perception of sensory attributes. This paper has further realised the value of tribological techniques in instrumentally understanding emulsion microstructure in relation to sensory perception, and highlights the physical properties involved in the perception of textural and mouthfeel attributes.

3.2. Introduction

Emulsions are common food structures. The nature of emulsions offers the opportunity to alter the physical characteristics of the system for desired functionality. Sensory properties, such as creaminess, are of great interest as they influence hedonic appeal, which crucially affects a consumer's repeat purchase habits. Despite the complexity of food oral processing, perceptions of texture-related sensory attributes appear to correlate with instrumental measures, such as bulk rheology and thin-film rheology (tribology) (Chen & Stokes, 2012).

3.2.1. Textural interactions and the perception of creaminess

Creaminess is considered a multi-influenced sensory attribute (Chen & Eaton, 2012), and the multi-faceted nature of the processes underlying its perception make it hard to define. To the consumer creaminess has been a term used to describe the appearance, flavour and/or texture of a food product, and is commonly used to describe products containing fat (Mela, 1988; Chen & Eaton, 2012). It is also a term that has a stronger influence on hedonic response than many other sensory attributes (Kilcast & Clegg, 2002). Textural attributes are considered to be predominant within the perception of creaminess. Early work investigating liquid systems showed that creaminess perception is largely dependent on the perception of both thickness and smoothness (Kokini, Kadane & Cussler, 1977). As a result of these findings, Cussler *et al.,* (1979) extended investigations using a range of foods with varying degrees of creaminess. Results showed that the sensory attributes thickness and smoothness were independent, and were insufficient to predict creaminess perception alone. Consequently, the authors concluded that creaminess might be a combination of both smoothness and thickness perception. Within a study predicting the texture of liquid and melting semi-solid foods, Kokini and Cussler (1983) showed that a prediction of creaminess perception could be obtained with regression analysis of data from scores of thickness and smoothness (see equation 3.1).

$$Creaminess = Thickness^{0.54} X\ Smoothness^{0.84}$$

Eq. 3.1

Within a similar study using liquid and semi-solid food, slipperiness was also considered a component of creaminess (Kokini *et al.*, 1984). Another equation to predict creaminess was generated, with regression analysis of data from scores of thickness, softness (smoothness) and slipperiness (see Equation 3.2).

$$\log Creamy = 0.52 \log Thick + 1.56 \log Soft - 0.32 \log Slippery$$

Eq. 3.2

Results of these studies highlight that multiple attributes (predominantly textural), that are independent of one another, are involved in creaminess perception (Kokini, 1987). It would be useful to be able to predict creaminess from instrumental measures, decreasing product development time by reducing the need for labour and time intensive sensory testing. This approach, however, is likely to be challenging given the complexity and variability of the sensory experience.

3.2.2. Rheological and tribological correlations with sensory perception

Instrumentally understanding a food's rheological behaviour can provide insight into flow behaviour during processing in the mouth. The food's state at a given level or range of shear deformation can be related to textural perception. We can, therefore, assume that if alterations in microstructure affect flow properties, oral processing behaviour and sensorial properties will alter.

The relationship between viscosity and oral assessment of thickness has been extensively investigated for fluid systems. Wood (1968) showed that the highest level of correlation between thickness perception and viscosity was at a shear rate of 50 s^{-1}. As such, 50 s^{-1} has been extensively used as a convenient value to represent oral processing shear rate of fluid foods. Subsequent investigations demonstrate that oral processing exerts a range of deformation and shear upon a food, and correlations between thickness and viscosity at a given shear rate differ dependent on the food's rheology (Cutler, Morris & Taylor, 1983; Kokini, Kadane & Cussler, 1977; Morris & Taylor, 1982; Morris, Richardson & Taylor, 1984; Shama & Sherman, 1973a; Shama & Sherman, 1973b), with strong correlations having been demonstrated at shear rates between 10-1000 s^{-1} (Shama & Sherman, 1973a, 1973b).

However, it is apparent that bulk rheology does not entirely describe the textural properties of foods. This is highlighted by the improvement in understanding of attributes, such as smoothness, fattiness, stickiness and creaminess (de Wijk & Prinz, 2005), when surface related properties are considered. During oral processing of fluid systems the compression of the tongue and palate against one another generates a thin fluid film. The evolution and presence of thin films within the oral cavity, in addition to the post-deglutition oral mucosa, is involved in sensory perception and can be related to surface properties. The influence of surface related properties such as lubrication has long been recognised as an important factor in sensory perception (Hutchings & Lillford, 1988; Kokini, Kadane & Cussler, 1977). Yet, the nature in which bulk viscosity is traditionally measured does not account for surface related properties. Therefore, additional approaches which do measure surface properties and provide insight into material properties that bulk rheology measures cannot deduce, are required, such as the use of tribology (thin-film rheology) (Malone, Appelqvist & Norton, 2003). Tribology is the science of adhesion, friction and lubrication. Correlations between tribological measures of lubrication behaviour and oral perception is an area of growing interest (Chen & Stokes, 2012).

Kokini's work (1977, 1983, 1987) is considered to be pioneering in food tribology research and led to the design of Kokini's model of oral lubrication (Kokini, 1987). This model quantified physical measures of friction and viscosity in relation to the perception of sensory attributes smoothness and slipperiness. "Smoothness" correlated with the reciprocal of the friction force between oral surfaces and "Slipperiness" with the reciprocal of the sum of the viscous force and the friction force. Within emulsion systems, tribology has been considered in relation to the perception of smoothness and slipperiness, in addition to other textural attributes (Bellamy *et al.,* 2009; Chojnicka-Paszun, de Jongh & de Kruif, 2012; de Hoog *et al.,* 2006; de Wijk & Prinz, 2005; Dresselhuis *et al.,* 2007; Dresselhuis *et al.,* 2008b; Malone, Appelqvist & Norton, 2003). Malone, Appelqvist and Norton (2003) related friction coefficients at different entrainment speeds to sensory perception of slipperiness. It was concluded that oral speeds during sensory assessments were most probably between 10 and 100 mm/s. Therefore, the median value within this range can act as a representative and convenient value of the entrainments speeds experienced during oral processing. However, friction coefficient alone is not sufficient to completely predict textural attributes. As such, there is a need to consider both bulk and surface properties.

3.2.3 Emulsion microstructure and composition in relation to sensory perception

Investigations have shown that oil droplet size within oil-in-water emulsions does not (or only minimally) affect sensory perception (Akhtar *et al.,* 2005; Vingerhoeds *et al.,* 2008). Kilcast and Clegg (2002), however, did show that creamy texture and thickness perception both increased with increasing droplet size, attributing these differences to the fact that larger droplet sizes increased viscosity. However, until now only a relatively narrow oil droplet size range has been investigated (0.5-2 µm, Akhtar *et al.,* 2005; 3-9 µm, Kilcast & Clegg, 2002; 0.5-6 µm, Vingerhoeds *et al.,* 2008), which limits the conclusions that can be drawn from such work.

Some studies have investigated the effect of oil droplet size on rheology (Akhtar *et al.,* 2005; Akhtar, Murray & Dickinson, 2006; Kilcast & Clegg, 2002) or tribology (de Wijk & Prinz, 2005) in relation to sensory attributes. However, studies are yet to consider a broad range of oil droplet sizes and the effects on physical properties and sensory perception. Therefore, the present study investigates the effect of oil-in-water emulsion droplet size on rheological and tribological behaviour and sensory perception, and how these measures were inter-related, using a very wide range of oil droplet sizes.

3.3. Materials and methodology

3.3.1. Emulsion materials and preparation

1 wt.% sodium caseinate (NaCas) (Excellion EM7, DMV International, The Netherlands), 2 wt.% sucrose (Silverspoon granulated, British Sugar Plc, UK) and 15 wt.% sunflower oil (Tesco Plc, UK) and distilled water were used to produce the model oil-in-water emulsion systems.

Emulsions were produced using two different methods dependent upon the required mean droplet size of the emulsion being produced: a high shear mixer (Silverson L5M, Silverson machines Ltd, UK) or a high-pressure homogeniser (GEA Niro Soavi Panda Plus 2000, GEA Niro Soavi, Italy). In a 600 ml beaker, 15 wt.% sunflower oil was added to 85 wt.% aqueous phase (1.1 wt.% NaCas, 2.2 wt.% sucrose, 96.6 wt.% distilled water solution). The whole sample was then emulsified for 5 minutes using the high shear mixer. Dependent on oil droplet size being produced the sample was subjected to a different rotational speed (rpm) and emulsor screen (fine (0.8 mm pores) or medium (1.6 mm pores)) (**50 µm**: *2500 rpm medium screen,* **40 µm:** *3500 rpm medium screen,* **20 µm**: *5000 rpm fine screen* and **12 µm:** *9000 rpm fine screen*). For emulsions produced using the high-pressure homogeniser, first a pre-emulsion was produced using the high shear mixer at 9000 rpm with a fine emulsor screen for 5 minutes. The pre-emulsion was then subjected to homogenisation, differing in pressure and number of passes (**6 µm**: *20 Bar 3 passes*, **2 µm:** *100 Bar 2 passes* and **0.2 µm:** *1250 Bar 4 passes*). This model system was used, as in preliminary work to select a model system, NaCas provided the widest droplet size range and most stable droplets at concentrations safe for human consumption, in comparison to WPI and Tween 80, and 15% oil volume fraction, as this provided the widest droplet size range whilst still being palatable, in comparison to 5, 10, 20, 30 ,40 and 50%. This model emulsion system, subject to slight variation, was used throughout this thesis. All samples were produced in 400 g batches, under clean and hygienic conditions on the day of evaluation, emulsion droplet size was stable when tested or assessed, and all samples were stored under refrigerated conditions at 2-5 ^{o}C.

3.3.2. Droplet size characterisation

Oil droplet size distributions were measured by dynamic laser light scattering (Malvern Mastersizer MS2000, Malvern Instruments, UK). All measurements were performed in triplicate and conducted on the day of emulsion manufacture. When oil droplet size was measured post tribometer experiment to assess potential break-up behaviour, a sample was taken following the measurement via pipette from the bulk and contact zone and droplet size was measured immediately.

3.3.3. Rheological measurements

A rheometer (Kinexus Pro, Malvern Instruments, Malvern, UK) was used to perform bulk rheology measurements. Measurements were taken with a stainless steel double gap geometry. For all experiments, the sample was poured into the rheometer cup, and allowed to thermally equilibrate prior to the measurement. Temperature was fixed at 37 °C to mimic oral temperature. A shear table was used to measure viscosity between shear rates of 0.1 and 200 s^{-1}. Measurements were performed in triplicate and a mean value derived.

3.3.4. Tribological measurements

A ball-on-disc tribometer (MTM, PCS Instruments, UK) was used to obtain tribology data. Measurements were taken using tribopair surfaces of a polished ¾ inch steel ball loaded onto to a silicone elastomer disk (diameter: 46 mm, thickness: 4 mm). For all tests, both contact surfaces were sonically cleaned (Branson 2210 Ultrasonic cleaner, Branson Ultrasonics Corporation, Danbury, USA) in ethanol for 5 minutes followed by distilled water for 5 minutes and then air dried. A new elastomer disk was used for each experiment. 40 ml of sample was used and temperature was fixed at 37 °C, simulating oral processing temperature. 3 applied forces were investigated: 0.5 N, 1 N and 3 N.

Friction coefficient (μ) is a dimensionless parameter (Hamrock, 1994) used as a representation of the samples' lubrication capacity, which can be calculated using Equation 3.3:

$$\mu = \sum \frac{F\mathrm{t}}{W}$$

Eq. 3.3

where F_t is the point of tangential friction force assessment and W is the ball load. μ was measured across 20 averaged entrainment speeds (U) (see Equation 3.4) logarithmically set between 1 to 1000 mm/s, using a slide-roll ratio (SRR) of 50% to mimic the complex sliding and rolling friction associated with oral processing conditions.

$$U = \sum \frac{U\text{disc} - U\text{ ball}}{\text{SRR}}$$

Eq. 3.4

Stribeck curves (μ versus *U*) were obtained in 6 consecutive sweeps, alternating between ascending and descending, in order to map samples under hydrodynamic, mixed and boundary lubrication regimes. For each sample, experiments were performed in triplicate, and Stribeck curves were generated from the mean of the 18 sweeps.

The deformation of an oil droplet within the emulsion samples while present in a thin-film under the tribological regime was calculated. Deformation was calculated at 44.7 *U*. 44.7 *U* was chosen as Malone, Appelqvest and Norton (2003) showed some limited sensory data for guar gum thickened solutions, indicating a significant level of correlation between lubrication properties and oral slipperiness perception at entrainment speeds between 10 and 100 mm/s. Although contributory work to this finding is limited, this work suggested that this entrainment speed range is most relevant to describe oral processing processes associated with slippery mouthfeel, an attribute regarded as significantly influenced by friction (Kokini, 1987), thus the median value between 10 and 100 mm/s (45 mm/s) was chosen for investigation. 44.7 *U* will be referred to as 45 *U* for the rest of the chapter.

A droplet's deformation parameter (Erni, Fischer & Windhab, 2005) was calculated using Equation 3.5, to identify droplet behaviour within the contact zone. The greater the deformation parameter value, the greater the degree of droplet deformation to an ellipsoidal shape.

$$\mathrm{D} = \frac{19\lambda_\mu + 16}{16\lambda_\mu + 16} Ca$$

Eq. 3.5

where *Ca* is the capillary number calculated using Equation 3.6 (Taylor, 1934) and λ_μ is the viscosity ratio of the continuous (μ_c) over the dispersed (μ_d) phase. Viscosity values were taken at 50 s^{-1} at 37 ^{o}C (average human body temperature).

$$Ca = \frac{\tau_{max} . R}{\sigma}$$

Eq. 3.6

where σ is the interfacial tension, *R* is the droplet radius in the absence of flow and τ_{max} is the shear stress experienced within the contact zone calculated using Equation 3.7.

$$\tau_{max} = \frac{W + F_F}{3A_r}$$

Eq. 3.7

where F_F is the frictional force at 45 *U* at a specific ball load and *W* is the normal force exerted by the ball, over the contact area (A_r). A_r was a constant for all samples at a given applied force (0.5 N: 9.57 x 10^{-6}, 1 N: 1.52 x 10^{-5}, 3 N: 3.16 x 10^{-5}). A_r was calculated using Equation 3.8.

$$A_r = \pi \left[\frac{3WR}{4E^*}\right]^{\frac{2}{3}}$$

Eq. 3.8

where W is the normal load applied by the ball, R is the radius of the stainless steel ball applied and $E*$ is the composite young's modulus calculated using Equation 3.9.

$$\frac{1}{E^*} = \frac{1 - \nu_{ball}^2}{E_{ball}} + \frac{1 - \nu_{disc}^2}{E_{disc}}$$

Eq. 3.9

where ν is the Poisson's ratio for the ball/disk, and E is the elastic modulus of ball/disk. Values for Poisson's ratio and elastic modulus for the ball, are that of stainless steel, and taken as E_{ball}= 216GPa, and ν_{ball}=0.3 (Hougardy *et al.*, 2012). Values for Poisson's ratio and elastic modulus for the disk, are that of silicone elastomer, and taken as E_{disc} = 0.5 MPa (Garrec and Norton, 2013) and a Poisson's ratio, ν_{disc}=0.5 (Ilg *et al*., 2012).

3.3.5. Interfacial tension measurements

Interfacial tension between the oil and the continuous phase was measured in triplicate using a Tensiometer K100 (Kruss, Hamburg, Germany) with the Wilhelmy plate method. The experiment was conducted over a time of 4,000 seconds with a surface detection speed of 15 mm/min. Data used within capillary number calculations (see Equation 3.6) was collected at 3,500 seconds (plateau of curve).

3.3.6. Sensory analysis

Sensory sessions were scheduled between 10 a.m. and 12 a.m. or 2 p.m. and 4 p.m., Monday to Friday, with sessions lasting approximately 1 hour. Participants were instructed to arrive having refrained from consuming any food or beverages, other than water, for 2 hours before their arrival. Subjects were seated in individual booths and were presented with 40 ml of sample in 60 ml pots coded with random 3-digit codes. Each random 3-digit code corresponded to a specific sample, which was only known by the investigators. All samples were served between 5-7 ^{o}C. Sample order was randomised. Sensory assessment was carried out using visual analogue scale (VAS) questions. Six 100 mm randomised VAS's representing each sensory attribute (see Table 3.1) were presented headed with the question "How [sensory attribute] is sample [sample code]?" anchored with opposing statements left-to-right e.g. "Not at all [target attribute]" (scored as zero) and "Extremely [target attribute]" (scored as 100). Some questions differed slightly, so they were grammatically correct. Participants were briefed with the description of each attribute pre-test (see Table 3.1 for details). A spittoon was provided and subjects were instructed to spit out the sample after their assessment had been made. Still bottled spring water and dry cream crackers (Aldi, Germany) were provided, and subjects were instructed to use these to refresh their palate between samples. Participants were given the opportunity to ask questions to clarify the sensory protocol before testing began.

The sensory properties of the emulsions were assessed by 24 untrained healthy non-smoking male assessors aged between 18-26 with a mean BMI of 22.8 ± 1.7 Kg/M^2. All participants gave written informed consent prior to participation. As data was not available to conduct a power analysis to determine appropriate sample size, sample size was set larger than similar studies investigating emulsions in sensory perception (Akhtar *et al.*, 2005 (n = 14); Akhtar, Murray and Dickinson, 2006 (n = 12); de Wijk and Prinz, 2005 (n = 8); Vingerhoeds *et al.*, 2008 (n = 8)) to try and account for appropriate power. To ensure sample characteristics were consistent throughout the thesis, females were excluded. This is because in chapters investigating satiety, females were excluded as they typically practice significantly higher levels of restricted eating, along with significant higher levels of other eating behaviours than males (Wardle, 1987). Ethical approval for the study was obtained from the University of Birmingham ethics committee.

Table 3.1 Sensory attributes used during sensory analysis, with description given to participants.

Attribute category	Sensory attribute	Description reference
Mouthfeel	Smoothness	Degree of absence of any particles, lumps, bumps etc within the sample
	Thickness	Viscous consistency within the mouth; *Water to yoghurt*
	Slipperiness	Degree to which the product slides over the tongue
	Creamy mouthfeel	Soft, smooth with flowing consistency; *Water to full fat cream*
Overall	Creaminess	Assessment of overall creaminess of the sample
	Oiliness	Assessment of overall oiliness of the sample

3.3.7. Data analysis

To assess and understand the strength and direction of correlation between droplet size, tribology and sensory ratings Pearson correlation (r) was performed, and to assess and understand the proportion of variation in the correlation, coefficient of determination (R^2) was performed. Correlations are linear unless otherwise stated. The strength of the correlation was characterised in accordance with Evans’s (1996) guide, so that 0 -19.9 = no correlation, 20 – 39.9 = weak correlation, 40 – 59.9 = moderate correlation, 60 – 79.9 = strong correlation and 80 -100 = very strong correlation. Data analysis was carried out using SPSS.

3.4. Results and discussions

3.4.1. Emulsion droplet size distribution

Eight different oil-in-water emulsions differing in oil droplet size were produced ($d_{4,3}$ µm mean ± standard deviation of triplicate measures accounting for emulsion production error): 0.16 ± 0.01, 1.84 ± 0.09, 6.22 ± 0.82, 12.41 ± 0.11, 20.20 ± 0.42, 37.01 ± 1 and 48.24 ± 0.82 (see Fig. 3.1). For the rest of this chapter samples will be referred to as 0.2, 2, 6, 12, 20, 40 and 50 µm, respectively. Span values, highlighted that with the exception of the 2 µm emulsion, droplet size uniformity within samples was largely comparable (**50 µm**: 1.40, **40 µm:** 1.33, **20 µm**: 1.31, **12 µm:** 1.68, **6 µm**: 1.93, **2 µm:** 2.59 and **0.2 µm:** 1.84), despite processing being via either high shear mixing (50, 40, 20 and 12 µm) or homogenisation (6, 2 and 0.2 µm). It seems sensible to suggest the larger span for the 2 µm emulsion sample could be attributed to fewer homogenisation passes, in comparison to emulsion samples containing 6 and 0.2 µm droplets.

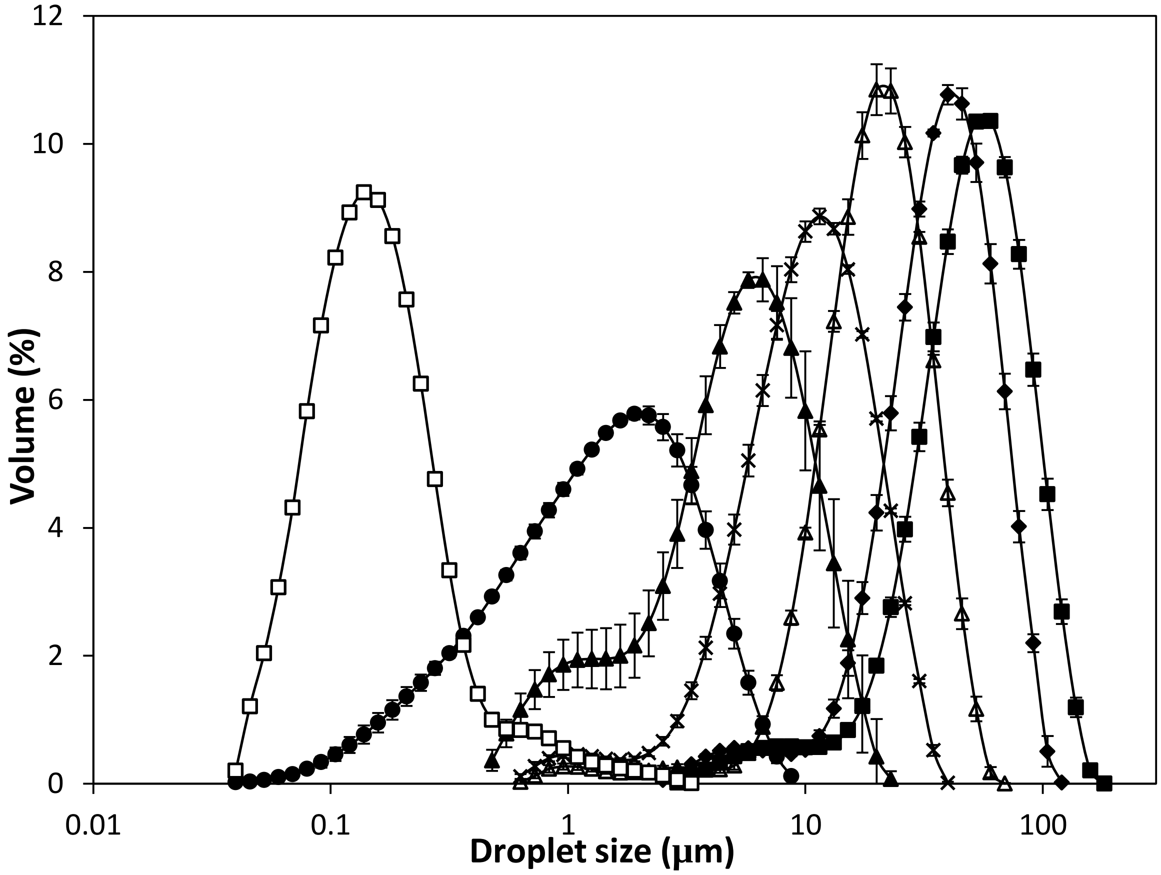


Fig. 3.1 Droplet size distributions of 1 wt.% NaCas, 15 wt.% sunflower oil-in-water emulsions: 0.2 µm (□), 2 µm (●), 6 µm (▲), 12 µm (X), 20 µm (△), 40 µm (◆) and 50 µm (■) . Error bars representative of one standard deviation.

3.4.2. Emulsion rheology

Rheological behaviour as a function of droplet size is shown in Figure 3.2.

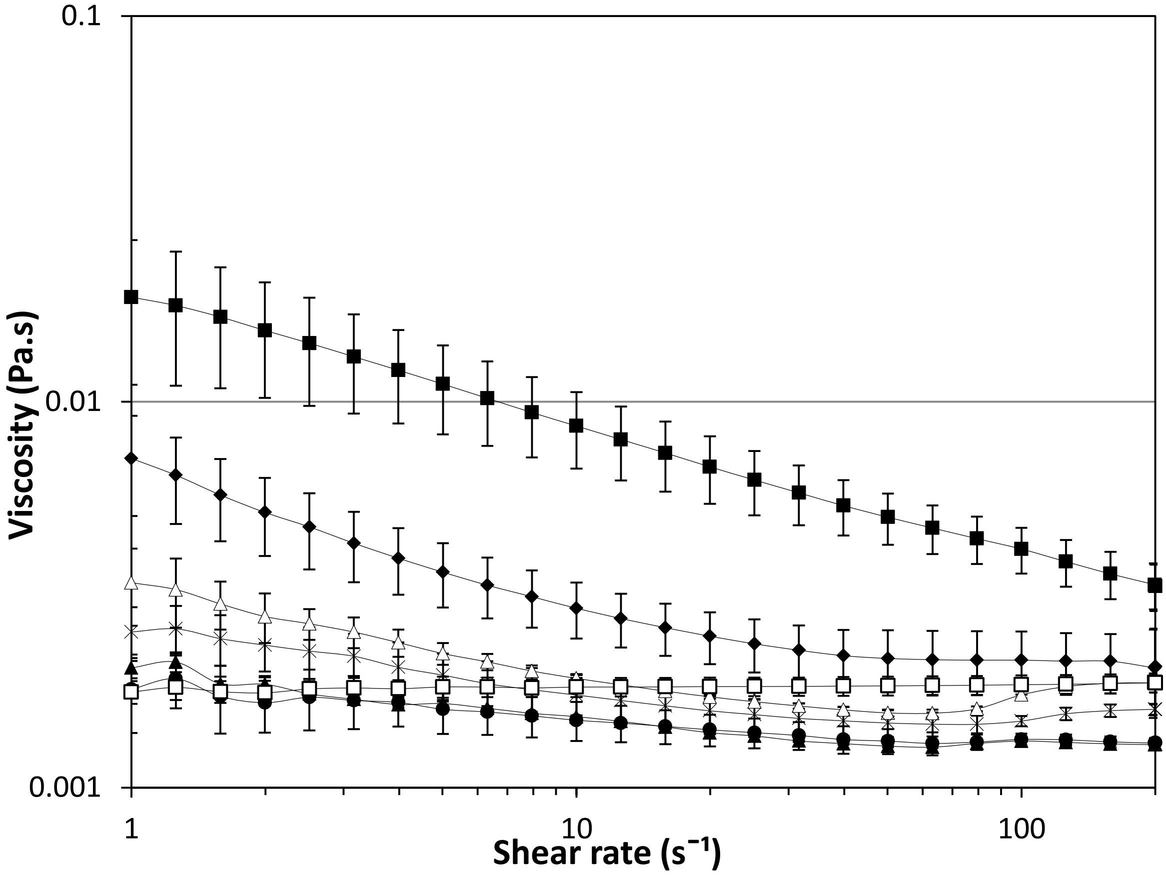


Fig. 3.2 Viscosity profile of 1 wt.% NaCas, 15 wt.% sunflower oil-in-water emulsions varying in average oil droplet size: 0.2 µm (□), 2 µm (●), 6 µm (▲), 12 µm (X), 20 µm (△), 40 µm (◆) and 50 µm (■). Error bars representative of one standard deviation.

The rheological behaviour of the emulsions was dependent on its droplet size (see Fig. 3.2). Flow curves demonstrate shear thinning behaviour for all emulsions except the emulsion with a droplet size of 0.2 µm, with flow curves becoming increasingly Newtonian with decreasing droplet size. This is behaviour is further highlighted by power law derived values (0.2 µm: 0.008, 2 µm: -0.068, 6 µm: -0.088, 12 µm: -0.102, 20 µm: -0.118, 40 µm: -0.223 and 50 µm: -0.333). It seems sensible to suggest the reason we see an increase in shear thinning behaviour with increasing droplet size is due to depletion flocculation. This is because with increasing droplet size, total dispersed phase surface area is reduce increasing the quantity of excess non-absorbed entities within the continuous phase i.e. NaCas emulsifier, which would promote attractive interaction between droplets and thus floc formation (Dickinson, 1989; Dickinson, 2010). Flocs increase the viscosity of a system and upon the application of increasing shear, these flocs break up, as the droplet-droplet interaction is only weak, producing the pronounced shear thinning behaviour observed (Somasundaran, 2006).

The emulsion with 50 µm droplets are shown to be the only sample which is distinguishably different in viscosity from any other droplet sizes at all shear rates. However, although the percentage difference in viscosity between extreme samples (2 and 50 µm) may seem large at all shear rates range (1s^{-1}: 90%, 10 s^{-1}: 83%, 50 s^{-1}: 74%, 100 s^{-1}: 68%, 200 s^{-1}: 61%), relative to previous studies investigating viscosity as a variable involved in sensory perception, the viscosity range of this sample set is considerably smaller (for example: 0.003-2.187 Pas (Christensen and Casper, 1987); 0.003-2.24 Pas (Smith *et al.*,1997)). Therefore, findings are most relevant to low-viscosity emulsion systems (Vingerhoeds *et al*., 2008). Viscosity data between 0.1-1 S^{-1} was omitted due to highly deviating data being produced, a common result of measuring low viscosity systems at very low shear rates.

In regards to the relationship between physical and orally perceived viscosity, a power function with an exponent of 0.34 was shown by Christensen and Casper (1987), using a number of sodium alginate thickened solutions of viscosities ranging from 0.003-2.187 Pas. Other authors using other fluid food models and this viscosity range have also shown that an exponent of approximately 0.3 characterises the relationship between physical and orally perceived viscosity (Christensen, 1979; Smith *et al.*, 1997; Smith *et al.*, 2006).

Cutler, Morris & Taylor (1983), used thickness perception as a sensory interpretation of orally perceived viscosity. Correlating viscosity and Thickness perception for our results, expresses a power function with an exponent of -0.15. An exponent of this value highlights that physical differences in viscosity between samples would not be substantial enough for participants to perceive differences in viscosity and therefore we can infer that based on viscosity data, samples would be perceptually iso-viscous."

The findings of this study clearly do not fall within previous exponent boundaries. The reason why our results do not fall within previous exponent boundaries and show a perceivable difference in viscosity, where other authors have, may be due to two main factors. Firstly, the viscosity difference between systems in this study is significantly smaller than previous studies. For example Smith and other authors (1997) used corn syrup thickened solutions from 0.003-2.24 Pas. Within Smith's study each preceding solution was approximately three times greater in viscosity than the subsequent, therefore providing a perceptually discriminative sample set. This perceivable incremental increase in viscosity was not observed within our sample set, therefore we can conclude that within this study, modulating oil droplet size within an emulsion with a 15% oil fraction does not produce the viscosity difference needed for perceptually discriminative samples in relation to sensory interpretations of viscosity such as Thickness, based on previous findings. This conclusion is additionally highlighted by Table 3.3 and has been found by other authors investigating oil droplet size in low-viscosity systems when oil volume fraction is constant (Vingerhoeds *et al*., 2008). Future studies considering droplet size in relation to perception should look to produce systems of higher overall viscosity and with a greater inter-sample viscosity difference, if viscosity effects on perception are of interest.

Secondly, previous exponent values have be derived by using a viscosity value taken at 100 s^{-1}. As we have established that in relation to previous literature the samples within this study were significantly lower in viscosity, we can make the assumption that effective oral shear rate would be greater (Cutler, Morris & Taylor, 1983), thus the replication of previous methods to interpret instrumental and oral viscosity relationships may not be entirely appropriate and higher shear rates should therefore be considered. However, correlating viscosity at 200 s^{-1} and Thickness perception, expresses a power function with an exponent of -0.19, even at 10 and 50 s^{-1} an exponent indicative of no relationship is seen (-0.11 and 0.04 respectively). In conclusion, samples are deemed to be perceptually iso-viscous, based on viscosity measures across the shear rate range investigated and therefore viscosity will not be further considered as a physical variable in perception.

3.4.3. Emulsion tribology

The effect of droplet size on friction coefficient at 45 *U* is shown in Figure 3.3. Previous investigations that consider the effect of oil droplet size on lubrication behaviour have shown a linear trend of increasing friction coefficient with increasing droplet size (r: 0.96). This was demonstrated by de Wijk and Prinz (2005) in a 40 wt.% oil-in-water soft-solid system (mayonnaise) with a droplet size range of 2-6 µm.

Our results in sodium caseinate liquid emulsion systems agree with de Wijk and Prinz (2005) for droplets between 2-6 µm. Increasing friction coefficient is observed with increasing droplet size at oral processing related entrainment speeds (45 mm/s) for a droplet size range of 0.2 to 6 µm, demonstrating a positive linear relationship (r = 0.8, R^2: 0.64). However, this trend would imply that friction coefficient increases with droplet size indefinitely. For these systems, our results demonstrate an alternative behaviour with increasing droplet size.

Figure 3.3 demonstrates that for droplet sizes larger than 6 µm the friction coefficient begins to decrease. This results in two extreme droplet sizes (0.2 and 50 µm) having comparable friction coefficients, (0.5 N: 0.24 and 0.12 μ, 1 N: 0.17 and 0.19 μ, 3 N: 0.11 and 0.16 μ, respectively). At all applied ball loads, 0.2 µm droplets exhibit a noticeable different friction coefficient from the negative linear trend observed between 6 – 50 µm (0.5 N: r = -0.99, R^2: 0.98, 1 N: r = - 0.98, R^2: 0.96 and 3 N: r = - 0.95, R^2: 0.91, respectively) (see Fig. 3.3). This indicates a unique behaviour of droplets of this size in relation to lubrication. Therefore, at this range of droplet sizes, friction coefficient in relation to droplet size demonstrates an increasing linear trend up to 6 µm and then a decreasing linear trend afterwards. This highlights two distinct lubrication mechanisms as a function of droplet size (0.2 - 6 µm droplets and 6 > – 50 µm droplets), providing equal lubrication at extreme droplet sizes.

Additionally, although friction coefficient shows load dependence, with increasing friction with decreasing load, the trend observed is independent of load. Increasing friction with decreasing load further supports other author's conclusions that deformable tribometer surfaces show this relationship, as oppose to non-deformable tribometer surfaces, such as glass, in which the opposite behaviour is observed (de Hoog *et al.*, 2006); A result of two non-deformable surfaces promoting quicker onset of boundary regime lubrication behaviour when a greater force is applied.

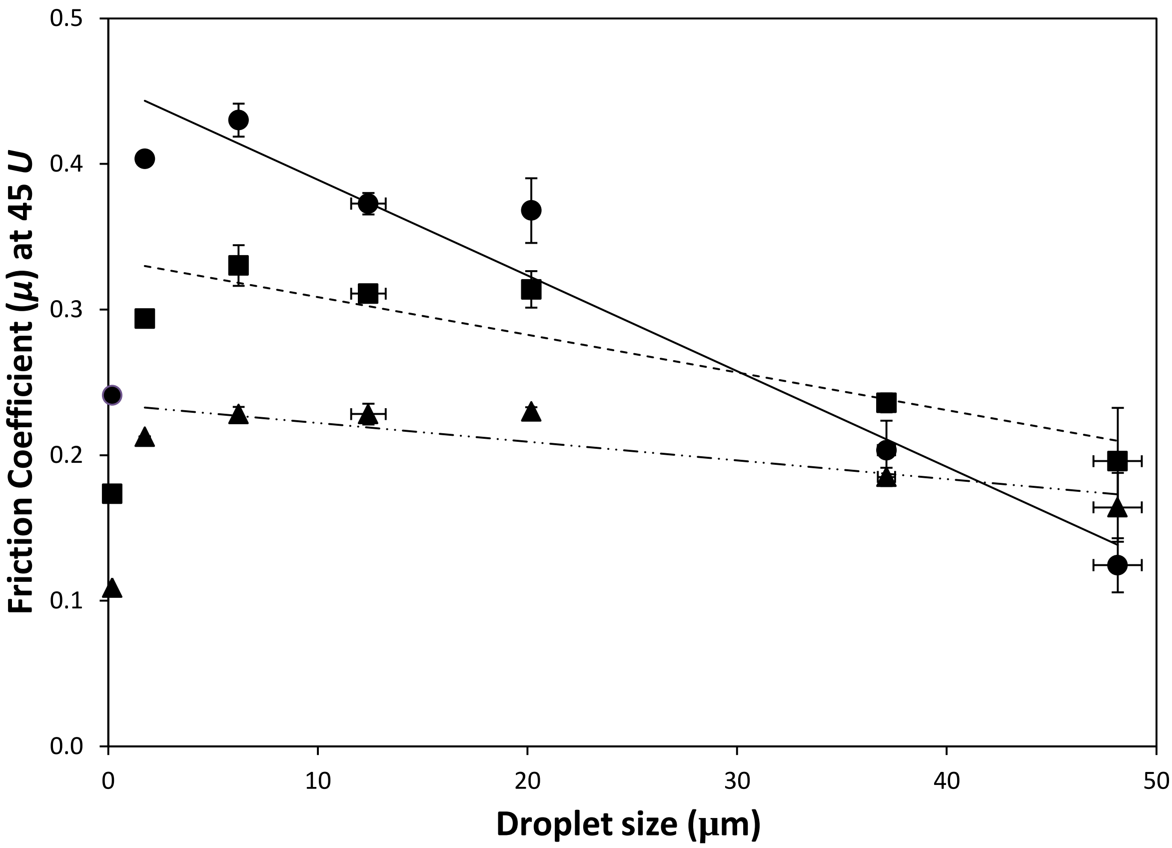


Fig. 3.3 Friction Coefficient (μ) at 45 *U*, from Stribeck curve, with a 0.5 N (●), 1 N (■) and 3 N (▲) ball load as a function of droplet size ($d_{4,3}$, 0.2-50 μm). Linear trend lines represent relationship between 2 – 50 μm. Error bars representative of one standard deviation.

3.4.3.1. 0.2 - 6 μm droplets lubrication behaviour

Our explanation for the observed lubrication behaviour between droplets of 0.2 to 6 μm is attributed to the manner in which the droplets behave within, and form, the lubricant film.

When lower droplet deformability is observed (see Eq. 3.5), the harder sphere behaviour offers a greater ability to repel the mating ball and disc within the contact zone, thus reducing the number of contacting sites between surfaces. Through calculating droplet deformation parameter (see Eq. 3.5 and Table 3.2), Greater resistance to deformation, therefore harder sphere behaviour, is observed with decreasing droplet size. However due to the nature in which friction coefficient is measured, droplet size may have changed throughout the experiment. In order to determine whether droplet size changed under test conditions, oil droplet size was measured post-tribometer experiment (see Fig. 3.4). Results indicate that emulsions with a droplet size of between 0.2 and 6 µm were resistant to droplet break-up in the contact zone of any significance under all test conditions (5-10% $d_{4,3}$ change). Applying droplet dynamic understanding (Stone, 1994), we can assume droplets of these sizes are resistance to significant size change due to their low deformability relative to droplets of a greater size, in which significant droplet break up is seen (see Table 3.2 and Fig. 3.4).

As a result, we conclude between 0.2 to 6 µm droplet size and its relative minimal droplet deformation, as droplet change is minimal and relatively constant, is responsible for the observed lubrication behaviour (see Fig. 3.3 and Fig. 3.5). Due to this conclusion, it is important to note deformation is shown to be force dependent (See Table 3.2). As increasing applied normal force from 0.5N to 3N showed to increase droplet deformation (0.2 µm: 48%; 2 µm: 44%; 6 µm: 43%; 12 µm: 47%; 20 µm: 48%; 40 µm: 57% and 50 µm: 61%) due to greater shear stress within the contact zone (see Eq. 3.7).

Table 3.2 Calculated deformation parameter for loads of 0.5, 1 and 3 N at 45 *U* as a function of droplet size ($d_{4,3,}$ µm).

$d_{4,3}$ (µm)	**Deformation Parameter (D)**		
	0.5 N	**1 N**	**3 N**
0.2	0.32	0.38	0.52
2	4.1	4.7	6.4
6	14	17	22
12	27	33	44
20	44	54	73
40	72	93	128
50	87	117	164

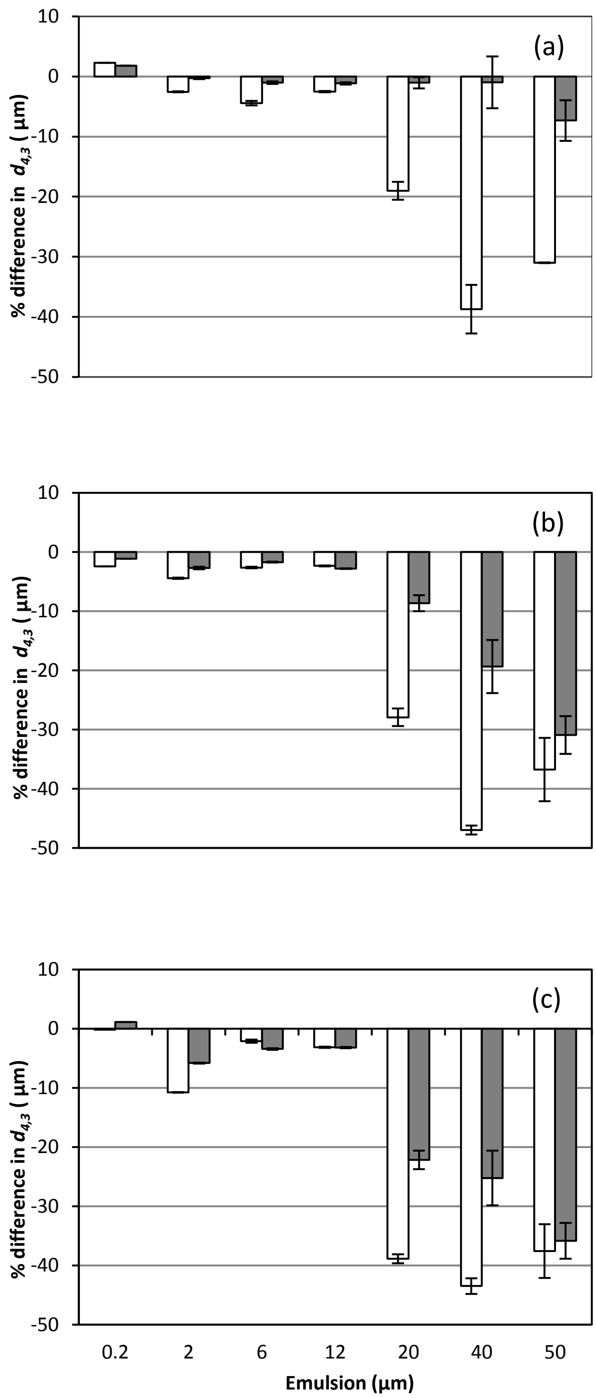


Fig. 3.4 Difference in Oil droplet size ($d_{4,3}$, µm) between pre-tribometer experiment and post tribometer experiment at 0.5 N (a), 1 N (b) and 3 N (c) both within the contact zone (open bars) and within the bulk (Filled bars). Error bars representative of one standard deviation.

Previous studies have also attributed deformability to a lubrication mechanism, however with gel particles in carrageenan fluid gels (Mills, Norton & Bakalis, 2013). The authors suggested that a reduction in deformability resulted in increased difficulty in particle inclusion into the contact zone, thus resulting in lower lubrication capacity. A similar behaviour might be occurring for the emulsions with oil droplets of 2 and 6 µm, given that they are the least lubricating across all entrainment speeds (see Fig. 3.5), and have minimal deformability (see Table 3.2). Especially at lower entrainment speeds (1 – 30 mm/s) (see Fig. 3.5) where tribopair surfaces are closer together and flow through the contact zone is reduced, resulting in conditions less supportive for contact zone droplet entrainment. It is important to note that we are not inferring droplets of these sizes are completely excluded from the contact zone, given lubrication behaviour is not that of the continuous phase alone (see Fig. 3.5). Instead results indicate that droplets of these sizes, which are included within the contact zone, increase friction, probably a result of droplet ordering behaviour, which has been shown to be of tribological importance (de Wijk & Prinz, 2005; Mills, Koay and Norton, 2013).

As highlighted previously, 0.2 µm droplets do not fit the negative linear trend observed between 2 – 50 µm (see Fig. 3.3), which is indicative of a lubrication mechanism specific to droplets of this size. To identify the lubrication mechanism of the emulsion containing 0.2 µm droplets, we must consider the tribopair roughness (silicone elastomer and stainless steel). Through characterising the surface roughness of tribometer surfaces (Garrec & Norton, 2013), silicone elastomer asperities appear to be sufficiently large enough to accommodate oil droplets of 0.2 µm. Previous work investigating the relationship between particle size and surface entrainment has typically considered gel particles, with particles of 0.97 - 1.34 µm being shown to be entrained within silicone elastomer asperities (Garrec & Norton, 2013; Garrec *et al.,* 2012). Given that emulsions containing 0.2 µm oil droplets display minimal deformation (See Table 3.2) and size change (See Fig. 3.4), this diameter would be maintained. As a result, 0.2 µm droplet entrainment at all speeds seems reasonable. A behaviour shown at most speeds and all ball loads, given a hysteresis is observed, with lower friction coefficient on reducing entrainment speed sweeps, than on increasing sweeps (Fig. 3.6). As decreasing entrainment speeds encourage boundary lubrication conditions, which are less supportive of droplet inclusion into the contact zone, it seems reasonable to suggest as entrainment speed decreases, droplets remain within the contact zone reducing the contact of the surfaces and therefore reducing friction. This behaviour is unique to 0.2 µm droplets, as all other droplet sizes show no hysteresis across a measurement; this is due to the other droplet sizes investigated being too large to be entrained within the silicone elastomer asperities (Garrec & Norton, 2013; Garrec *et al.*, 2012).

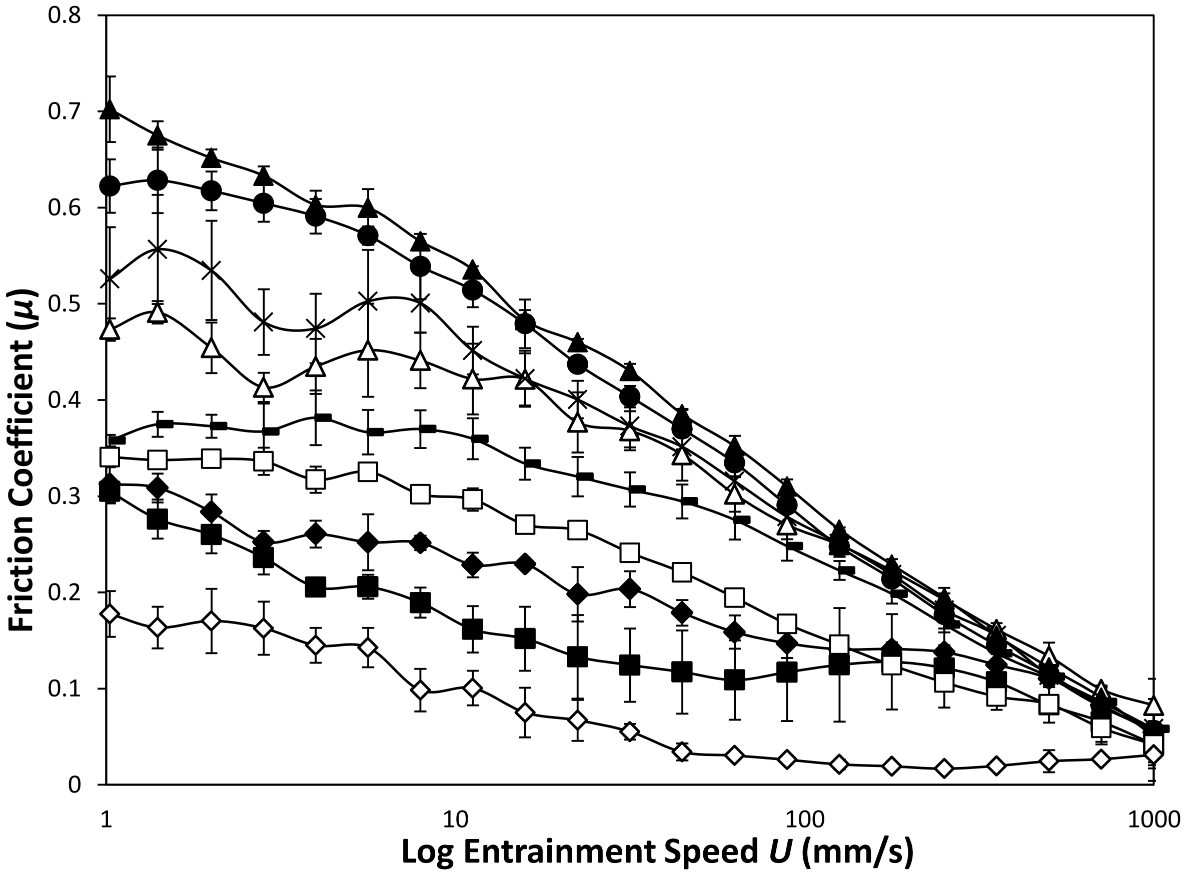


Fig. 3.5 Stribeck curves at 0.5 N of 1% NaCas 15% sunflower oil-in-water emulsions varying in average oil droplet size: Continuous phase (-), Sunflower oil (◊) 0.2 µm (□), 2 µm (●), 6 µm (▲), 12 µm (X), 20 µm (△), 40 µm (◆) and 50 µm (■).Error bars representative of one standard deviation.

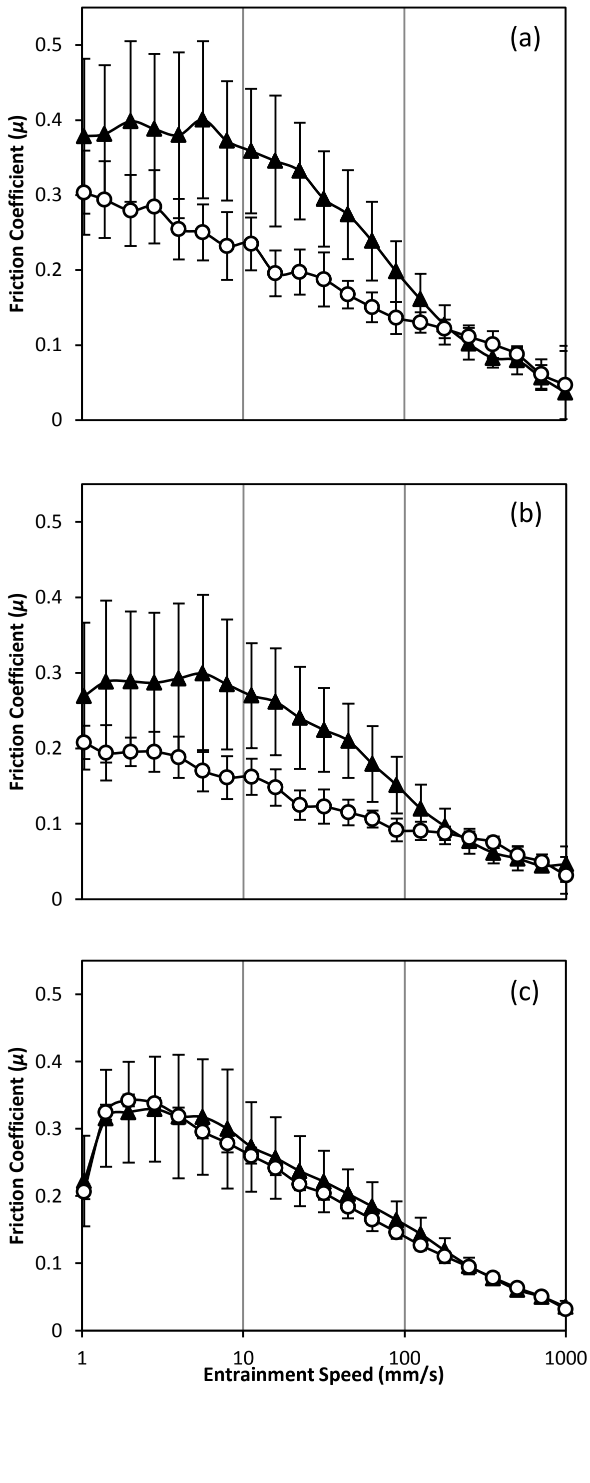


Fig. 3.6 Stribeck curve at 0.5 N (a), 1 N (b) and 3 N (c) for increasing (▲) and decreasing (○) entrainment speed of 1% NaCas 15% sunflower oil-in-water emulsions with an average oil droplet size of 0.2 µm. Error bars representative of one standard deviation.

3.4.3.2. 12 - 50 µm droplets lubrication behaviour

A negative linear trend of decreasing friction coefficient with increasing droplet size is observed at all ball loads at 45 *U*, for emulsions containing droplets greater than 6 µm (See Fig 3.3).

Given our conclusion for the minimal deformation mechanism in which emulsions with 0.2 – 6 µm droplets lubricate, we can deduce that for droplets above these sizes, a change in droplet behaviour must occur. Otherwise we would expect a continual increase in friction coefficient with increasing droplet size until complete droplet exclusion from the contact zone is reached, as non-deformable droplets would not be able to be entrained to provide lubrication due to their larger size. Droplet exclusion from the contact zone would result in a friction coefficient comparable to that of pure continuous phase, a result not observed within this study (see Fig. 3.5), but has previously been shown with larger fluid gel particles (Gabriele *et al.*, 2010; de Vicente, Stokes, & Spikes, 2006). However, unlike fluid gel particles, which are less deformable, with increasing oil droplet size a linear increase in droplet deformability (see Table 3.2) and droplet break-up in both the bulk and contact zone (see Fig. 3.4) is observed. Deformation would promote droplets within the contact zone to be present in an ellipsoidal shape, generating a lubricant film comprised of deformed ellipses, with greater elongational deformation being achieved with increasing droplet size. Additionally, droplet break up would reduce droplet size promoting easier inclusion into the contact zone. As such, it is reasonable to suggest that droplet deformability and break-up within the contact zone are key factors in relation to lubrication behaviour of droplets greater than 6 µm.

Stribeck curves (see. Fig 3.6) highlight that between entrainments speeds of 20 and 1000 mm/s, emulsions containing 12 and 20 µm droplets are comparable in lubrication to emulsions containing 2 and 6 µm droplets. This indicates at these entrainment speeds, like 2 and 6 µm droplets, 12 and 20 µm droplets also order within the contact zone, in a way which increases friction above that of the continuous phase alone (see section 3.4.3.1). However, between 1 and 15 mm/s, friction coefficient is lower. The decrease in droplet size post-tribometer test for emulsions with 12 µm droplets was non-significant at all applied loads (5-10% $d_{4,3}$ reduction). However, emulsions with 20 µm droplets show a 20 – 40% $d_{4,3}$ reduction dependent on load applied (see. Fig. 3.4). Droplet deformability is also higher than 2 and 6 µm droplets, due to their greater size (See Table 3.2).This suggests that at lower entrainment speeds 12 µm droplet lubrication is predominantly a function of deformability and 20 µm droplet lubrication is predominantly a function of droplet break-up to a droplet size comparable to 12 µm, where after lubrication would be largely controlled by droplet deformation. It seems sensible to suggest, both of these behaviours are promoting favourability to their inclusion within the contact zone in comparison to 2 and 6 µm droplets at lower entrainment speeds.

As well the largest droplet deformation (see Table 3.2), Significant droplet break-up within the contact zone (40% $d_{4,3}$ reduction) is also observed in emulsions consisting of 40 and 50 µm droplets, at all applied loads (see. Fig. 3.4) (Droplet span: **40 µm**: pre-test: 1.16, 0.5 N: 2.2, 1 N: 2.35, 3 N: 1.79; **50 µm**: pre-test: 1.46, 0.5 N: 2.8, 1 N: 2.76, 3 N: 2.75). As droplet span increases we can deduce that droplet break-up was not uniform. The lubricate film would therefore be expected to be comprised of droplets of varied sizes, all capable of deforming to fit within the contact zone, but also capable of fitting around other droplets within in the contact zone. This can be deduced as fluid drag associated with an increase in friction is observed between 125 and 350 mm/s (see Fig. 3.5) (Chen & Stokes, 2012), a function determined by lubricant viscosity, which would be expected to locally increase within the contact zone with increasing droplet packing. Additionally given that span values show that droplet break-up resulted in no sub-micron sized droplets, and no hysteresis is shown in the measurement, the presence of droplets small enough to fit in surface asperities within the contact zone seems unlikely. Furthermore, a droplet “oil pool” effect, resulting in a contact zone comprised of predominantly oil phase seems improbable, given droplet size decreased post-tribometer test (see Fig. 3.4) and the emulsions were not as lubricating as pure sunflower oil at any load or entrainment speed (see Fig. 3.5). As a result, we can conclude that the extent of droplet break-up and deformation behaviours in 40 and 50 µm droplets, relative to other droplet sizes, seems to be providing the lubrication mechanism resulting in the greatest reduction in friction coefficient (see Fig. 3.5).

Despite the relatively low viscosity ratio ($\lambda\mu$: 0.05) droplet break-up via droplet elongation will only occur when critical conditions are present. Large droplet deformation is required to promote droplet break-up, which is dependent on factors such as viscosity ratio, applied shear, flow type, flow conditions, and/or droplet features, such as interfacial tension and/or size (Stone, 1994). Size may explain why in the contact zone 1) 20, 40 and 50 µm droplets display a greater reduction in $d_{4,3}$ post-tribometer experiment and 2) why the reduction in $d_{4,3}$ is observed to be greater with increasing droplet size (see Fig. 3.4). Firstly, droplets >20 µm must be an adequate size to give a sufficient level of elongation deformation to promote significant break-up, under the given conditions. Secondly, as a greater volume of oil per droplet is present and greater deformation is seen with increasing droplet size (see Table 3.2), we can presume that larger droplets break-up into a greater number of smaller droplets (see Fig. 3.4).

Droplet size clearly is of tribological importance in relation the lubrication behaviour. Given our suggested mechanisms, the behaviour of how droplets from 0.2 - 50 µm lubricate should therefore shift dependent on factors affecting droplet dynamics, an area to consider for further investigation.

3.4.4. Emulsion oil droplet size and sensory perception

Modulating oil droplet size resulted in different perceptions of texture and mouthfeel related sensory attributes (see Table 3.3). Relationships were analysed as a whole group of samples (0.2 - 50 µm), and to allow the discussion to be compared to previous literature (Akhtar *et al.*, 2005; Kilcast & Clegg, 2002; Vingerhoeds *et al.*, 2008), droplet size and sensory data were further split into a smaller group (0.2 – 6 µm).

Table 3.3 Coefficient of determination (R^2) and Pearson correlation (r) for linear trends between droplet sizes of (a) 0.2 - 50 µm and (b) 0.2 - 6 µm and sensory attributes.

	(a) 0.2 - 50 µm		**(b) 0.2 - 6 µm**	
Attribute (mm)	$\mathbf{R^2}$	**r**	$\mathbf{R^2}$	**r**
Thickness	[a]0.24	- 0.49	0.00	- 0.05
Smoothness	[a]0.30 [b]0.81	- 0.55	0.02	0.13
Slipperiness	[a]0.03 [b]0.87	- 0.17	0.61	- 0.78
Creaminess	[a]0.86	- 0.93*	0.06	- 0.24
Oiliness	[a]0.03 [b]0.66	0.17	0.81	- 0.90

*correlation coefficient is significant at $p < 0.05$.

'[a]' denotes a linear trendline, and '[b]' denotes a polynomial (order:2) trendline.

3.4.4.1 Creaminess

Creaminess perception was shown to very strongly, significantly negatively correlate with droplet size (see Table 3.3a and Fig. 3.7d). Therefore, by simply changing the dispersion of oil within the system, perceptions of Creaminess can be significantly changed. This offers the potential to alter Creaminess of food products by introducing a processing step which results in a smaller droplet size (for example, higher shear/pressure processing). However, Creaminess perception is only significantly different when droplets from 0.2 - 50 µm are considered, with no significant difference in perception between 0.2 – 6 µm. This non-significant effect on perception is in line with the results of others who have previously investigated droplets 0.2 – 6 µm in similar systems (Akhtar *et al*., 2005; Kilcast & Clegg, 2002; Vingerhoeds *et al*., 2008). Creamy Mouthfeel data was removed from discussion as Creamy Mouthfeel and Creaminess scores were extremely similar (R^2: 0.98). This could be attributed to untrained participants perceiving foods in a less sensitive manner, assessing the totality of the food, i.e. Overall Creaminess, in comparison to trained panels which assess food analytically i.e. Overall Creaminess, Creamy Mouthfeel, Creamy flavour (Frost and Janhoj, 2007). Overall Creaminess will only be discussed in later sections of this chapter.

3.4.4.2 Smoothness, Slipperiness and Oiliness

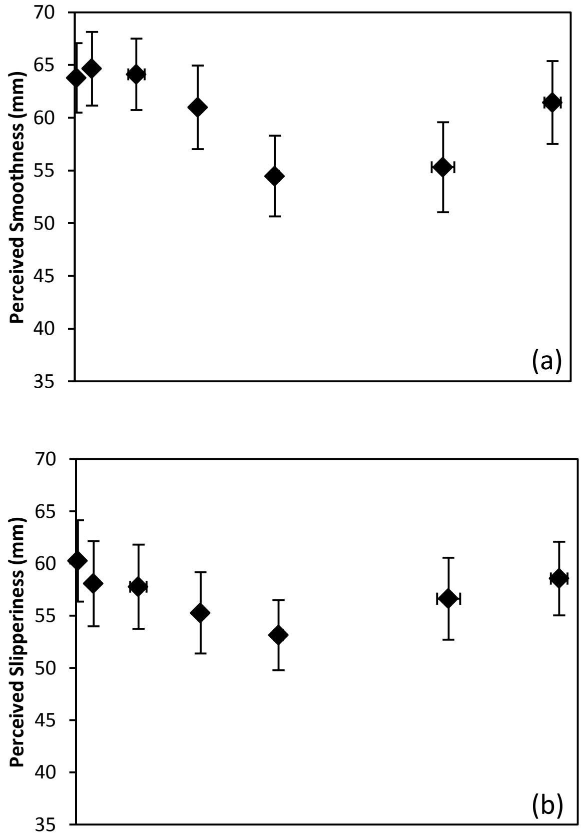
Attributes considered to be related to friction such as Smoothness and Slipperiness (Kokini, 1987; Kokini & Cussler, 1983; Kokini, Kadane & Cussler, 1977; Malone, Appelqvist & Norton, 2003) and Oiliness (section 3.4.5) all exhibited moderate to weak or no linear correlation with droplet size, but instead all three ratings showed stronger polynomial relationships with droplet size between 0.2 - 50 µm (See Table 3.3a). Extreme droplets sizes resulted in comparable ratings of these particular attributes, with perception intensity reducing with mid-range droplet sizes (see Fig 3.7a, b and c). Therefore it seems reasonable to suggest, given friction coefficient in relation to droplet size demonstrates an increasing linear trend up to 6 µm and then a decreasing linear trend afterwards (see Fig. 3.3), that the relationship between droplet size and Slipperiness and Oiliness perception at the full droplet size range considered could be a result of friction.

However, it is clear from figure 3.7 (a, b and c) that perception of Smoothness, Slipperiness and Oiliness in 40 and 50 µm droplet samples does not follow the negative linear trend of decreasing friction coefficient with increasing droplet size completely (see Fig. 3.3). This could be a result of the differences between oral conditions and instrumental conditions, and may explain why although a very strong polynomial correlation for Smoothness is shown for the 0.2 - 50 µm droplet size range, no linear relationship between 0.2 – 6 µm is shown (see

Table 3.3b) highlighting possible dependence on additional influences such as droplet oral behaviour. Nevertheless, droplet behaviour during oral processing could somewhat reflect similar behaviours discussed in relation to instrumental data, as friction coefficient and friction related attributes Slipperiness and Oiliness both show similar trends.

3.4.4.3 Thickness

The weak and non-significant correlation between droplet size and Thickness reinforces our discussion in section 3.4.2 that samples were perceptually iso-viscous, which can be attributed to the small difference in sample rheological behaviour (See Fig. 3.2).



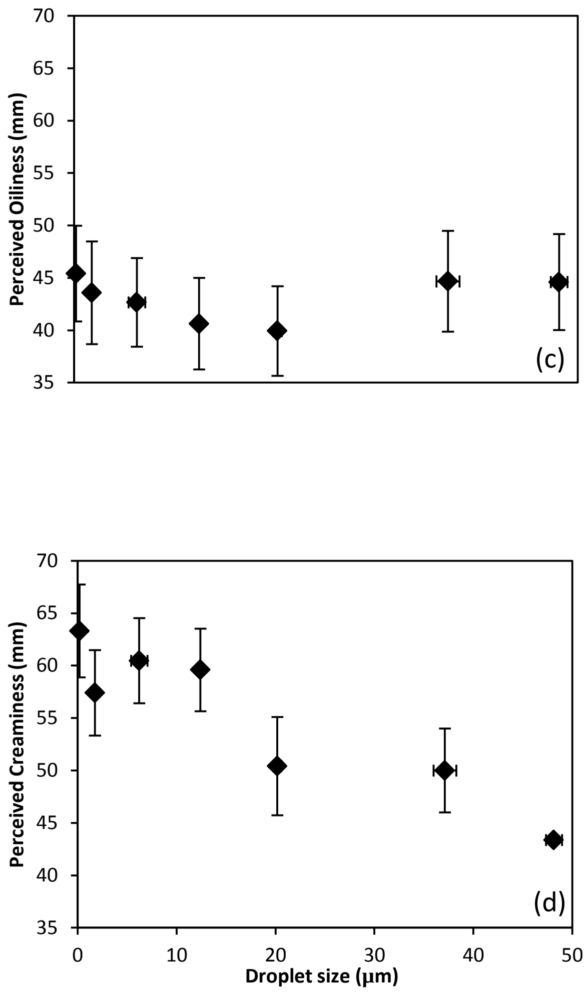


Fig. 3.7 Relationship between droplet size ($d_{4,3}$, μm ± standard deviation) and ratings of perceived Smoothness (a), Slipperiness (b), Oiliness (c) and Creaminess (d). Sensory rating error bars representative of one standard error of the mean.

3.4.5. Tribology and sensory perception

The correlation between friction coefficient at assumed oral entrainment speed (45 *U*) and the 3 ball loads (0.5, 1 and 3 N) with sensory ratings are shown in Table 3.4. As systems were deemed perceptually iso-viscous, the only measured variable to change perception was friction.

Table 3.4 Coefficient of determination (R^2) (linear) and Persons correlation (r) of friction coefficient taken at different ball loads (0.5, 1 and 3 N) as a function of a sensory attribute for droplets between 0.2 – 50 µm (a) and 0.2 - 6 µm (b).

(a) 0.2 - 50 µm						
Attribute (mm)	**R^2 value**			**r value**		
	0.5N	**1N**	**3N**	**0.5N**	**1N**	**3N**
Thickness	0.05	0.00	0.03	0.22	0.00	- 0.18
Smoothness	0.05	0.01	0.05	0.23	- 0.08	- 0.23
Slipperiness	0.14	0.44	0.56	- 0.37	- 0.66	- 0.75
Creaminess	0.37	0.05	0.00	0.61	0.22	- 0.05
Oiliness	0.42	0.66	0.65	- 0.65	- 0.81*	- 0.81*
(b) 0.2 - 6 µm						
Attribute (mm)	**R^2 value**			**r value**		
	0.5N	**1N**	**3N**	**0.5N**	**1N**	**3N**
Thickness	0.41	0.32	0.42	- 0.64	- 0.57	- 0.65
Smoothness	0.5	0.4	0.51	0.70	-0.63	0.71
Slipperiness	1	0.99	1	- 1*	- 0.99*	- 1*
Creaminess	0.61	0.52	0.62	- 0.78	- 0.72	- 0.79
Oiliness	0.96	0.99	0.96	- 0.98	- 0.99*	- 0.97

*correlation coefficient is significant at $p < 0.05$.

3.4.5.1 Slipperiness and Oiliness

Frictional properties and lubrication behaviour of droplets (see Fig. 3.3), are suspected to be responsible for the polynomial trend demonstrated for 0.2 – 50 µm droplets, within the perception of Slipperiness and Oiliness (see Table 3.3a, Fig 3.7b and c). Table 3.4 shows that with decreasing friction we observe increasing Slipperiness and Oiliness. Our results demonstrate that Oiliness strongly correlates with both droplet size ranges investigated, with significance shown in most relationships (see Table 3.4a and b). Importantly, Oiliness significantly correlated with friction between 0.2 – 6 µm. The only other previously investigated attribute in the literature to significantly correlated with friction between 0.2 – 6 µm was Slipperiness (Akhtar *et al.*, 2005; Vingerhoeds *et al.*, 2008). Due to samples being perceptually iso-viscous, our findings indicate that the predominant physical property in Oiliness perception is friction. However, it is important to note other factors, not measured within this study, such as flavour, may also be involved in Oiliness perception. For example, greater droplet diameter promotes greater lipophilic flavour release (Van Ruth, King and Giannouli, 2002), which theoretically would increase consumers awareness to oil within the mouth and therefore may impact upon Oiliness intensity. Additionally, behaviours such as droplet spreading and retention could be influential and would be of interest for further investigation (de Hoog *et al.*, 2006).

Slipperiness demonstrates strong correlations with friction coefficient between 2 - 6 µm. However, only a weak relationship is observed when the entire range is considered (See Table 3.4a), with the correlation strength between Slipperiness perception and friction coefficient increasing with ball load. Friction involvement in Slipperiness perception agrees with previous findings (Kokini, 1987), however a weak relationship is observed when the entire range is considered, indicating alternative influences other than friction in Slipperiness perception, such as the difference in droplet stability during oral processing (de Hoog *et al.*, 2006).

3.4.5.2 Smoothness

Smoothness only demonstrates weak and moderate correlations with friction coefficient (see Table 3.4), which is surprising considering Kokini (1987) concluded that friction is the only physical property involved in Smoothness perception. A very strong polynomial correlation for Smoothness perception and droplet size was observed (See Fig. 3.7a), which suggests a relationship with friction coefficient however no direct correlation is observed. The similarity in correlation between Figure 3.3 and Figure 3.7a, seems too coincidental in perceptually iso-viscous systems to dismiss friction in Smoothness perception. However, it seems reasonable to suggest that additional influences, not measured or considered within this study, may be involved in its perception, skewing correlation strength.

3.4.5.3 Thickness

Thickness perception was not significantly correlated with friction (see Table. 3.4). Based on the strength of previous findings, we have highlighted systems were perceptually iso-viscous, and therefore can assume Thickness perception is only influenced by viscosity. However, these results do contribute to understanding by indicating that friction is not involved in the perception of Thickness, which agrees with the literature consensus and Kokini's model of oral lubrication (Kokini, 1987).

3.4.5.4 Creaminess

Our results show only non-significant weak correlations between friction and Creaminess (see Table. 3.4a) at the full droplet size range investigated. However, between 0.2 - 6 µm, the range which has been typically investigated, Creaminess shows strong correlations with friction coefficient at all ball loads (see Table 3.4b and 3.6c). Between 0.2 - 6 µm the relationship indicates that as friction increases, Creaminess decreases, a trend reflecting findings by other authors (Kokini, 1987). The weak correlations found between Creaminess and friction coefficient for 0.2 – 50 µm (see Table 3.4a) could be explained by Kokini and Cussler's (1983) Creaminess predication equation (see equation 3.1), as both Thickness and Smoothness also show weak correlations with friction coefficient, and viscosity perceptually was not influential in this study. This conclusion reinforces Kokini and Cusslers's (1983) findings that Creaminess is indirectly influenced by physical properties such as friction via perception of other attributes (Smoothness and Thickness).

3.4.5.5. Technique considerations

It would be expected that as these emulsions were liquid, mating of the oral surfaces would encourage the emulsion to be present within the oral cavity as a thin-film. In light of the points discussed, as droplets of varying sizes behave differently within a thin-film (section 3.4.3), it would be beneficial for future work to consider these potential droplet behaviours, to gain further understanding of how oral and tribometer contact zone behaviour differs. Additionally, it should be noted that results were obtained on silicone elastomer, in which there are substantial differences in surface behaviours, presence of oral secretions and macroscopic roughness between that of oral surfaces such as the tongue (Dresselhuis *et al.*, 2008a), which are likely to have an effect on sensory perception meaning that the strength of correlation between instrumental measures and perception is likely to be affected.

3.5. Conclusions

This work has demonstrated that oil droplet size within sodium caseinate stabilised oil-in-water emulsions does affect the perception of certain sensory attributes.

The oil droplet size investigated was important in regards to perception and tribological behaviour, and extends previous understanding only considering oil droplet sizes between 0.2 - 6 µm. This approach additionally highlighted why previously investigated droplet size ranges may have only shown minor non-significant differences in perception.

Friction was shown to be involved in the perception of certain attributes (Oiliness and Slipperiness), further demonstrating the value of tribological techniques in understanding the physical make-up of sensory attributes and how modification of emulsion microstructure affects a foods sensory profile. Limitations in only using these techniques were additionally highlighted, with certain attributes not being fully explained by physical properties. However, indications that some of the differences in perception might be a result of droplet behaviour during oral processing, were highlighted. Further understanding of physical measures in relation to perception is still required, and this work reinforces the need for new approaches to understand the physical constitutes of sensory attributes.

Chapter 4. Iso-viscous and Iso-friction oil-in-water emulsions: The contribution of viscosity and friction in sensory perception

Data and discussions contained within this chapter are in preparation to be submitted for publication within:

Lett, A.M., Norton, J.E., Yeomans, M.R. and Norton, I.T. (In preparation). Iso-viscous and Iso-friction oil-in-water emulsions: The contribution of viscosity and friction in sensory perception. *Food Hydrocolloids*

4.1. Abstract

Both viscosity and frictional properties experienced within the oral cavity during the consumption of liquid foods are considered to be the prominent physical properties that influence textural perception. A series of liquid oil-in-water emulsions, stabilised by sodium caseinate, which varied in droplet size ($d_{4,3}$: 0.1 - 55 µm) were formulated to either be iso-viscous or iso-frictional, using the thickener guar gum. These emulsions were assessed with respect to sensory perception (Thickness, Smoothness, Slipperiness and Creaminess) by male participants (n =16). Viscosity was matched at 50 s^{-1} and friction coefficient was matched at 45 mm/s. This approach was taken to evaluate the independent contribution of viscosity and friction in perception of textural attributes, and then assess where these physical properties have a combined effect on sensory perception. Sensory results showed that: 1) Thickness was the sensory interpretation of viscosity, 2) Smoothness perception was mediated by droplet behaviour, which varied as a function of droplet size, and 3) Creaminess perception was influenced by both viscosity and droplet behaviour, where Thickness and Smoothness are the sensory interpretation of these two physical variables. This is in agreement with earlier findings that showed that Creaminess is a combination of these independent sensory variables. Overall, participants were not able to perceive sensory differences as a result of the manipulation in frictional properties, however significant differences in perception were still shown in iso-viscous emulsions. This is thought to be as a result of droplet behaviour during consumption, which was a function of droplet size. This study has highlighted the influence of viscosity and droplet behaviour during oral processing on textural perception.

4.2. Introduction

Oral processing is a dynamic process, which exerts a range of shears, stresses and forces upon a food (Stokes, Boehm & Baier, 2013; van Aken, Vingerhoeds & de Hoog, 2007). Various oral physical forces can somewhat be replicated using instrumental measures (Stokes, Boehm & Baier, 2013). Through research of this nature insight in to what physical forces are applied in the mouth, and the resulting effect on sensory perception has been gained.

Food texture is largely determined by the foods material behaviour upon applied forces in the mouth (Akhtar, Murray & Dickinson, 2006; Foster et al., 2011; Kokini, 1987). This has been deduced by correlating sensory scores of textural attributes with physical properties measured under replicated physical conditions to those experienced in the mouth. The earliest work of this nature considered the physical property viscosity in textural perception. By using hydrocolloid thickened soups, Wood (1968) showed that 50 s^{-1} is representative of shear rates applied in the mouth. Since this finding, 50 s^{-1} has been extensively used as a convenient value to represent the shear rate experienced during oral processing of fluid foods. Further studies have shown strong correlations between viscosity and the perceptions of Thickness, with this relationship being one of the most extensively investigated in fluid foods (Akhtar, Murray & Dickinson, 2006; Cutler, Morris & Taylor, 1983; Kokini, Kadane & Cussler, 1977; Morris & Taylor, 1982; Morris, Richardson & Taylor, 1984; Shama & Sherman, 1973a; Shama & Sherman, 1973b).

Viscosity has also been shown to be influential in perception of other sensory attributes, such as Slipperiness and Creaminess (Kokini & Cussler, 1984; Kokini, 1987). However, unlike Thickness perception, the perception of Slipperiness and Creaminess are not fully explained by viscosity, and instead are dependent on additional physical properties. Creaminess, for example, is thought to be indirectly dependent on viscosity and friction, through perception of ‘‘Thickness’’ and ‘‘Smoothness’’ respectively, the two independent sensory attributes that Kokini (1987) used to model Creaminess perception.

Typically, in the latter stages of oral processing, emulsions are often present within a thin-film. Therefore, frictional sensations, a result of thin-film rheological behaviour, are influential in textural perception (Chen & Stokes, 2012; Stokes, Boehm & Baier, 2013). Until recently, friction has been largely disregarded as a physical variable involved in sensory perception, with only more recent work considering its influence (Chen & Stokes, 2012). The influence of friction on sensory perception was drawn from conclusions that the perception of Smoothness correlated with the reciprocal of the friction force between oral surfaces, and Slipperiness correlated with the reciprocal of the sum of the viscous force and the friction force (Kokini, 1987). More recent work has developed our understanding of the influence of friction on perception, how friction occurs during oral processing, and has also looked to identify the effect of both formulation and food structure on friction. This has increased our understanding of the sensory attributes that are not completely explained by viscosity measurements. For food emulsions, attributes such as Smoothness, Slipperiness, Fattiness, Stickiness and Creaminess have been further understood, with some work even taking in to account the complex droplet and colloidal destabilisation behaviours which may occur during oral processing (Bellamy *et al.*, 2009; Chojnicka-Paszun, de Jongh & de Kruif, 2012; de Hoog *et al.*, 2006; de Wijk & Prinz, 2005; Dresselhuis *et al.*, 2008; Lett et al., 2016; Malone, Appelqvist & Norton, 2003).

As a result, it is agreed that viscosity and friction are two independent physical properties, both of which affect sensory perception. However, there is a clear deficiency in the literature. Previous research has not isolated these physical properties to investigate how they independently affect perception. For emulsions, work to date has isolated viscosity by designing iso-viscous samples using a hydrocolloid thickener (Akhtar, Murray & Dickinson, 2006; Malone, Appelqvist & Norton, 2003). The work mainly focused on the effect of fluid viscosity on oral shear rate, and/or thickener type and oil volume fraction on perception. Only Malone, Appelqvist and Norton (2003) have quantified and evaluated friction in iso-viscous emulsions and related it to sensory perception. Surprisingly, isolating friction in emulsions by designing iso-frictional samples has yet to be achieved. However, such research would be valuable in identifying the independent contribution of friction in sensory perception.

Therefore, the present study separates the influence of viscosity and friction in sensory perception by using oil-in-water emulsions that vary in oil droplet size that are either iso-viscous or iso-frictional, by using a hydrocolloid thickener. The work evaluates the contribution of 1) viscosity, 2) friction and 3) a combination of viscosity and friction, in perception of textural attributes.

4.3. Materials and methodology

4.3.1. Design and participants

Male participants were recruited via advertisement and screened for food allergies, smoking habits, body mass index (BMI), current medical status and dietary habits (restricted eating) via Dutch eating behaviour questionnaire (DEBQ) (van Strien, *et al.*,1986). Females were excluded as they typically practice significantly higher levels of restricted eating, along with significant higher levels of other eating behaviours than males (Wardle, 1987). The restricted eating DEBQ consisted of 10 questions each having a five-option response format: never (1), seldom (2), sometimes (3), often (4), and very often (5). A restraint score was obtained by summing the scores for the 10 items and dividing by 10. A higher score indicates greater dietary restraint. Potential participants were prevented from participating if they 1) indicated any food allergies, 2) had a history of smoking, 3) had a BMI above 24.9 Kg/M^2 or below

18.5 Kg/M^2, 4) were taking medication known to interfere with sensory perception or food intake or 5) had a DEBQ restricted eating score of >2.4 indicative of occasionally or more often exercising restricted eating behaviour. 19 participants were recruited, however only 16 respondents met the study criteria and were included in the study. Participants were aged 19 - 31, with a mean BMI of 22.6 ± 1.5 Kg/m^2 and DEBQ restricted eating score of 1.8 ± 0.6. All participants gave written informed consent prior to participation. The study was described as an investigation into the sensory analysis of emulsions. Ethical approval for the study was obtained from the University of Birmingham ethics committee.

4.3.2. Measurement of sensory perception

Test sessions were scheduled between 10 am and 12 am or 2 pm and 4 pm, Monday to Friday, with session lasting 1 to 1.5 hours. Participants were instructed to arrive on one occasion having refrained from consuming any food or beverages other than water 2 hours before their arrival. Participants were seated in individual sensory booths and were presented with 40 ml of sample in 60 ml pots coded with random 3 digit codes. Each sample type was tested in duplicate, using replicate samples, blind to the participant. All samples were served between 5-7 ^{o}C and were visually identical. Sample order was randomised differently for all assessors. Inter-sample duration was at the participant discretion and ranged from approximately 1-3 minutes. A spittoon was provided and subjects were instructed to spit out the sample after their assessment had been made. One 250 ml bottle of still water and 3 dry crackers were provided, and participants were instructed to use these to refresh their palate between samples.

Measurements of sensory attributes were made using visual analogue scales (VAS). Ten 100 mm randomised VAS's anchored with opposing statements left-to-right e.g. "Not at all <target attribute>" (scored as zero) and "Extremely [target attribute]" (scored as 100) representing each attribute were presented (See Table 4.1), headed with questions acquiring information about the intensity of the sensory perception e.g. “How <attribute> is sample <code>?”. Questions differed slightly in order to be grammatically correct. Before testing, all sensory attributes were discussed individually with participants in accordance to the description shown in Table 4.1. Participants were also given the opportunity to ask any questions about the study and its protocol in order to clarify issues, queries or definitions before the test began.

Table 4.1 Attributes used during measurements of sensory perception with the description discussed verbally with participants.

Attribute category	**Sensory attribute**	**Description reference**
Mouthfeel	Thickness	Viscous consistency within the mouth
	Smoothness	Degree of absence of any particles, lumps, bumps etc within the sample
	Slipperiness	Degree to which the product slides over the tongue
	Creamy	Soft, smooth with flowing consistency
Overall	Creaminess	Assessment of overall creaminess of the sample

4.3.3. Emulsions

Samples consisted of oil-in-water emulsions varying in their oil droplet size, designed to be either iso-viscous or iso-frictional. 1 wt.% sodium caseinate (NaCas) (Excellion EM7, DMV International, The Netherlands), 2 wt.% sucrose (Silverspoon granulated, British Sugar Plc, UK) and 15 wt.% sunflower oil (Tesco Plc, UK), dispersed in distilled water, formed the initial oil-in-water emulsions, in which guar gum was then added too to produce the iso-viscous or iso-frictional emulsion sets.

Emulsion droplet size was varied via processing. Two different methods, dependent upon the required mean droplet size of the emulsion being produced were used: a high shear mixer

(Silverson L5M, Silverson machines Ltd, UK) or a high-pressure homogeniser (GEA Niro Soavi Panda Plus 2000, GEA Niro Soavi, Italy). In a 600 ml beaker, 15 wt. % sunflower oil was added to an aqueous phase containing the NaCas and sucrose in a distilled water solution. The whole sample was then emulsified for 5 minutes using the high shear mixer. Dependent on oil droplet size being produced the sample was subjected to a different rotational speed (rpm) and emulsor screen (fine (0.8 mm pores) or medium (1.6 mm pores)) (**55 µm**: *2500 rpm medium screen* and **20 µm**: *5000 rpm fine screen*). For emulsions produced using the high-pressure homogeniser, first a pre-emulsion was produced using the high shear mixer at 9000 rpm with a fine emulsor screen for 5 minutes using the high shear mixer. The pre-emulsion was then subjected to homogenisation, differing in pressure and number of passes (8 **µm**: *20 Bar 3 passes* and **0.1 µm:** *1250 Bar 4 passes*).

Once the emulsion had be produced varying concentrations of guar gum were sieved (200 mesh 3500 cps, Impexar, UK) into the emulsions and mixed using a magnetic stirrer and flea, in order to either: 1) match viscosity at 50 s^{-1} (in order to produce iso-viscous emulsions), as this has been extensively used as a value to represent oral shear rate of fluid foods (Akhtar *et al.*, 2005; Akhtar, Murray & Dickinson, 2006; Shama & Sherman, 1973a; Stanley & Taylor, 1993; Wood, 1968) or 2) match friction coefficient at 45 mm/s (in order to produce iso-frictional emulsions), as Malone, Appelqvest and Norton (2003) showed some, if limited, sensory data for guar gum thickened solutions that indicated a significant correlation between lubrication properties and Slipperiness perception at entrainment speeds between 10 and 100 mm/s. This work suggested that this entrainment speed range is most relevant to oral processing associated with slippery mouthfeel, which is an attribute regarded as significantly influenced by the frictional properties of the food (Kokini, 1987). Thus the median value between 10 and 100 mm/s (45 mm/s) was chosen as value at which all iso-frictional emulsions should be matched. Iso-viscous and iso-frictional samples of the same droplet size produced for instrumental and sensory analysis used the same initial emulsion; samples were then taken and their respective guar gum concentrations added to achieve the required viscosity or friction.

The final formulation were as such (droplet size, guar gum concentration): 1) iso-viscous emulsions: 0.1 µm: 0.25 wt.%; 8 µm: 0.255 wt.%; 20 µm: 0.24 wt.%; 55 µm: 0.235 wt.%, and 2) iso-frictional emulsions: 0.1 µm: 0.15 wt.%; 8 µm: 0.5 wt.%; 20 µm: 0.43 wt.%; 55 µm: 0.125 wt.%. All samples were produced in 400 g batches, under clean and hygienic conditions on the day of evaluation and stored under refrigerated conditions at ~2-5 ^{o}C.

Matching emulsion viscosity and friction was achieved on a trial-and-error basis, with all emulsions of varying droplet size being subject to viscosity measurements at guar gum concentrations of 0 – 0.5%, increasing in 0.05 increments. Once viscosities or friction coefficients were within a similar range, guar gum concentration was manipulated further in 0.01% increments in order to more closely match viscosity or friction coefficient.

4.3.4. Droplet size characterisation

Oil droplet size distributions were measured using dynamic laser light scattering (Malvern Mastersizer MS2000, Malvern Instruments, UK). All measurements were performed in triplicate (using a new sample each measurement) and conducted on the day of emulsion manufacture. Oil droplet size $d_{4,3}$ values were recalculated to remove guar gum particle size distribution.

4.3.5. Rheological measurements

A rheometer (Bohlin Gemini HR Nano, Malvern Instruments, Malvern, UK) was used to perform bulk rheology measurements. Measurements were taken with a stainless steel four blade vane-in-cup geometry. Temperature was fixed at 37 °C to mimic oral temperature.

For all experiments, the sample was poured into the rheometer cup, and allowed to thermally equilibrate prior to the measurement. A series of viscosity ramps were used to measure viscosity between shear rates of 0.1 and 1000 s^{-1}. Viscosity ramp time was 5 minutes, with a measurement time of 5 seconds at each shear rate. Measurements were performed in triplicate (using a new sample each measurement), and the mean value and standard deviation were derived.

4.3.6. Tribological measurements

A ball-on-disc tribometer (MTM, PCS Instruments, UK) was used to obtain tribology data. Measurements were taken using tribopair surfaces of a polished ¾ inch steel ball loaded onto a silicone elastomer disk (diameter: 46 mm, thickness: 4 mm). For all tests, both contact surfaces were sonically cleaned (Branson 2210 Ultrasonic cleaner, Branson Ultrasonics Corporation, Danbury, USA) in ethanol for 5 minutes, followed by distilled water for 5 minutes, and then air-dried. A new elastomer disk was used for each experiment. 40 ml of sample was used and temperature was fixed at 37 °C, again simulating oral processing temperature.

Friction coefficient (μ) was used as a representation of an emulsions lubrication capacity. μ was measured across 20 averaged entrainment speeds (U) logarithmically set between 1 to

1000 mm/s, using an applied force of 0.5 N and a slide-roll ratio (SRR) of 50% to mimic the complex sliding and rolling friction associated with oral processing conditions.

Stribeck curves (μ versus U) were obtained in 6 consecutive sweeps, alternating between ascending and descending sweeps, in order to map the emulsion under hydrodynamic, mixed and boundary lubrication regimes. For each sample, experiments were performed in triplicate (using a new sample each experiment). Mean values were generated from the 18 sweeps and standard deviation from the three mean values.

4.3.7. Data Analysis

Data and statistical analysis were carried out using IBM SPSS Statistic (SPSS, SPSS Inc., Chicago, US). The effect of emulsion type on sensory perception was analysed via general linear model repeated-measures ANOVA. Test-within subject's sphericity assumed significance was taken at 95% confidence interval and degrees of freedom and P values are presented. If P value was considered significant ($p < 0.05$), a pairwise comparison post-hoc Bonferroni test was performed to reveal the nature of the differences. Replicate samples which significantly differed in perception were excluded from post hoc Bonferroni tests. To assess relationships between iso-viscous and iso-frictional emulsions and sensory attributes, coefficient of determination (R^2) was performed. All correlations presented are linear. The strength of the correlation was characterised so that 0 - 0.19 = no correlation, 0.2 – 0.39 = weak correlation, 0.4 – 0.59 = moderate correlation, 0.6 – 0.79 = strong correlation and 0.8 - 1 = very strong correlation. Principal component analysis (PCA) was performed, via XLSTAT v.2015 (Addinsoft, Paris, France), on the sensory data from the iso-viscous and iso-frictional emulsions in order to give an overview of the magnitude in which viscosity or friction influence perception and where and to what degree these physical properties combine to influence perception.

4.4. Results and discussion

4.4.1. Emulsion characterisation

4.4.1.1. Physical characterisation

To assess the contribution of both viscosity and friction in sensory perception, oil-in-water emulsions varying in droplet size were successfully designed in order to either be iso-viscous (at a shear rate of 50 s^{-1}) or iso-frictional (at an entrainment speed of 45 mm/s) (see Fig. 4.1 a and b).

There was no significant difference in viscosity for iso-viscous emulsions ($p > 0.05$), but iso-viscous emulsion friction coefficient significantly differed with droplet size ($p < 0.001$), with significant differences being between the emulsions with 0.1 µm droplets and emulsions with 8 µm ($p = 0.018$) and 20 µm droplets ($p = 0.012$) and between emulsions with droplet sizes of 55 µm and 8 µm ($p = 0.047$). Iso-friction emulsions did not significantly differ in friction coefficient ($p > 0.05$) but the viscosities of these emulsions significantly differed as a function of droplet size ($p < 0.001$), with significant differences between the emulsion with 0.1 µm droplets emulsions with 8 µm ($p = 0.002$) and 20 µm ($p = 0.002$), and between the emulsion with 55 µm droplets and those with 8 µm ($p = 0.004$) and 20 µm ($p = 0.007$) droplets. Across all emulsion samples viscosity was successfully matched at 30 mPas (see Fig. 4.1a), a similar viscosity to cream (Vega & Mercade-Prieto, 2011), and so to was friction coefficient at 0.14 μ (see Fig. 4.1b). This viscosity and friction coefficient was chosen as it ensured that all iso-viscous and iso-frictional samples contained a sufficient percentage of guar gum, as not to be considerably different in formulation to each other. Additionally, this produced 1) iso-viscous samples which were significantly different in viscosity from iso-frictional emulsions, and 2) iso-viscous samples which had a median viscosity value between viscosities of iso-friction emulsions (see Fig. 4.1a and b). Viscosity profiles of iso-friction emulsions (See Fig. 4.2a) and Stribeck curves of iso-viscous emulsions (See Fig 4.2b) have been provided, as an example of viscosity and friction behaviour differences between samples of the same iso-criterion. To the best of our knowledge this is the first paper to successfully design iso-frictional emulsions, and use systems that separate viscosity and friction cleanly for their assessment in terms of sensory perception.

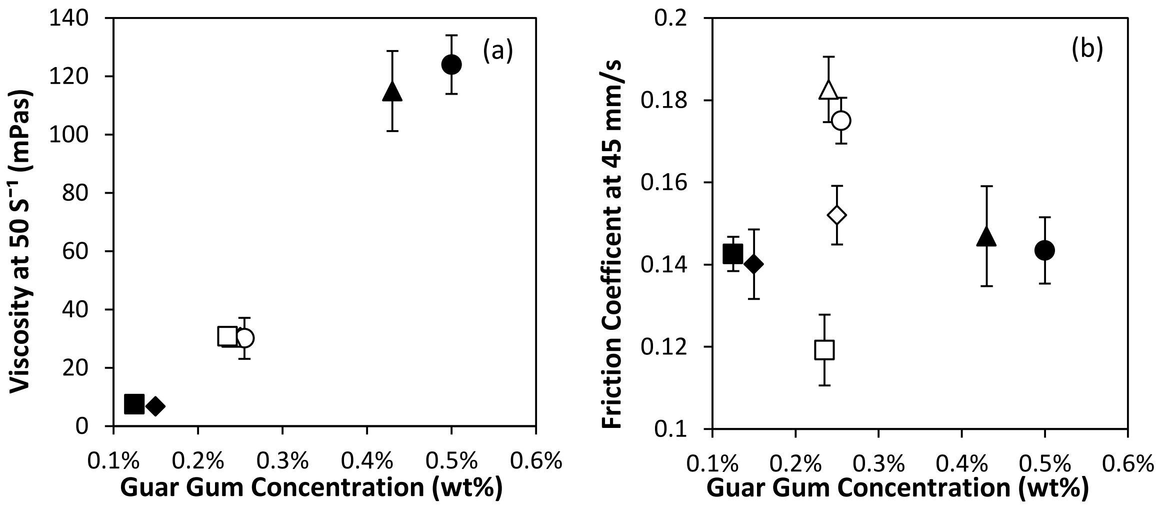
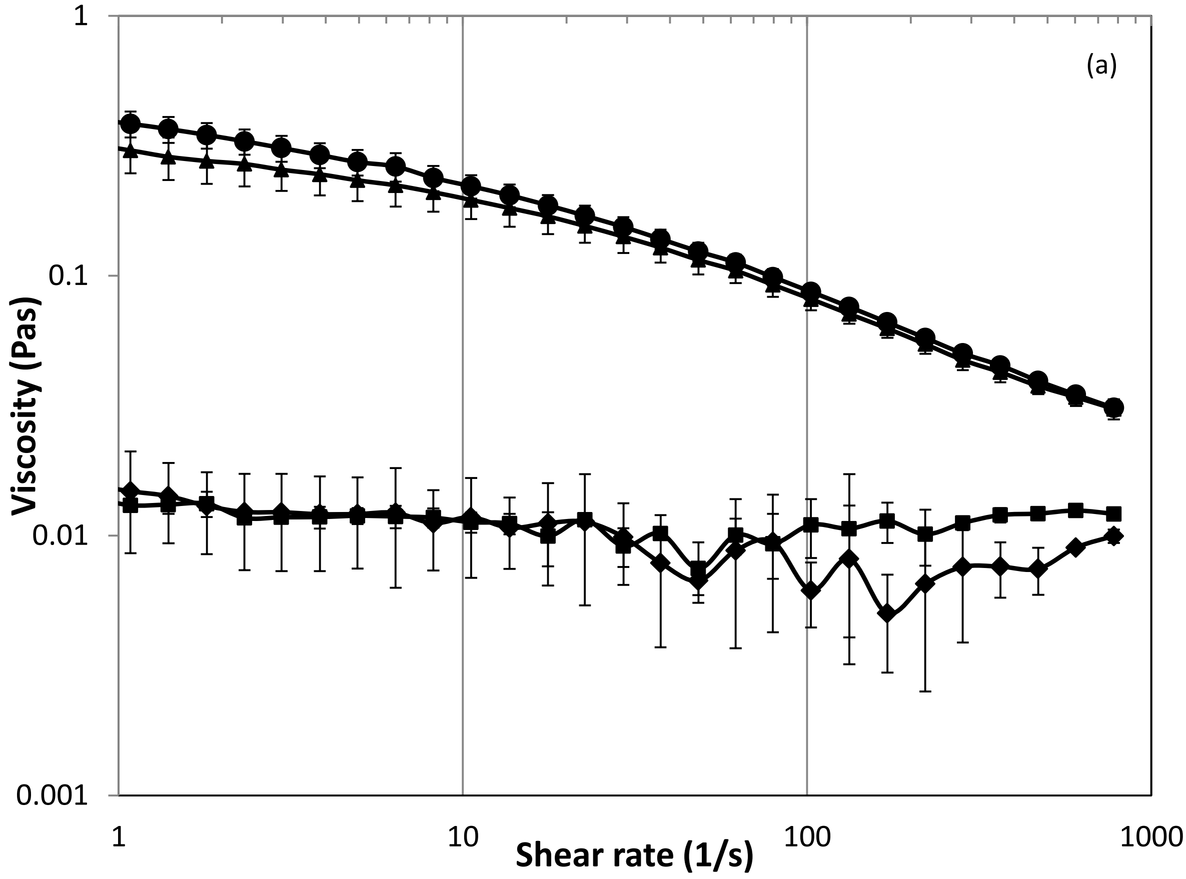


Fig. 4.1. Viscosity at 50 s^{-1} (a) and Friction coefficient at 45 mm/s (b) for Iso-viscous emulsions (0.1 µm (◇), 8 µm (○), 20 µm (△) and 55 µm (□)) and Iso-friction emulsions (0.1 µm (◆), 8 µm (●), 20 µm (▲) and 55 µm (■)). Data shown is mean ± standard deviation of triplicate measurements, (Decorrelation = R^2:0.01).

Within both sets of emulsion types, reproducible monomodal size distributions, at each droplet size, were shown. Mean emulsion droplet sizes ($d_{4,3}$) of iso-viscous emulsions were 0.2 ± 0.03, 7 ± 0.2, 21 ± 0.1 and 57 ± 1 and iso-frictional emulsions were 0.2 ± 0.02, 9 ± 2, 20 ± 0.2 and 56 ± 1. In all subsequent sections of this chapter, droplet sizes will be referred to as 0.1, 8, 20 and 55 µm.

The achievement of similar droplet size distributions for the two different emulsion sets can be attributed to the technique used to form the emulsion samples. As both iso-viscous and iso-frictional samples of the same droplet size were produced from the same initial emulsion before guar gum was added, no significant difference in droplet size between the emulsion types ($p > 0.05$) highlights that the addition of guar gum, and the technique used to disperse it, was mild enough not to induce significant droplet break-up or coalescence.



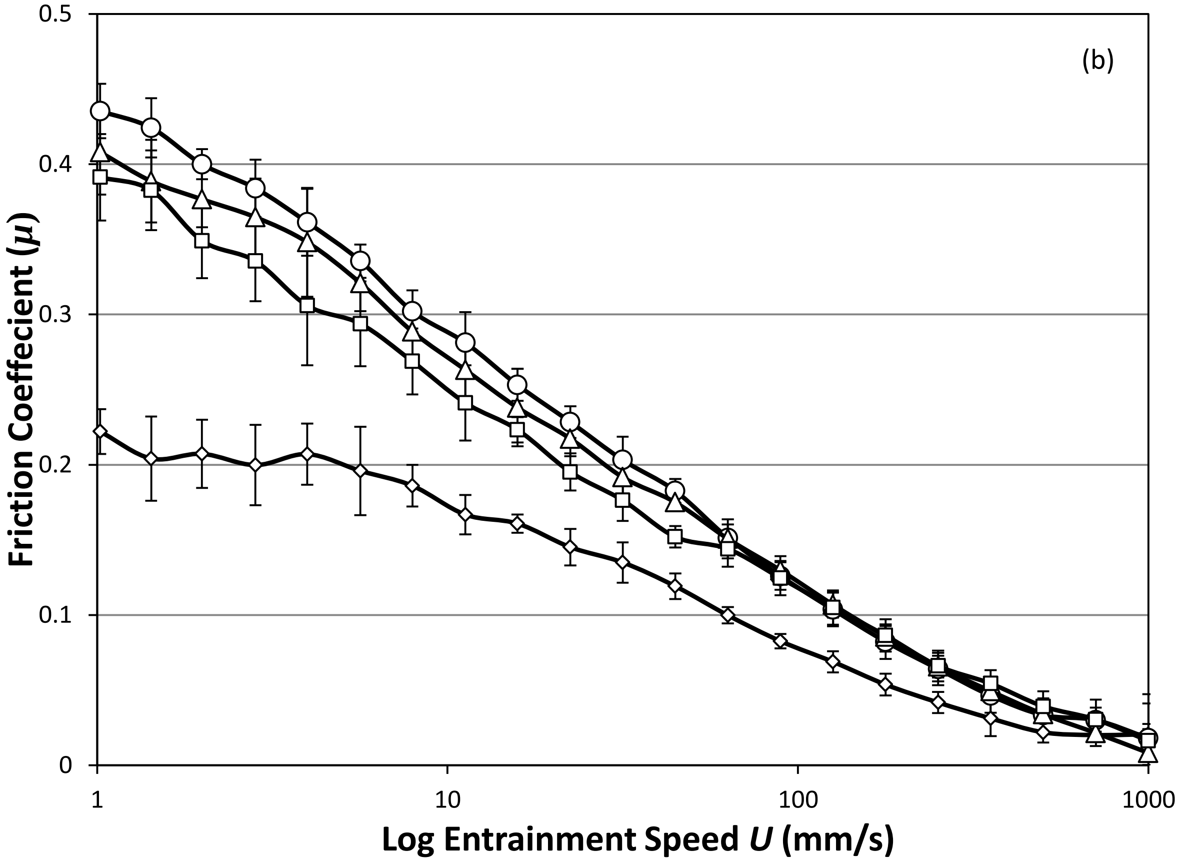


Fig. 4.2. (A) Viscosity profile of and Iso-friction emulsions (0.1 µm (◆), 8 µm (●), 20 µm (▲) and 55 µm (■)). (B) Stribeck curves at 0.5 N of Iso-viscous emulsions (0.1 µm (◇), 8 µm (○), 20 µm (△) and 55 µm (□)). Data shown is mean ± standard deviation of triplicate measurements. Provided as examples of behaviour differences between samples of the same iso-criterion.

4.4.1.2. Sensory characterisation

Significant differences in the perception of sensory attributes between the iso-viscous and iso-friction emulsions, oil droplet sizes and replicate samples were shown, with a group type*oil droplet size interactions also shown (See Table 4.2).

Table 4.2 ANOVA results for each of the sensory attributes investigated attributes used during measurements of sensory perception with the description discussed verbally with participants.

	Group (iso-viscous vs. iso-frictional samples)	**Oil Droplet Size**	**Group*Oil Droplet Size interaction**	**Replicate Samples**	**Group* Replicate Interaction**	**Oil Droplet Size * Replicate Interaction**	**Group* Droplet Size * Replicate Interaction**
Thickness	$p = 0.02$* F (1, 1) = 6.79	$p < 0.001$* F (1, 3) = 12.42	$p < 0.001$* F (1, 3) = 11.87	$p > 0.05$ F (1, 1) = 2.82	$p > 0.05$ F (1, 1) = 1.86	$p > 0.05$ F (1, 3) = 0.31	$p > 0.05$ F (1, 3) = 2.82
Smoothness	$p > 0.05$ F (1, 1) = 1.39	$p = 0.001$* F (1, 3) = 7.14	$p > 0.05$ F (1, 3) = 0.25	$p = 0.034$* F (1, 1) = 5.43	$p > 0.05$ F (1, 1) = 0.55	$p > 0.05$ F (1, 3) = 0.38	$p > 0.05$ F (1, 3) = 0.84
Slipperiness	$p > 0.05$ F (1, 1) = 1.07	$p > 0.05$ F (1, 3) = 0.40	$p > 0.05$ F (1, 3) = 1.58	$p > 0.05$ F (1, 1) = 5.74	$p > 0.05$ F (1, 1) = 0.63	$p > 0.05$ F (1, 3) = 0.838	$p > 0.05$ F (1, 3) = 1.64
Creamy mouthfeel	$p > 0.05$ F (1, 1) = 0.436	$p < 0.001$* F (1, 3) = 16.43	$p = 0.019$* F (1, 3) = 3.65	$p > 0.05$ F (1, 1) = 0.03	$p > 0.05$ F (1, 1) = 0.78	$p > 0.05$ F (1, 3) = 1.96	$p > 0.05$ F (1, 3) = 0.78
Creaminess	$p > 0.05$ F (1, 1) = 0.03	$P < 0.001$* F (1, 3) = 12.19	$p = 0.025$* F (1, 3) = 3.41	$p > 0.05$ F (1, 1) = 0.83	$p > 0.05$ F (1, 1) = 1.0	$p > 0.05$ F (1, 3) = 1.72	$p > 0.05$ F (1, 3) = 1.79

* Significant at $p < 0.05$

After the significant differences were identified using ANOVA analysis (see Table 4.2), Bonferroni post-hoc test identified where the significant differences occurred. Post hoc analysis for each of the significant attributes (i.e. Thickness, Smoothness, Creamy Mouthfeel and Creaminess) will be discussed in turn below. Slipperiness perception intensity did not significantly differ under any condition.

4.4.1.2.1 Thickness

Post hoc analysis determined that for Thickness perception there were significant differences between a number of the iso-frictional emulsions (i.e. that varied in viscosity) dependent on oil droplet size: the emulsion with 0.1 μm sized droplets was significantly different in Thickness perception from both the 8 μm sized emulsion ($p < 0.001$) and the 20 μm sized emulsion ($p < 0.001$); the emulsion with 55 μm droplets was significantly different from both the 8 μm sized emulsion ($p < 0.001$) and the 20 μm emulsion ($p < 0.001$). There were no significant differences between samples within the iso-viscous group.

There were also significant differences in Thickness perception between emulsions of the same droplet size, but different sample type (i.e. between iso-viscous and iso-frictional groups): 8 μm ($p < 0.001$), 20 μm ($p = 0.001$) and 55 μm ($p = 0.018$).

There were additional significant differences in Thickness perception as a function of a group type (i.e. iso-viscous or iso-frictional) droplet size interaction. The emulsion with 0.1 μm sized droplets from the iso-viscous grouping was significantly different from the iso-frictional emulsions with 8 μm droplets ($p < 0.001$), 20 μm droplets ($p = 0.002$), and 55 μm droplets ($p = 0.002$). The iso-viscous emulsion with 8 μm droplets was also significantly different from the iso-frictional emulsion with 20 μm droplets ($p < 0.001$). The emulsion with 20 μm sized droplets from the iso-viscous grouping was significantly different from the iso-frictional emulsions with 0.1 μm droplets ($p = 0.041$), 8 μm droplets ($p < 0.001$), and 55 μm droplets ($p = 0.002$). Finally, the iso-viscous emulsion with 55 μm droplets was significantly different from the iso-frictional emulsions with 8 μm droplets ($p < 0.001$) and 20 μm droplets ($p < 0.001$).

4.4.1.2.2 Smoothness

There was significant difference in Smoothness perception between replicates of iso-viscous 0.1 μm and 8 μm samples ($p = 0.031$ and $p = 0.032$, respectively) as such they were not used within post-hoc analysis. Apart from these samples, for Smoothness perception post hoc analysis determined that there were significant differences as a result of droplet size, whereby the emulsion with 0.1 μm droplets from the iso-frictional group was significantly different from both iso-frictional emulsions with 20 μm ($p = 0.001$) and 55 μm droplets ($p = 0.006$), and the emulsion with 8 μm droplets from the iso-frictional group was significantly different from the iso-frictional emulsion with 20 μm droplets ($p = 0.038$). There were no significant differences between samples, due to oil droplet size, within the iso-viscous group.

4.4.1.2.3 Creamy Mouthfeel

Droplet size also had an effect on Creamy Mouthfeel perception, with significant differences in both iso-viscous emulsions and iso-friction emulsions. For the iso-viscous emulsions there were significant differences between the emulsion with 0.1 μm droplets and both the emulsion with 20 μm droplets ($p = 0.04$), and the emulsion with 55 μm droplets ($p = 0.001$), and between the 8 μm sized emulsion and the 55 μm sized emulsion ($p = 0.022$). For the iso-frictional emulsions, there were significant differences in Creamy Mouthfeel perception between emulsions with 0.1 μm droplets and both emulsions with 8 μm droplets ($p = 0.005$) and 55 μm droplets ($P < 0.001$), between the emulsion with 8 μm droplets and the both emulsions with 20 μm droplets ($p = 0.006$) and with 55 μm droplets ($p < 0.001$), and finally between the 20 μm emulsion and the 55 μm emulsion ($p < 0.001$).

Again, there were significant differences as a function of a group type (i.e. iso-viscous or iso-friction) droplet size interaction, with differences between the iso-viscous emulsion with 0.1 μm droplets and the iso-frictional emulsion with 55 μm droplets ($p < 0.001$), between the iso-viscous emulsion with 8 μm droplets and iso-frictional emulsions with 55 μm droplets ($p < 0.001$), between the iso-viscous emulsion with 20 μm droplets and both the iso-frictional emulsion with 8 μm droplets ($p = 0.001$) and the iso-friction emulsion with 55 μm droplets ($p = 0.001$), and finally between the iso-viscous emulsion with 55 μm droplets and iso-friction emulsions with droplet sizes of 8 μm ($P < 0.001$) and 20 μm ($p = 0.009$).

4.4.1.2.4 Creaminess

Post hoc analysis determined that for Creaminess perception there were significant differences in perception as a function of droplet size. For emulsions within the iso-viscous group there were differences between the emulsions with 0.1 μm droplets and both the emulsion with 20 μm droplets ($p = 0.047$), and the emulsion with 55 μm droplets ($p = 0.001$), and between the emulsions with 8 μm droplets and 55 μm droplets ($p = 0.021$). For emulsions within the iso-frictional group there were significant differences between the emulsion with 0.1 μm droplets and both the emulsion with 8 μm droplets ($p = 0.001$) and the emulsion with 55 μm droplets ($p = 0.006$), as well as between the emulsion with 8 μm droplets and both 20 μm droplets ($p = 0.006$) and 55 μm droplets ($p < 0.001$), and finally between the emulsions with 20 and 55 μm droplets ($p = 0.001$).

There were also significant differences as a function of a group type (i.e. iso-viscous or iso-friction) and droplet size interaction. There were significant differences between the iso-viscous emulsion with 0.1 μm droplets and iso-friction emulsions with both 8 μm droplets ($p = 0.02$) and 55 μm droplets ($p < 0.001$), between the iso-viscous emulsion with 8 μm droplets and the iso-frictional emulsion with 55 μm droplets ($p < 0.001$), between the iso-viscous emulsion with 20 μm droplets and iso-friction emulsions with 8 μm ($p < 0.001$) and 55 μm ($p < 0.001$) droplets, and finally between the iso-viscous emulsion with 55 μm droplets and iso-friction emulsions with 8 μm ($p < 0.001$) and 20 μm ($p = 0.039$) droplets.

4.4.1.2.4 Sensory result considerations

As previous observations (Frost & Janhoj, 2007; Lett et al., 2016), have assumed that Creaminess and Creamy Mouthfeel are considered as the same attribute by participants, we correlated scores of these attributes. Creaminess correlated very strongly with Creamy Mouthfeel for both sets of emulsions (iso-viscous R^2:0.99; iso-frictional R^2:0.98). Additionally, for both attributes, similar significance is shown in all conditions (see Table 4.2). Given the strength of this correlation, and previous observations in systems with a similar protocol, participant group and droplet sizes investigated (Lett et al., 2016), we conclude Creaminess and Creamy Mouthfeel were assessed as the same attribute. Nevertheless, this observation highlights that Creaminess was predominantly influenced by textural cues, a conclusion also reached by other authors using liquid systems (Frost and Janhoj, 2007). In all subsequent sections of this chapter, only the results of Creaminess perception will be discussed.

As replicate samples in Smoothness perception significantly differed, they were omitted from post-hoc analysis. Data for iso-viscous 0.1 μm emulsion and 8 μm emulsion for Smoothness perception was therefore unavailable for full comparison; as such, they are not included in discussions (see Fig. 4.3b, 4.4b, 4.5b, section 4.4.2.2 and 4.4.3.2).

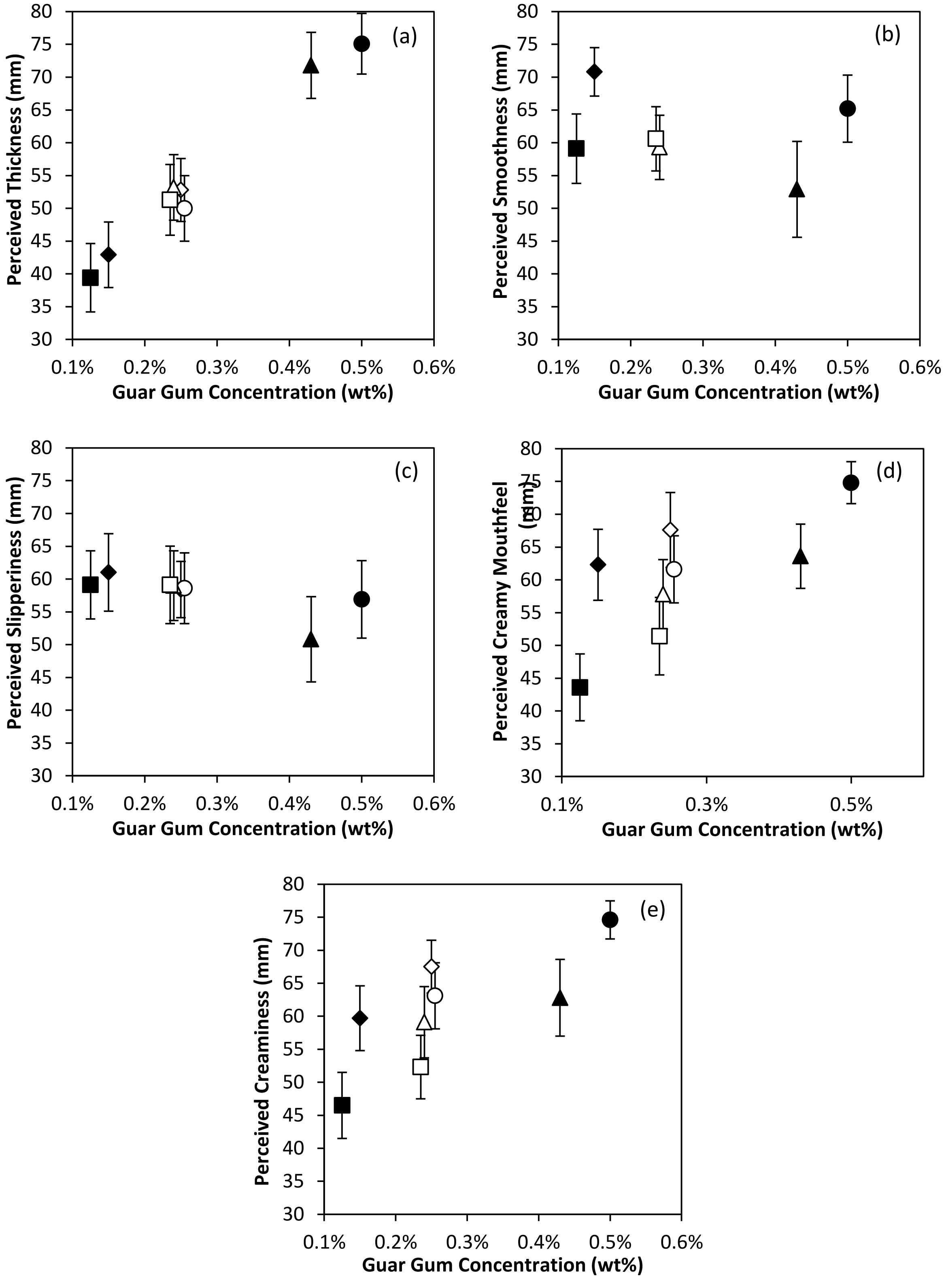


Fig. 4.3. Mean ratings of Thickness (a), Smoothness (b), Slipperiness (c), Creamy mouthfeel (d) and Creaminess (e) for Iso-viscous emulsions (0.1 µm (◇), 8 µm (○), 20 µm (△) and 55 µm (□)) and Iso-friction emulsions (0.1 µm (◆), 8 µm (●), 20 µm (▲) and 55 µm (■)) as a function of guar gum concentration. Error bars representative of one standard error of the mean.

4.4.2. Viscosity influences in sensory perception

4.4.2.1. Thickness

Iso-viscous emulsions were not significantly different in Thickness perception, with significant differences in Thickness perception only being shown when viscosity was a variable (i.e. for iso-friction emulsions and group * droplet size interactions (See Table 4.2)). The relationship demonstrated between Thickness perception and viscosity is linear and positive (R^2 = 0.96) (see Fig. 4.4a). Thickness perception is shown here to be a sensory interpretation of viscosity, a relationship that is commonly described in the literature (Cutler, Morris & Taylor, 1983; Kokini, Kadane & Cussler, 1977; Morris & Taylor, 1982; Morris, Richardson & Taylor, 1984; Shama & Sherman, 1973a; Shama & Sherman, 1973b). This relationship is independent of droplet size, and dependent on guar gum concentration (R^2 = 0.98), which influences overall emulsion viscosity (see Fig. 4.3a).

4.4.2.2. *Smoothness*

Recent studies that use advanced sensory techniques such as Temporal Dominance Sensation, which captures the intensity of textural attributes over the duration of oral processing (Lenfant *et al.*, 2009), highlight that during oral processing initially rheological properties dominate in terms of influencing sensory perception, then with time frictional properties dominate (Chen & Stokes, 2012; Stokes, Boehm & Baier, 2013). These techniques support instrumental conclusions that perception of sensory attributes are dependent on both the rheological and surface related properties of the food bolus (de Wijk & Prinz, 2005; Kokini, 1987). It highlights why attributes such as Smoothness have been difficult attributes to characterise both instrumentally and in terms of sensory perception (Chen & Stokes, 2012). Practical restraints in this study meant that oral processing time was not controlled. As such, the time point of Smoothness assessment was at the participant’s discretion and consequently may highlight why iso-viscous 0.1 μm and 8 μm replicate samples significantly differed in Smoothness (see section 4.4.1.2.2).

As liquids have lower oral residence time than semi-solid or solid foods (Ertekin and Aydogdu, 2003), and consequently lower sensory exposure time (Hogenkamp *et al.,* 2011), the transition from rheological to thin-film influences on perception may have been less defined perceptually. Droplet size in the iso-frictional emulsions significantly influenced Smoothness perception (see Table 4.2). Differences in perception intensity between emulsions with droplet sizes of 0.1 μm and 55 μm and 8 μm and 20 μm respectively, indicate that in both relationships, they are independent of viscosity and frictional influences, as at the investigated shear rate and traction speed both viscosity and friction are comparable (See Fig. 4.1a and b). This observation highlights that droplet size is important, with smaller droplets increasing Smoothness perception (see Fig. 3b, 4b, 5b). Emulsions with smaller droplets have a greater number of droplets with lower deformability and break up (see section 3.4.3). However, how the physical characteristics of emulsions affect both oral processing and texture perception would be of interest for further investigation.

4.4.2.3. *Creaminess*

A moderate positive linear correlation between viscosity and Creaminess perception was shown (R^2:0.43) (see Fig. 4.4e). The relationship observed highlights two interesting behaviours. Firstly, when viscosity is varied, as seen with iso-friction emulsions, Creaminess is largely dependent on viscosity (R^2:0.64). This agrees with previous studies that conclude that viscosity is the predominant variable that influences Creaminess perception (Akhtar *et al.,* 2005; Frost & Janhoj, 2007; Kilcast & Clegg, 2002). This explains why Thickness is considered an attribute which contributes to the multitude of sensory influences which amalgamate to form Creaminess (Kokini and Cussler, 1984; Kokini *et al.,* 1984). Secondly, when viscosity is constant, as seen with iso-viscous emulsions, Creaminess perception varies with oil droplet size, with emulsions with smaller oil droplets always being perceived as creamier (see Fig. 3e, 4e, 5e) (discussed further in section 4.4.3.3). The second behaviour described is also observed in iso-friction emulsions, when comparing 1 μm and 55 μm samples and 8 μm and 20 μm samples, where friction is the same and viscosity is coincidentally similar in both relationships (Fig. 4.1a and b).

This highlights an alternative behaviour that influences perception, which is a function of oil droplet size. This alternative "behaviour" is undetectable through the instrumental methods used with our study, and thus must be initiated by chemical or mechanical factors occurring during oral processing. A possibility may be the difference in the flocculation behaviour of protein stabilised oil droplets when in the presence of saliva (Vingerhoeds *et al.,* 2005; Vingerhoeds *et al.,* 2009), which could be an area for future investigation.

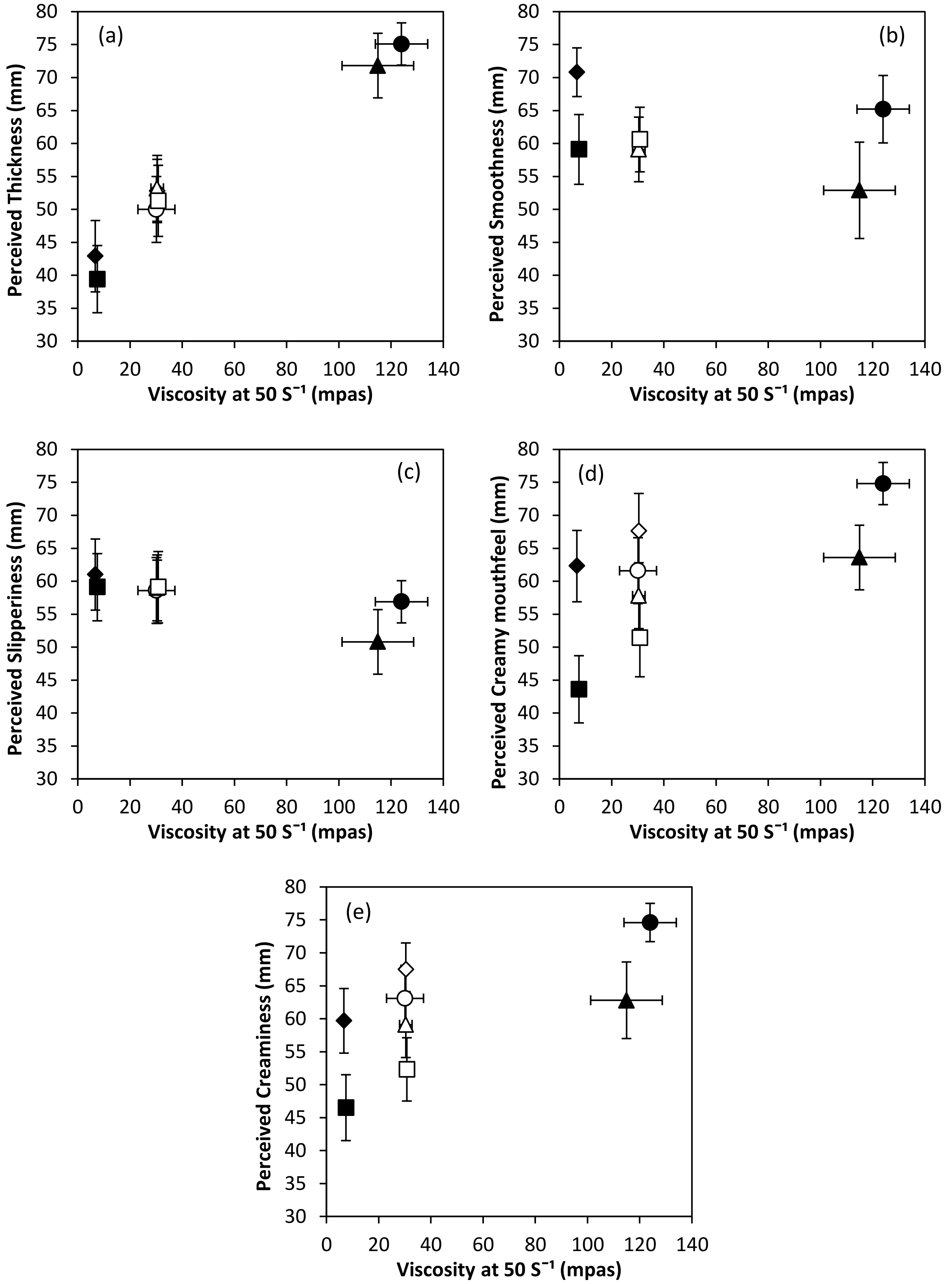


Fig. 4.4 Mean ratings of Thickness (a), Smoothness (b), Slipperiness (c), Creamy mouthfeel (d) and Creaminess (e) for Iso-viscous emulsions (0.1 µm (◇), 8 µm (○), 20 µm (△) and 55 µm (□)) and Iso-friction emulsions (0.1 µm (◆), 8 µm (●), 20 µm (▲) and 55 µm (■)) as a function of viscosity at 50 S^{-1}. Y-axis error bars represent one standard error of the mean. X-axis error bars represent one standard deviation of the mean.

4.4.3. Frictional influences in sensory perception

4.4.3.1. Thickness

Varying friction (i.e. for the iso-viscous emulsions) did not affect the perception of Thickness ($R^2 = 0.02$) (see Fig. 4.5a), reinforcing previous understanding that Thickness perception is mediated by viscosity (see section 4.4.2.1), and independent of friction.

4.4.3.2. Smoothness

Smoothness perception is not thought to be directly influenced by viscosity, but instead is thought to correlate with the reciprocal of the friction force between surfaces (Kokini, 1987). As such, it would be expected that iso-viscous emulsions would highlight frictional influences in Smoothness perception, however this is not shown (see Fig. 4.5b). Interestingly, differences in Smoothness perception only occurred for the iso-frictional emulsions, where viscosity was varied (see Fig. 4.4b). However, it is thought that this effect is in fact independent of viscosity, and a result of behaviour of the droplets during oral processing (see section 4.4.2.2).

Two key things should be considered before interpreting the Smoothness results. Firstly, oral behaviour was not controlled. Kokini (1987) concluded that “Smoothness is the reciprocal of the friction force between surfaces”, and controlled oral behaviour by instructing participants to move their tongue lightly across the food being judged. When a liquid food is being consumed, this controlled oral behaviour would promote a lubricated contact (i.e. a thin-film) between the tongue and the roof of the mouth. At this point during oral processing the sensation of “Smoothness” was assessed. Other studies have also manipulated oral behaviour in a similar manner to investigate Smoothness (Pascua, Koc and Foegeding, 2013; van Aken, Vingerhoeds and de Hoog, 2007). Within the current study, oral behaviour was at the participant’s discretion and therefore is representative of natural oral behaviour. It can therefore be presumed that due to the typical rapid oral processing behaviour of liquid foods, samples were only present within the mouth for around 2 seconds (Ertekin and Aydogdu, 2003), and would of only be present in a thin-film for a short period before swallowing. The degree of frictional influence on perception would be expected to be considerable less than observed by Kokini (1987), especially if participants assessed the samples at initial oral-sample contact, where viscosity influences are dominant (Chen & Stokes, 2012; Stokes, Boehm & Baier, 2013). Secondly, two (0.1 μm and 8 μm) of the four iso-viscous emulsions were omitted from post-hoc analysis, reducing the extent to which we can discuss frictional influences on Smoothness.

4.4.3.3. Creaminess

Only a weak positive linear correlation between friction coefficient and Creaminess perception was shown (R^2:0.27) (see Fig. 4e). However, as highlighted in Section 4.4.2.3, when viscosity is constant Creaminess perception varied as a function of oil droplet size, with smaller oil droplets increasing Creaminess (R^2:0.95) (see Fig. 3e, 4e, 5e). This relationship has been previously observed in model emulsion systems with a similar droplet size range (Lett et al., 2016). Such as with Smoothness perception, it seems sensible to suggest the behaviour of oil droplets during oral processing is affecting Creaminess perception, especially as Smoothness is thought to be involved in Creaminess perception (Frost and Janhoj, 2007; Kokini, 1987). We have already highlighted possible droplet destabilisation mechanisms, which could be occurring during oral processing and affecting perception. For example, emulsions that coalescence have been shown to increase Creaminess perception (Chojnicka-Paszun, de Jongh and de Kruif, 2012; Dresselhuis *et al.,* 2008); the relationship was shown using tribology, where greater coalescence on tribo-surfaces (where it is assumed that a similar relationship would be observed in the mouth) lowered friction, and as such would increase Creaminess.

Coalescences under quiescent conditions is dependent on the rheology of the continuous phase, the aggregation state of the droplets and the droplet-size distribution (Dickinson, 1992). For the iso-viscous emulsions viscosity of the continuous phase was constant, and aggregation was not measured. Although the effect of droplet size on coalescence is complex, decreasing droplet size increases collision frequency, which can increase the rate of coalescence (McClements, 1998). This would explain why Creaminess perception was a function of droplet size; however, oral processing conditions also need to be taken into account.

For an emulsion of this nature, under quiescent conditions coalescence rate would be extremely slow; however, the application of force and shear (i.e. as experienced during oral processing) would increases coalescence rate (McClements, 1998). Additionally, for these protein stabilised emulsions the presence of saliva would facilitate droplet flocculation (Vingerhoeds *et al.*, 2005; Vingerhoeds *et al.*, 2009), which would increase coalescence rate through increasing droplet contact. However, due to the extremely short oral residence time of fluids within the mouth (Ertekin and Aydogdu, 2003) it seems unlikely that coalescence would be able to occur at a sufficient rate to significantly affect perception. As such, it seems sensible to suggest flocculation, instead of coalescence, is the primary droplet behaviour facilitating the change in Creaminess perception. Flocculation would occur almost instantaneously under the chemical conditions of the mouth (Vingerhoeds *et al.*, 2005), in comparison to the much slower rate of coalescence (McClements, 1998). Emulsions containing smaller droplets, would possess 1) a greater surface area available for floc formation, increasing the number of flocs present and 2) greater droplet integrity (see section 3.4.3), reducing the likelihood of coalescence, in relation to larger droplet, when within a floc. Droplet flocculation affects perception as it affects properties such as viscosity (Vingerhoeds *et al.*, 2009), which is recognised as dominant factor in Creaminess perception in this study (see section 4.4.2.3) and others (Akhtar *et al.*, 2005; Frost & Janhoj, 2007; Kilcast & Clegg, 2002; Mela, Langley, & Martin, 1994; Tarrega & Costell, 2006; De Wijk & Prinz, 2007). Therefore, if we discount any dilution by saliva, we can assume that orally processed emulsions had a higher viscosity than measured prior to oral processing due to saliva-induced flocculation, which in turn affected perception. In order to confirm this conclusion future studies should look to measure flocculation and viscosity following oral processing.

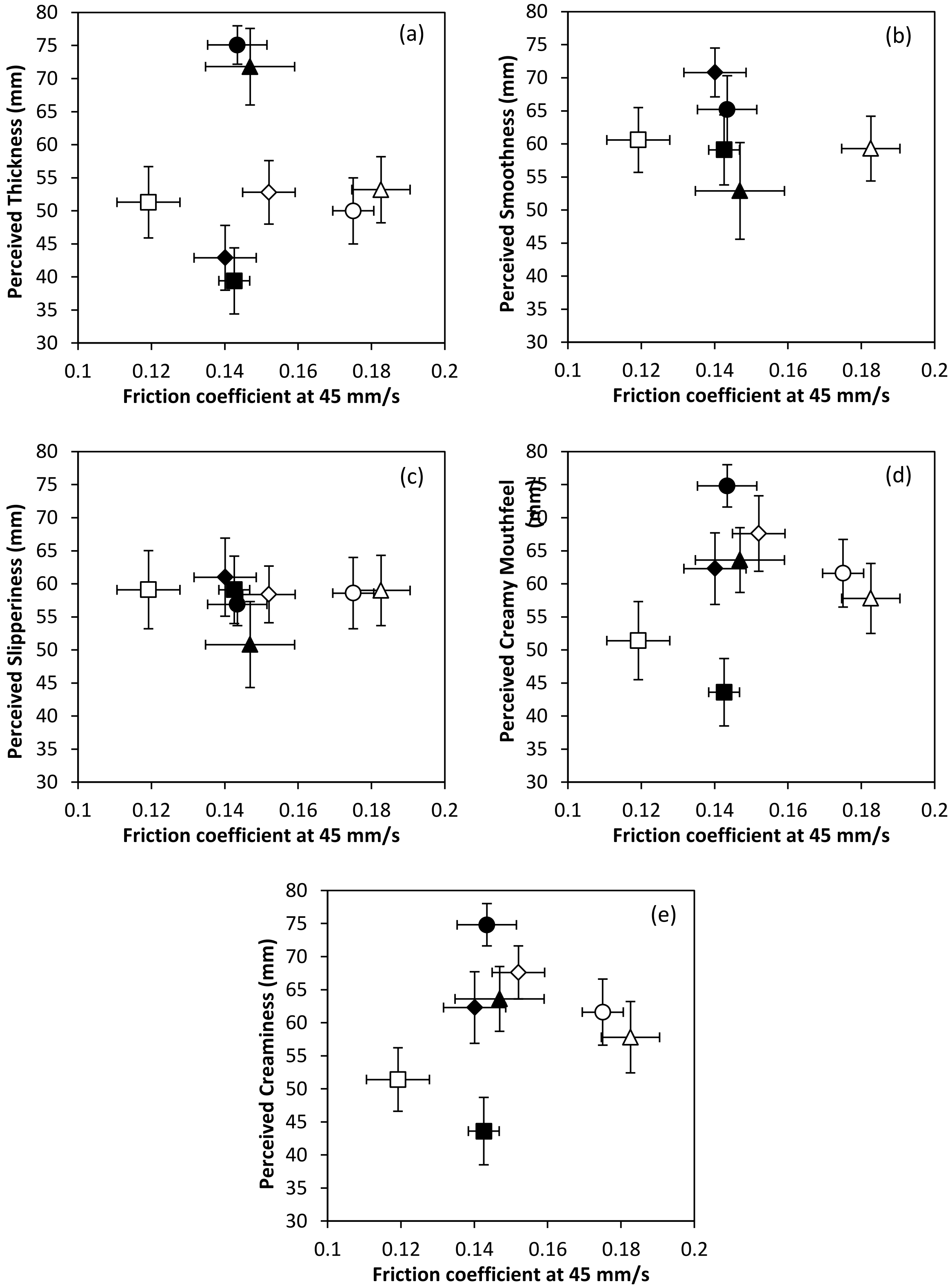


Fig. 4.5 Mean ratings of Thickness (a), Smoothness (b), Slipperiness (c), Creamy mouthfeel (d) and Creaminess (e) for Iso-viscous emulsions (0.1 µm (◇), 8 µm (○), 20 µm (△) and 55 µm (□)) and Iso-friction emulsions (0.1 µm (◆), 8 µm (●), 20 µm (▲) and 55 µm (■))as a function of friction coefficient at 45 mm/s. Y-axis error bars represent one standard error of the mean. X-axis error bars represent one standard deviation of the mean.

4.4.4. Combined influence of Friction and Viscosity in Sensory Perception

The sensory results for the iso-viscous and iso-frictional emulsions is summarised in a PCA bi-plot (Fig. 4.6). The plot highlights that the distribution of the emulsions across the sensory space is reflective of their viscosity (See Fig. 4.1) on the viscosity plane, but droplet size on the friction plane. This indicates that either differences in friction could not be perceived, or, and more probable, for the iso-viscous emulsions droplet behaviour during oral processing had an effect on perception as a function of droplet size (see Sections 4.4.2 and 4.4.3).

4.4.4.1. Thickness

Previous discussions (see Sections 4.4.2.1 and 4.4.3.1) have established that Thickness perception was related to viscosity, and independent of friction. This relationship is further reflected in the PCA bi-plot (Fig. 4.6), where Thickness is at the extreme of the viscosity plane, with samples that had the greatest viscosity (see Fig. 4.1), but distanced from the friction plane, highlighting the independence.

4.4.4.2. Smoothness

It was concluded that Smoothness is independent of viscosity and frictional influences at the investigated shear rate and traction speed (See Fig. 4.1a and b), and the observations were a result of droplet behaviour during oral processing; this is reflected in the PCA bi-plot where Smoothness sits distanced from both plane extremes.

4.4.4.3 Creaminess

Creaminess exists within the sensory space between the extremes of friction and viscosity (see Fig. 4.6). However, from analysis and discussion of the data, we know the physical variables involved within this study to be viscosity (see section 4.4.3.3) and droplet behaviour during oral processing, with smaller droplet increasing Creaminess perception (see section 4.4.2.3).

Interestingly, within this study, the sensory interpretations of these two physical properties shown to be involved in Creaminess are: 1) Thickness, a sensory interpretation of viscosity (see sections 4.4.2.1, 4.4.3.1, 4.4.4.1) and 2) Smoothness, dependent on droplet behaviour during oral processing as a function of droplet size. This conclusion reflects previous literature, that defined Creaminess as a combination of the two independent sensory variables ‘‘Thickness’’ and ‘‘Smoothness’’ (see Fig 4.6) (Kokini & Cussler, 1984; Kokini, 1987). This relationship is also reflected in the PCA bi-plot (see Fig. 4.6). This therefore indicates that a combination of physical and sensory influences are involved in Creaminess perception. Also, as flavour was a constant within this study these results reflect Creaminess is predominantly a textural attribute. It must be noted, droplet size has also been shown to effect flavour release (van Ruth, King and Giannouli, 2002), with implications on perceptions of Creaminess (Frost & Janhoj, 2007; Weenen, Jellema and de Wijk, 2005).

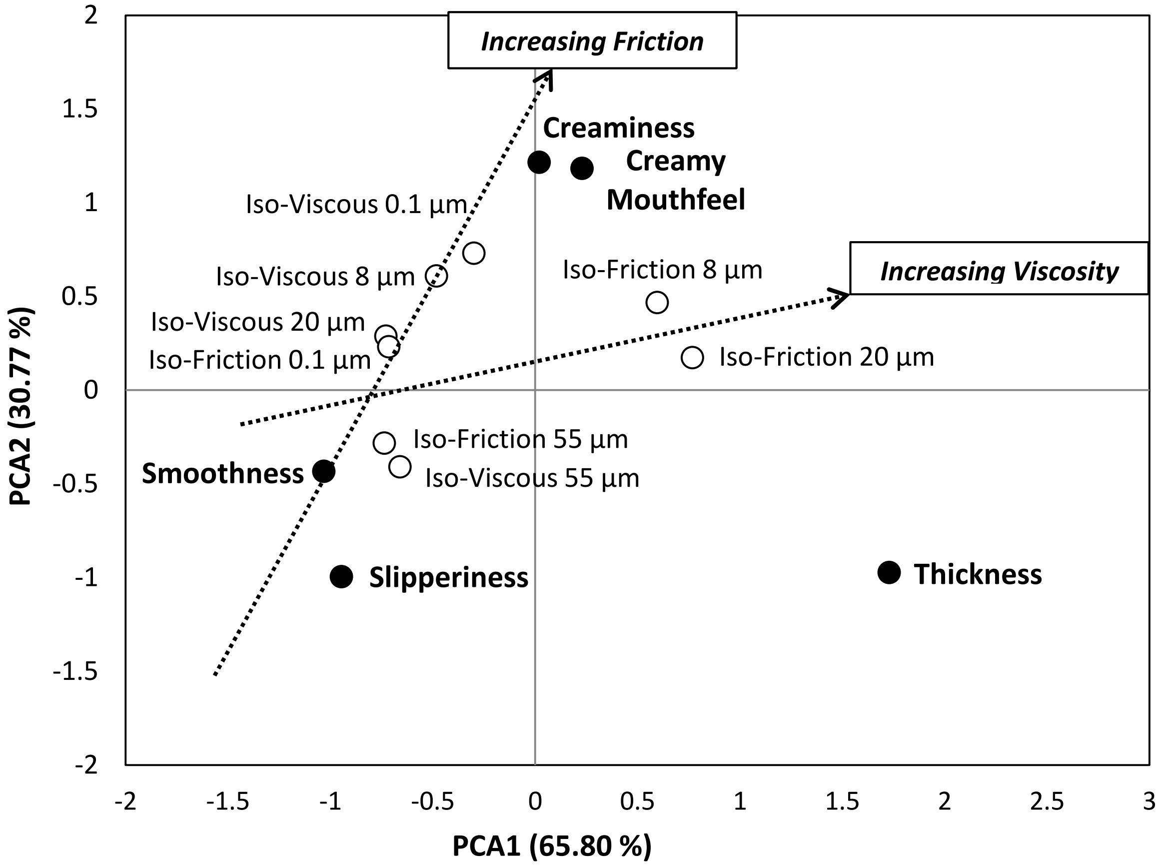


Fig.4.6 Principal component analysis (PCA) Bi-plot of sensory attributes in respect to viscosity and friction influences. Lines indicate predicted viscosity and friction planes based on physical data (see Fig. 4.1).

4.5. Conclusions

Using this novel approach, textural perception of emulsions has been shown to be dependent on both viscosity and droplet behaviour during oral processing. Viscosity was the dependent variable affecting Thickness perception. Friction did not affect the perception of any attribute, however this may be because any frictional differences between emulsions could not be perceived. However, perception differed in iso-viscous systems, where friction was varied. As such, it was concluded that droplet behaviour during oral processing affected perception, and was a function of droplet size. This was demonstrated in perception of Smoothness and Creaminess, and highlights that when viscosity is kept constant, behaviour of oil droplets during oral processing could be manipulated in order to change the perception of these attributes.

Furthermore, interpretation of the PCA bi-plot highlighted the combined influence of viscosity and droplet behaviour in Creaminess perception, which is thought to be a combination of Thickness and Smoothness, confirming conclusions made in the literature.

Chapter 5. Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour

Data and discussions contained within this chapter have been published within:

Lett, A.M., Yeomans, M.R., Norton, I.T. and Norton, J.E. (2016). Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour. *Food Quality and Preference, 47,* 148-155

5.1. Abstract

Food reformulation, either to reduce nutrient content or to enhance satiety, can negatively impact upon sensory characteristics and hedonic appeal, whilst altering satiety expectations. Within numerous food systems, perception of certain sensory attributes, known as satiety-relevant sensory cues, have been shown to play a role in food intake behaviour. Emulsions are a common food structure; their very nature encourages reformulation through structural design approaches. Manipulation of emulsion design has been shown to change perceptions of certain sensory attributes and hedonic appeal, but the role of emulsions in food intake behaviour is less clear. With previous research yet to identify emulsion designs which promote attributes that act as satiety-relevant sensory cues within emulsion based foods, this paper investigates the effect of oil droplet size ($d_{4,3}$: 0.2 - 50 µm) and flavour type (Vanilla, Cream and No flavour) on sensory perception, hedonics and expected food intake behaviour. By identifying these attributes, this approach will allow the use of emulsion design approaches to promote the sensory characteristics that act as satiety-relevant sensory cues and/or are related to hedonic appeal. Male participants (n =24) assessed the emulsions. Oil droplet size resulted in significant differences ($p < 0.05$) in ratings of Vanilla and Cream flavour intensity, Thickness, Smoothness, Creamy Mouthfeel, Creaminess, Liking, Expected Filling and Expected Hunger in 1 hours' time. Flavour type resulted in significant differences ($p < 0.05$) in ratings of Vanilla and Cream flavour intensity, Sweetness and Liking. The most substantial finding was that by decreasing oil droplet size, Creaminess perception significantly increased. This significantly increases hedonic appeal, in addition to increasing ratings of Expected Filling and decreased Expected Hunger in 1 hours' time, independently of energy content. If this finding is related to actual eating behaviour, a key target attribute will have been identified which can be manipulated through an emulsions droplet size, allowing the design of hedonically appropriate satiating foods.

5.2. Introduction

With the increasing prevalence of global obesity and its related non-communicable diseases, new strategies to promote weight loss and reduce the risk of weight gain are urgently needed. The food industry is increasingly being encouraged to contribute to the alleviation of the obesity burden through product reformulation and the development of the next generation of foods (Norton, Moore and Fryer, 2007). One approach involves increasing the satiating power of foods and beverages, reducing consumption quantity, and thus energy intake (Blundell, 2010; van Kleef *et al.*, 2012).

Sensory characteristics experienced during eating have been shown to impact upon consumption (de Graaf, 2012). Even subtle differences in sensory characteristics have an impact on eating behaviour (de Graaf and Kok, 2010; McCrickerd *et al.*, 2012; Yeomans and Chambers, 2011; Zijlstra *et al.*, 2009a; Zijlstra *et al.*, 2009b). This indicates that certain sensory characteristics, such as Thickness (Hogenkamp *et al.*, 2011; Mattes and Rothacker, 2001; McCrickerd *et al.*, 2012; Zijlstra *et al.*, 2009b), and the degree to which these are perceived during eating, act as satiety-relevant sensory cues, changing the food or beverages capacity to generate satiety expectations. Identifying satiety-relevant sensory cues and designing foods with these sensory attributes should increase their satiating power.

The mechanism by which satiety-relevant sensory cues appear to work suggests that people learn to associate the sensory characteristics of a food or beverage with the subsequent experience of satiety post-consumption (Brunstrom, Shakeshaft and Scott-Samuel, 2008; Yeomans *et al*., 2014). As such, on later occasions, when presented with the same product or a novel food or beverage with similar sensory characteristics, expectations are made about how satiating the food or drink will be based on its sensory profile. As a result of how these satiety expectations are generated, an indication of how a novel food or beverage may impact on actual satiety can be simply acquired by measuring the resulting satiety expectations generated after presenting a novel food.

Nonetheless, disadvantages of using satiety-relevant sensory cues as a reformulation or design approach have been highlighted: 1) as learned sensory cues are associated with a given caloric value and satiety expectation, producing low-energy dense foods with sensory characteristics (such as Thickness and Creaminess) indicative of a greater energy content, which is not delivered by the food, typically results in compensatory intake (Yeomans and Chambers, 2011); and 2) palatability has been shown to be inversely correlated to satiating power (Drewnowski, 1998; de Graaf, de Jong and Lambers, 1999; Holt *et al.,* 1995), a commercial disadvantage when we consider that hedonic appeal is a driver in consumer purchasing habits (Dhar and Wertenbroch, 2000).

If hedonic properties can be maintained, or even enhanced, an effective formulation or design approach would be to increase the satiating power of foods independently of energy content. Typically, energy dense foods associated with nutrients such as fat have a strong hedonic appeal (Prentice and Jebb, 2003). Within food systems, fat is often structured in the form of an emulsion. An emulsion is comprised of two immiscible liquids, the most common food emulsion being oil dispersed in water (e.g. mayonnaise, milk, dressings, creams), known as an oil-in-water emulsion.

Microstructural reformulation approaches have been shown to alter sensory characteristics and hedonics in model and applied emulsion food systems (Akhtar *et al.,* 2005; Akhtar, Murray and Dickinson, 2006; de Wijk and Prinz, 2005; Kilcast and Clegg, 2002;; Mela, Langley and Martin, 1994; Moore *et al.,* 1998; van Aken, Vingerhoeds and de Wijk, 2011; Vingerhoeds *et al.,* 2008). Subsequently, through the manipulation of microstructural properties, the capability to change the capacity to which satiety expectations are generated could be realised, through altering perception intensity of sensory characteristics that act as satiety-relevant cues.

We report: 1) how microstructural differences in emulsion based food systems change perceptions of sensory attributes; 2) sensory attributes that promote hedonic appeal; and 3) sensory attributes that act as satiety-relevant sensory cues, within emulsion systems. Taking a multidisciplinary approach, combining understanding of food engineering, sensory science, nutrition and food psychology, the work identifies the microstructural properties of emulsion food systems that promote individual sensory attributes and expected food intake behaviours. Most importantly, we aim to identify emulsion designs which may be used to maintain or enhance sensory and hedonic properties, but increase the satiating power of emulsion based foods.

5.3. Materials and methodology

5.3.1. Design and participants

The present study investigated the effect of oil droplet size and flavour type within model oil-in-water emulsions on the perception of sensory attributes, hedonics and expected food intake behaviours.

Male participants were recruited via advertisement and screened for food allergies, smoking habits, body mass index (BMI), current medical status and dietary habits (restricted eating) via Dutch eating behaviour questionnaire (DEBQ) (van Strien, *et al.*,1986). Females were excluded as they typically practice significantly higher levels of restricted eating and other eating behaviours than males (Wardle, 1987). The restricted eating DEBQ consisted of 10 questions having a five-option response format: never (1), seldom (2), sometimes (3), often (4), and very often (5). A restraint score was obtained by summing the scores for the 10 items and dividing by 10. A higher score indicates greater dietary restraint. Potential participants were prevented from participating if they indicated any food allergies, history of smoking, had a BMI above 24.9 Kg/M^2 or below 18.5 Kg/M^2, were taking medication known to interfere with sensory perception or food intake or had a DEBQ restricted eating score of >2.4 indicative of the participant occasionally or more often exercising restricted eating behaviour. 24 respondents met the study criteria and were included in the study. Participants were aged 18 - 26, with a mean BMI of 22.8 ± 1.7 Kg/m^2 and DEBQ restricted eating score of 1.8 ± 0.2. All participants gave written informed consent prior to participation. To guard against expectancy effects, the study was described as an investigation into the sensory analysis of emulsions. Ethical approval for the study was obtained from the University of Birmingham ethics committee.

5.3.2. Test Samples

Samples consisted of an oil-in-distilled water emulsion (1 wt.% sodium caseinate (Excellion EM7, DMV International, The Netherlands)), 2 wt.% sucrose (Silverspoon granulated, British Sugar Plc, UK)) and 15 wt.% sunflower oil (Tesco Plc, UK)) with one of three flavours dependent on flavour condition: 1 wt.% vanilla extract (Nielsen-Massey Vanillas International LLC, The Netherlands), 0.05 wt.% cream flavouring (Frontier Natural Products Co-op, USA) and No flavour.

Emulsions were produced using two different methods dependent upon the required mean droplet size of the emulsion being produced: a high shear mixer (Silverson L5M, Silverson machines Ltd, UK) or a high-pressure homogeniser (GEA Niro Soavi Panda Plus 2000, GEA Niro Soavi, Italy). In a 600ml beaker, 15 wt.% sunflower oil was added to 85 wt.% aqueous phase (1.1 wt.% NaCas, 2.2 wt.% sucrose, 96.6 wt.% distilled water solution). The whole sample was then emulsified for 5 minutes using the high shear mixer. Dependent on oil droplet size being produced the sample was subjected to a different rotational speed (rpm) and emulsor screen (fine (0.8 mm pores) or medium (1.6 mm pores)) (**50 μm**: *2500 rpm medium screen,* **40 μm:** *3500 rpm medium screen,* **20 μm**: *5000 rpm fine screen* and **11 μm:** *9000 rpm fine screen*). For emulsions produced using the high-pressure homogeniser, first a pre-emulsion was produced using the high shear mixer at 9000 rpm with a fine emulsor screen for 5 minutes using the high shear mixer. The pre-emulsion was then subjected to homogenisation, differing in pressure and number of passes (**6 μm**: *20 Bar 3 passes,* **2 μm:** *100 Bar 2 passes* and **0.2 μm:** *1250 Bar 4 passes*). All samples were produced in 400 g batches, under clean and hygienic conditions on the day of evaluation and stored under refrigerated conditions at 2-5 ^{o}C.

5.3.3. Measurement of sensory perception and expected food intake behaviours

Test sessions were scheduled between 10 am and 12 am or 2 pm and 4 pm, Monday to Friday, with sessions lasting 1 hour to 1 hour 30 minutes. Participants were instructed to arrive on one occasion having refrained from consuming any food or beverages other than water 2 hours before their arrival. Participants were seated in individual sensory booths and were presented with 21 40 ml samples in 60 ml twist closure lid pots coded with random 3 digit codes. All samples were served between 5-7 ^{o}C and were visually identical. To minimise volatile loss, all samples were served with the lids closed; participants were instructed only to remove the lid of the relevant sample during its analysis and then replace the lid once sample analysis was complete. Sample order was randomised differently for all assessors. Inter-sample duration was at the participant discretion and ranged from approximately 1-3 minutes. A spittoon was provided and subjects were instructed to spit out the sample after their assessment had been made. A bottle containing 400 g of water with 4 wt.% blue food colouring (Silverspoon blue food colouring liquid, British Sugar Plc, UK) was provided to act as a visual portion size reference for food intake expectation questions, which requires the participant to imagine they were to consume a bottle of the specific sample presented. 1 250 ml bottle of still water and 3 dry crackers were provided, and participants were instructed to use these to refresh their palate between samples. In addition to the randomised presentation of samples for each participant, to further minimise the impact of consuming the water and crackers on predicted food intake ratings, participants were instructed to rinse and spit with the water and ensure crackers were completely consumed by the end of the study (this worked out to be 1-2 bites of cracker after each sample).

Measurements of perceived intensity of sensory attributes, hedonics and expected food intake behaviours were made using visual analogue scales (VAS). Fifteen 100 mm randomised VAS's acquiring information about the intensity of the sensory perception or level of expected intensity of the specific food behaviour e.g. "How <attribute> is sample <code>?" or "Imagine you consumed an entire bottle of sample <code> right now, how strong would your <intake behaviour> be in <time period>?" were presented. These questions were anchored with opposing statements left-to-right e.g. "Not at all <target attribute>" (scored as zero) and "Extremely [target attribute]" (scored as 100) (see Table 5.1). Questions differed slightly in order to be grammatically correct. Pre- and post-test participants rated their mood and appetite via VAS's comprised of a series of questions in the form ''How <word> do you feel?''. The evaluations rated were Full, Hungry, Desire to Eat, Prospective Consumption, Clearheaded, Calm, Happy, Anxious, Tired and Alert, in random order. Before testing, all sensory attributes were discussed individually with participants in accordance to the description shown in Table 5.1. Participants were also given the opportunity to ask any questions about the study and its protocol to clarify issues, queries or definitions before the test began.

Table 5.1. Assessment attributes used during measurements of sensory perception, hedonics and expected food intake behaviour analysis, with description.

Attribute category	Sensory attribute	Description reference
Flavour	Vanilla flavour intensity	Degree of perceived vanilla flavour
	Cream flavour intensity	Degree of perceived cream flavour
	Sweetness	Degree of sweet taste associated with table sugar
Mouthfeel	Smoothness	Degree of absence of any particles, lumps, bumps etc within the sample
	Thickness	Viscous consistency within the mouth; *Water to yoghurt*
	Slipperiness	Degree to which the product slides over the tongue
	Creamy	Soft, smooth with flowing consistency; *Water to full fat cream*
Overall	Creaminess	Assessment of overall creaminess of the sample
	Oiliness	Assessment of overall oiliness of the sample
	Liking	Overall liking of the sample
Expected food intake behaviour	Filling	Measure of expected satiation if to consume 400g, referenced to 400g water portion
	Hunger in 1 hours' time	Measure of expected satiety if to consume 400g, referenced to 400g water portion
	Prospective Consumption in 1 hours' time	Measure of expected quantity consumed, if to consume 400g now of the sample and 400g again in 1 hours' time, referenced to 400g water portion
	Desire to Eat immediately	Measure of expected appetite if to consume 400g, referenced to 400g water portion
	Desire to Eat in 1 hours' time	Measure of expected appetite in 1 hours' time if to consume 400g, referenced to 400g water portion

5.3.4. Data analysis

Data and statistical analysis were carried out using IBM SPSS Statistic (SPSS Statistics 21, SPSS Inc., Chicago, US). The effect of emulsion design (oil droplet size and flavour condition) on sensory perceptions, hedonics and expected food intake behaviour were analysed via general linear model repeated-measures ANOVA. Test-within subject's sphericity assumed significance was taken at 95% confidence interval and degrees of freedom and P values are presented. If a P value was considered significant, a pairwise comparison post-hoc Bonferroni test was performed to reveal the nature of the differences. Pre- and post-test mood ratings were compared via paired t-test with significance being taken at 95%

confidence interval. To assess the direction and variability of relationships between microstructural components and attributes, or attributes and attributes, Pearson's correlation (r) and coefficient of determination (R^2) were performed. Correlations are linear unless stated. Means and standard error of the mean (SEM) are presented throughout.

5.4. Results

5.4.1. Emulsion droplet size

Seven different emulsions varying in droplet size were produced. The volume weighted mean droplet sizes ($d_{4,3}$, µm) were 0.19 ± 0.02, 1.6 ± 0.17, 5.9 ± 0.65, 11.2 ± 0.38, 20.2 ± 0.83, 37.1 ± 0.94 and 48.1 ± 3.3. All samples displayed a unimodal oil droplet size distribution (See section 3.4.1). In all subsequent sections of this chapter, droplet sizes will be referred to as 0.2, 2, 6, 11, 20, 40 and 50 µm for simplicity.

5.4.2. Evaluations of emulsions

The mean sensory and expected food intake ratings are presented in table 5.2.

Table 5.2. Mean (± SEM) sensory and expected food intake ratings (mm) of samples for droplet size (a) and flavour (b) as variables.

	Emulsion sample (Droplet size μm)						
	0.2	**2**	**6**	**11**	**20**	**40**	**50**
Vanilla Flavour	50.7 ± 3.8	48.1 ± 3.5	48.4 ± 3	50.8 ± 3.2	46.9 ± 3	46.3 ± 3.2	39.4 ± 3.7
Cream Flavour	62.3 ± 3	56.4 ± 3.1	57.6 ± 2.4	56.4 ± 2.7	50.8 ± 3.4	49.7 ± 3.9	45.9 ± 3.4
Sweetness	51.5 ± 3.6	48.9 ± 3.4	47.9 ± 3.3	52.2 ± 3.6	44.7 ± 3.6	47.3 ± 3.5	46.5 ± 3.8
Smoothness	61.8 ± 3	63.4 ± 2.9	62.3 ± 3	60.4 ± 3.2	53.3 ± 3.4	54.7 ± 3.7	60.1 ± 3.8
Thickness	43.3 ± 3.8	40.2 ± 3.4	41.5 ± 3	39.8 ± 3.1	38.5 ± 3	43.4 ± 3.3	32.8 ± 2.9
Slipperiness	59.3 ± 3.7	58 ± 3.7	56.7 ± 3.5	56 ± 3.2	54.1 ± 2.9	56.6 ± 3.2	58.1 ± 3
Creamy Mouthfeel	63.3 ± 3.4	58.7 ± 3.7	59.8 ± 3.3	58.7 ± 2.9	51.8 ± 3.4	53.3 ± 4	44.6 ± 3.6
Creaminess	65.5 ± 3.7	59.2 ± 4.1	61.7 ± 3.6	60.3 ± 4	51.1 ± 3.6	50.2 ± 4.1	43 ± 3.5
Oiliness	45.4 ± 3.9	43.6 ± 4.3	43.6 ± 3.4	40.6 ± 3.6	40.8 ± 3.8	44.7 ± 4.2	44.6 ± 3.7
Liking	53.8 ± 2.9	47.8 ± 3.4	48 ± 3	50.1 ± 2.6	43.7 ± 2.5	41.9 ± 3.8	40.4 ± 3.7
Filling	63.2 ± 3.2	61.1 ± 3.7	60 ± 3.7	58.1 ± 2.9	56.4 ± 3.3	57.7 ± 4.3	50.8 ± 4
Hunger in 1 hours' time	44.2 ± 5.4	44.9 ± 5.1	45 ± 4.6	49 ± 4.4	45 ± 4.6	46.3 ± 4.8	57.4 ± 4
Prospective Consumption in 1 hours' time	57.4 ± 5.3	54.2 ± 5.2	59.3 ± 4.3	59.4 ± 5.3	58.3 ± 4.6	59.9 ± 5	59.3 ± 4.5
Desire to Eat immediately	42.4 ± 4.4	41.8 ± 4.7	42.2 ± 4.3	42 ± 4.6	41.9 ± 4.3	44 ± 4.6	48.3 ± 4.2
Desire to Eat in 1 hours' time	48.8 ± 4.4	46.8 ± 4.6	49.6 ± 4.4	51.9 ± 4.1	49.4 ± 4.5	51.1 ± 4.2	54.7 ± 4.1

(a)

	Emulsion sample (Flavour)		
	Vanilla	**Cream**	**No flavour**
Vanilla Flavour	57.7 ± 2.7	46.9 ± 3.9	37 ± 3.3
Cream Flavour	56.3 ± 3.2	57 ± 2.9	49.2 ± 2.5
Sweetness	53 ± 3.2	50.8 ± 4.2	41.5 ± 3.2
Smoothness	61.1 ± 3	58.3 ± 2.9	58.8 ± 2.7
Thickness	41.1 ± 2.6	40 ± 3	38.7 ± 2.4
Slipperiness	57.9 ± 2.6	56.8 ± 3.1	56.3 ± 3.5
Creamy Mouthfeel	54.6 ± 3.4	57.5 ± 3.1	55.1 ± 3.3
Creaminess	56.2 ± 3.4	56.7 ± 3.4	54.6 ± 3.4
Oiliness	43.4 ± 3.6	43.5 ± 3.7	43 ± 3
Liking	52.3 ± 3	46 ± 3.3	41.2 ± 2.7
Filling	57.7 ± 3.1	59.8 ± 3.5	57.1 ± 3.2
Hunger in 1 hours' time	49.2 ± 4.5	46.3 ± 4.7	46.7 ± 4.1
Prospective Consumption in 1 hours' time	61.1 ± 4.6	58.1 ± 5.2	55.6 ± 4.6
Desire to Eat immediately	42.6 ± 4.6	43.4 ± 4.5	43.6 ± 4
Desire to Eat in 1 hours' time	50.5 ± 4.5	49.8 ± 4.5	50.7 ± 3.7

(b)

5.4.2.1. Flavour evaluations

The intensity of rated Vanilla flavour was dependent on both oil droplet size (F (1, 6) = 3.18) and flavour condition (F (1, 2) = 18.53, $p < 0.001$), with no significant interaction (F (1, 12) = 0.63, $p > 0.05$). Vanilla flavoured emulsions were perceived as having significantly greater Vanilla flavour compared to Cream ($p = 0.047$) and No flavour ($p < 0.001$) emulsions. Additionally, Cream flavoured emulsions were perceived as having significantly greater Vanilla flavour than No flavour emulsions ($p < 0.001$). However, perception of Vanilla flavour also decreased significantly with increasing droplet size ($R^2 = 0.73$, $p = 0.006$), with a significant difference between droplets of 50 μm and 11 μm ($p = 0.013$).

The intensity of rated Cream flavour was dependent on both oil droplet size (F (1, 6) = 8.14) and flavour condition (F (1, 2) = 7.87, $p = 0.001$), with no significant interaction (F (1, 12) = 0.54, $p > 0.05$). Cream flavour emulsions were perceived as having significantly greater Cream flavour compared to No flavour emulsions ($p = 0.004$), however not the Vanilla flavoured emulsions ($p > 0.05$). Additionally, Vanilla flavoured emulsions were perceived as having a significantly greater Cream flavour than No flavour emulsions (P 0.03). However, the perception of Cream flavour also decreased significantly with increasing droplet size (R^2 = 0.73, P 0.006) with 50 μm droplets being rated as less creamy than 0.2, 2, 6 or 11 μm emulsions ($p < 0.001$, $p = 0.006$, $p = 0.003$, $p = 0.001$, respectively).

Sweetness intensity was dependent on flavour condition (F (1, 2) = 8.27, $p < 0.001$), but not droplet size (F (1, 6) = 2.01, $p > 0.05$), with no significant interaction (F (1, 12) = 0.47, $p > 0.05$). Vanilla and Cream flavoured emulsions were perceived as significantly sweeter (p = 0.001, $p = 0.02$, respectively) than the No flavour emulsions.

5.4.2.2. Mouthfeel and texture evaluations

Thickness perception intensity was dependent on droplet size (F (1, 6) = 2.6, p = 0.02), but not flavour condition (F (1, 2) = 0.8, p >0.05), with no significant interaction (F (1, 12) = 0.71, p >0.05). Thickness significantly decreased with increasing droplet size (r = - 0.58, R^2 = 0.34), with a significant difference between droplets of 50 μm and 40 μm (p = 0.049).

Creamy Mouthfeel intensity depended on droplet size (F (1, 6) = 9.69, p < 0.001), but not flavour condition (F (1, 2) = 0.84, p >0.05), with no significant interaction (F (1, 12) = 0.98, p >0.05). Creamy Mouthfeel intensity significantly decreased with increasing droplet size (r = - 0.92, R^2 = 0.85, with emulsions with 50 μm droplets being rated as having a less creamy mouthfeel than those with 0.2, 2, 6 or 11 μm droplets (p < 0.001, p = 0.004, p = 0.003, p = 0.001, respectively) and 0.2 μm having a creamier mouthfeel than 20 μm droplets (p = 0.029).

Smoothness perception intensity was dependent on droplet size (F (1, 6) = 3.69, p = 0.002), but not flavour condition (F (1, 2) = 1.4, p >0.05), with no significant interaction (F (1, 12) = 0.69, p >0.05). The significant difference at a 94% confidence interval was between droplets of 2 μm and 20 μm (P 0.059). The trend between droplet size and ratings of Smoothness is interesting; a strong polynomial relationship (R^2 = 0.76) between droplet size and smoothness was demonstrated, despite there being a weak linear relationship (R^2 = 0.29). The polynomial relationship appears to be a result of the increase in perception intensity at 50 μm. A strong linear relationship is observed when 50 μm is removed (R^2 = 0.73); however, the order 2 polynomial relationship also increases in strength (R^2 = 0.82).

Slipperiness perception did not significantly (p >0.05) differ as a function of droplet size (F (1, 6) = 0.55), flavour condition (F (1, 2) = 1) or interaction (F (1, 12) = 0.72).

5.4.2.3. Overall sensory evaluations

Creaminess perception intensity was dependent on droplet size (F (1, 6) = 10.47, p < 0.001), but not flavour condition (F (1, 2) = 0.37, p >0.05), with no significant interaction (F (1, 12) = 0.76, p >0.05). Creaminess significantly decreased with increasing droplet size (r = - 0.94, R^2 = 0.89), with emulsions with droplets of 50 μm being rated as significantly less creamy than those with 0.2, 2, 6, 11 or 20 μm droplets (p <0.001, p = 0.003, p = 0.008, p = 0.006, p = 0.037, respectively). Emulsions with 0.2 μm droplets were also significantly creamier than those with 20 or 40 μm droplets (p = 0.019, p = 0.011, respectively).

Oiliness perception intensity did not depend on oil droplet size (F (1, 6) = 0.07, p >0.05) or flavour condition (F (1, 2) = 0.76, p >0.05), but a flavour condition*droplet interaction was observed (F (1, 12) = 2.803, p = 0.001). Contrasts revealed significant differences in oiliness between 20 μm No flavour emulsions and 6, 20, 40 and 50 μm Vanilla emulsions (p = 0.011, p = 0.001, p = 0.01, p = 0.001, respectively) and 0.2, 2, 6, 11, 40 and 50 μm Cream emulsions (p = 0.011, p = 0.008, p = 0.025, p = 0.008, p = 0.003, p = 0.047, respectively), 0.2 μm No flavour emulsions and 2 and 11 μm Vanilla flavoured emulsions (p = 0.008, p = 0.018, respectively) and 50 μm Cream flavoured emulsions (p = 0.043), 11 μm No flavour emulsions and 20 and 50 μm Vanilla flavoured emulsions (p = 0.021, p = 0.005, respectively) and 20 μm Vanilla emulsions and 50 μm Cream flavoured emulsions (p = 0.04).

Liking was dependent on both droplet size (F (1, 6) = 5.53, p < 0.001) and flavour condition (F (1, 2) = 8.23, p = 0.001), with no significant interaction (F (1, 12) = 0.99, p >0.05). Vanilla flavoured emulsions were liked significantly more than Cream (p = 0.046) and No flavour (p = 0.008) emulsions, but liking of Cream and No Flavour emulsions was similar (p >0.05). However, Liking significantly decreased with increasing droplet size (r = - 0.89, R^2 = 0.79), with 0.2 μm droplet emulsions being liked more than 20, 40 and 50 μm emulsions (p =

0.012, p = 0.011, p = 0.01, respectively) and 11 μm emulsions being more liked than 20 and 50 μm emulsions (p = 0.006, p = 0.045, respectively).

5.4.2.4. Expected food intake evaluations

Expected Filling was dependent on droplet size (F (1, 6) = 3.08, p = 0.007), but not flavour condition (F (1, 2) = 0.67, p >0.05), with no significant interaction (F (1, 12) = 0.8, p >0.05). Expected Filling significantly decreased with increasing droplet size (r = - 0.9, R^2 = 0.8), the significant difference being between emulsions with droplets of 0.2 μm and 50 μm (p = 0.025).

Expected Hunger in 1 hour was dependent on droplet size (F (1, 6) = 5.8, $p < 0.001$

), but not flavour condition (F (1, 2) = 2, p >0.05), with no significant interaction (F (1, 12) = 1.1, p >0.05). Expected Hunger in 1 hour significantly increased with increasing droplet size (r = 0.76, R^2 = 0.57). The significant difference being between emulsions with droplets of 50 μm and those with 2, 6 and 20 μm droplets (p = 0.017, p = 0.008, p = 0.008, respectively).

Expected Desire to Eat in 1 hour was unaffected by oil droplet size (F (1, 6) = 2.18, p >0.05) or flavour condition (F (1, 2) = 0.1, p >0.05), but there was a significant flavour condition*droplet interaction (F (1, 12) = 2.33, p = 0.007). Contrasts revealed significant differences in Expected Desire to Eat in 1 hour for 0.2 μm Vanilla flavoured emulsions and 20 and 50 μm Cream flavoured emulsions (p = 0.034, p = 0.013, respectively) and 11 μm No flavour emulsions (p = 0.048), 2 μm Cream flavoured emulsions and 11, 20 and 50 μm No flavour emulsions (p = 0.026, p = 0.048, p = 0.028, respectively), 6 μm Cream flavoured emulsions and 2 μm No flavour emulsions (p = 0.048), 20 μm Cream flavoured emulsions and 11 and 50 μm No flavour emulsions (p = 0.011, p = 0.009 respectively) and 40 μm Vanilla flavour emulsions (p = 0.025), 40 μm Cream flavoured emulsions and 2 μm No flavour emulsions (p = 0.037) and 50 μm Cream flavoured and 0.2, 2, 6 and 40 μm No flavour emulsions (p = 0.042, p = 0.005, p = 0.015, p = 0.048, respectively).

Ratings of Prospective Consumption and Desire to Eat immediately did not significantly differ as a function of droplet size (F (1, 6) = 1.08, p >0.05; F (1, 6) = 1.94, p >0.05, respectively), flavour condition (F (1, 2) = 2.26, p >0.05; F (1, 2) = 1.94, p >0.05, respectively), or an interaction (F (1, 12) = 1.08, p >0.05; F (1, 12) = 1.15, p >0.05, respectively).

5.4.3. Sensory attribute – expected food intake behaviour correlations

Attribute-Attribute correlations (see Table 5.3) highlight the relationship between sensory attributes and prandial outcome expectations.

Table 5.3. Pearsons correlation (r) Coefficient of determination (Linear R^2) of mean sensory attribute, hedonic and expected food intake ratings as a function of one another.

r

	Thickness	Smoothness	Slipperiness	Creamy Mouthfeel	Creaminess	Oiliness	Liking
Filling	0.85*	0.40	0.28	0.96*	0.92*	0.14	0.84*
Hunger in 1 hours' time	- 0.85*	0.16	0.03	- 0.77*	- 0.70	0.09	- 0.55
Prospective Consumption in 1 hours' time	- 0.09	-0.36	- 0.46	- 0.36	- 0.36	- 0.10	- 0.35
Desire to Eat immediately	- 0.71	0.31	- 0.07	0.80*	- 0.78*	0.44	- 0.66
Desire to Eat in 1 hours' time	- 0.63	- 0.22	- 0.05	- 0.71	- 0.66	0.01	- 0.54
Liking	0.56	0.36	0.58	0.92*	0.96*	- 0.02	

R^2

	Thickness	Smoothness	Slipperiness	Creamy Mouthfeel	Creaminess	Oiliness	Liking
Filling	0.73	0.08	0.08	0.92	0.85	0.02	0.70
Hunger in 1 hours' time	0.73	0.00	0.03	0.59	0.50	0.01	0.30
Prospective Consumption in 1 hours' time	0.01	0.21	0.13	0.13	0.13	0.10	0.12
Desire to Eat immediately	0.50	0.01	0.09	0.65	0.61	0.19	0.44
Desire to Eat in 1 hours' time	0.40	0.05	0.00	0.50	0.44	0.00	0.29
Liking	0.32	0.34	0.13	0.85	0.92	0.00	

*correlation coefficient is significant at $p < 0.05$.

5.4.4. Mood ratings

Participants' mood rating scores were not significantly different pre- and post-test (p >0.05). Therefore, differences in sensory ratings were as a result of sample differences and not participants' mood.

5.5. Discussion

The results of this study indicate that participants, who were untrained, were able to perceive significant differences in flavour, mouthfeel, texture, hedonics and expectations of food intake behaviour as a result of differences in emulsion design: flavour type and oil droplet size.

The microstructural property that had the predominant effect on perceived sensory characteristics, food intake expectations and sample hedonics was oil droplet size. Thus, our findings suggest that greater consideration should be given to this structural component during reformulation of emulsion-based food products. In comparison to previous studies investigating oil droplet size (Akhtar *et al.*, 2005; de Wijk and Prinz, 2005; Vingerhoeds *et al.*, 2008), in this work a larger range of droplet sizes was considered. Our results demonstrate that when a larger oil droplet size range is investigated, many findings emerge that were not evident with narrower range of droplet sizes (2 – 6 µm).

Flavour intensity (Vanilla and Cream) significantly decreased with increasing droplet size. For the flavour within the dispersed phase (Cream), this observation may relate to the greater surface area with smaller droplets. As, the increased contact between the oil phase and the surface of the mouth, could have enhanced flavour intensity, in line with previous findings in other contexts (Malone, Appelqvist & Norton, 2003). For the flavour within the continuous phase (Vanilla), this highlights the dispersed phase effects perception of the continuous phase. Although not well reported in the literature, there is some evidence to suggest that, as NaCas concentrations was a constant, this maybe a result of the greater number of excess protein entities within the continuous phase, with increasing droplet size, binding with continuous phase flavour molecules and reducing their perception intensity (Taylor and Linforth, 2010). However, the observed relationship for both Cream and Vanilla was mainly due to decreased perception of these properties with 50 μm droplets, a finding which highlights a future opportunity to decrease flavour intensity. An interesting observation is that a greater number of oil droplet sizes were significantly different to the sample with 50 μm droplets in the Cream flavoured emulsions, which contained an oil-soluble flavour, than the Vanilla flavoured emulsions that contained a water-soluble flavour. This highlights a potential difference in flavour intensity dependent on the phase location of the flavour within an emulsion system and a surface area effect of droplet size on oil-soluble flavour perception. This would be an interesting area for further investigation.

The main sensory attribute types in which significant differences in perception were generated as a function of oil droplet size were related to mouthfeel and textural sensations. Studies considering Thickness perception and oil droplet size often report increasing Thickness perception with decreasing droplet size. Commonly this is shown to be a result of increasing viscosity with decreasing droplet size, since a strong correlation between viscosity and Thickness perception has been shown previously (Cutler, Morris and Taylor, 1983; Kokini, Kadane and Cussler, 1977; Shama and Sherman, 1973a; Shama and Sherman, 1973b; Wood, 1968). Our observations highlighted a weak linear relationship, with Thickness perception decreasing as droplet size increased, although this was only significant between two oil droplets of adjacent sizes, and so should be interpreted with some caution. This could be a result of the sensory protocol and/or the systems themselves, as suggested in Chapter 3, since only subtle viscosity differences in emulsions of these droplet sizes exist, identifying a perceivable difference in Thickness may be challenging to untrained participants.

Our observations do suggest that droplet size effects Smoothness perception, which agrees with previous observations (de Wijk and Prinz, 2005). Our results using a droplet size range of 0.2 - 50 μm highlight significant differences, but only at a 94% confidence interval. This suggests that although statistical significance is shown, oil droplet size may have a lesser influence on Smoothness than the other attributes. However, the trend between oil droplet size and Smoothness was complicated. At the full droplet size range investigated a polynomial trend was shown; on omitting 50 μm droplets (whose data seemed not to fit the trend for other emulsions) a linear increase in smoothness was shown, however a polynomial trend remained and strengthened. Given the known strength of the correlation between friction coefficient and Smoothness perception (de Wijk and Prinz, 2005; Kokini *et al.,* 1984), the polynomial second order trend with friction coefficient with emulsions of these droplet sizes (see Chapter 3) and our current observations that the significant difference in perception occurs between a small and median size droplets, suggests that with such a large droplet size range the relationship between Smoothness and droplet size is polynomial, but why this is so remains unclear.

Creaminess perceptions of the emulsions were not significantly influenced by flavour type, a relationship also demonstrated by Kilcast and Clegg (2002). Instead our observations show that Creamy Mouthfeel and overall Creaminess increases significantly with decreasing droplet size. Given the strength of correlation between Creaminess and Creamy Mouthfeel (r: 0.99, R^2: 0.98), this strongly suggests overall Creaminess and Creamy Mouthfeel were assessed as the same attribute. This could be attributed to the synthetic manner in which ordinary consumers, as represented the untrained participants, perceived food, assessing the totality of an attribute, instead of assessing attributes analytically when requested (Frost and Janhoj, 2007). Nevertheless, this observation highlights that Creaminess was predominantly influenced by textural/mouthfeel attributes, a conclusion also reached by Frost and Janhoj (2007) in liquid systems. This further suggests that the mechanism through which oil droplet size modified Creaminess was through altered mouthfeel. When hedonics and expected food intake behaviour is also considered, this observation provides an extremely interesting finding which can be related to a modifiable emulsion design property (Table 5.2a).

As previously observed in liquid dairy products (Richardson-Harman *et al.,* 2000) and semi-solids (Daget, Joerg and Bourne, 1987; Elmore *et al.*, 1999) and shown here in liquid emulsions, Creaminess is strongly and significantly positively correlated with the sample's hedonic appeal. When we regard expected food intake behaviours, our results in relation to Creaminess demonstrate a novel and substantial finding.

Expected Filling significantly increased with decreasing droplet size and Expected Hunger significantly decreased with decreasing droplet size. In regards to a predominant sensory characteristic that would be driving these differences, the attribute Thickness (Hogenkamp *et al.*, 2011; Mattes and Rothacker, 2001; McCrickerd *et al.,* 2012; Zijlstra *et al.,* 2009b) displays a strong significant correlation with Expected Filling and hunger in 1 hours' time (see Table 5.3), despite potential erroneous data due to subtleties in viscosity. However, Thickness does not show the strongest correlation (see Table 5.3). Additionally, Smoothness, Slipperiness and oiliness were not shown to be directly involved in hedonics or any expected food intake behaviours (see Table 5.3). Instead, the strongest significant correlation for both Expected Filling and hunger was with Creaminess (see Table 5.3). This suggests with increasing Creaminess we see an increase in Expected Filling and a decrease in Expected Hunger. Therefore, Creaminess, as well as being a predominant influence in hedonics (see Table 5.3), can also generate greater expectations of filling and decreased hunger. If this observation translated to actual eating behaviour, this would highlight Creaminess as a key target attribute, which would allow foods to be engineered via droplet size manipulations to modify eating behaviour, but also maintain hedonic properties (see Table 5.2a). Clearly, future work should determine if expected ratings translate to real behaviour.

Given our earlier discussion regarding participants considering Creaminess as a textural/mouthfeel attribute, this difference in expected food intake behaviour mediated by Creaminess is suggested to be related to textural/mouthfeel sensations. This could be because texture is one sensory characteristic that reliably predicts nutrient content (Drewnowski, 1990) especially for attributes such as Creaminess which are typically associated with fat content (de Wijk, Rasing and Wilkinson, 2003; Frost and Janhoj, 2007). Thus, for energy-dense foods containing structures such as the oil-water emulsions used here, modifying droplet size could lead to enhanced satiety expectations that could enhance the degree to which participants subsequently respond to the ingested fat, in line with evidence that increased satiety expectations increase satiety generated by other macronutrients (Bertenshaw, Lluch and Yeomans, 2013; McCrickerd, Chambers, & Yeomans, 2014; Yeomans and Chambers, 2011). However, if the increase in expected satiety generated by manipulated droplet size was not matched by adequate nutrient ingestion, data suggests there might be a risk of rebound hunger (Yeomans and Chambers, 2011), and so the use of modified droplet size to generate satiety expectations in the context of low-energy products should be approached with caution. Nevertheless, the observation that droplet size affects expected satiety is important in relation to actual short-term eating behaviour when we consider the effect of expectations on eating behaviour mediators such as ghrelin response, which has been demonstrated to be significantly lower if the preload is assumed to be caloric (Crum *et al.*, 2011). Furthermore, our results still highlight an interesting finding that Creaminess may also provide a functional benefit in relation to actual eating behaviour.

With regards to flavour type, the flavour manipulations were included primarily as a positive control to ensure that the ratings used were significantly sensitive to detect effects, guarding against the possibility than droplet manipulations may have had no effects (although in practice droplet size had very clear effects). As expected, a significant increase in ratings of

Vanilla and Cream flavour intensity were observed with the addition of the respective flavour. Interestingly, just the presence of a flavour significantly increased Sweetness and Vanilla and Cream flavour intensities. It is generally considered Sweetness intensity is enhanced by odour, when sweet congruent odours are added to sugar solutions (Cliff and Noel, 1990; Frank and Byran, 1988; Frank, Ducheny and Mize, 1989; Valentin, Chrea and Nguyen, 2006). Odorants like Vanilla and Cream flavours are themselves rated as "sweet" tasting (even though they contain no specific sweet tastants). This enhancement of Sweetness through the presence of odorants has been demonstrated in protocols where samples are swallowed and spat out by participants, as seen within our protocol (Frank, Ducheny and Mize, 1989).

Additionally, our findings highlight a significant increase in Liking was achieved with the addition of Vanilla flavour, compared to No flavour or Cream flavour. Independent of flavour related questions, flavour type did not independently significantly affect the perception of mouthfeel or texture, and did not affect overall or expected food intake behaviour. However, flavour type significantly influenced expected Desire to Eat in 1 hours' time and oiliness in an interaction with droplet size. An unexpected result given that an oil droplet*flavour interaction is not shown in any other expected appetite or satiety attributes. However, findings regarding oiliness are more in line with other findings. Chapter 3, found that frictional properties form a part of Oiliness perception; however, other influences such as flavour could be involved within the formation of the multi-influenced attribute Oiliness. The findings of this chapter support this conclusion, with oiliness perception being a result of an oil droplet*flavour interaction, independent of just flavour or oil droplet size alone. Additionally, as results indicate that flavour only significantly affected perceived flavour intensities, and oil droplet independently affected mouthfeel and textural perceptions. An interaction between the two variables would be expected for a significant difference in perception of an attribute which is comprised of textural and flavour perceptions. Given our observations, using flavour type as a reformulation technique, should only be considered in emulsion based food products when looking to produce a specific flavour or to manipulate Oiliness intensity.

5.6. Conclusions

The present study has shown that changing oil droplet size significantly altered flavour intensity, Thickness, Smoothness, Creamy Mouthfeel, Creaminess, Liking, Expected Filling and Expected Hunger in 1 hours' time. Altering the flavour of these emulsions using odour-based flavourants only significantly changed flavour intensity, Sweetness and Liking. The most important observation highlighted in this study is that by altering the emulsion design through decreasing oil droplet size, perceived Creaminess can be significantly enhanced which as a result significantly increases Hedonic appeal as well as increasing Expected Filling and reducing Expected Hunger, independent of energy content. If shown to relate to actual eating behaviour, this would provide a key target attribute which can be manipulated through emulsion design, to produce hedonically appropriate satiating foods.

Chapter 6. Emulsion oil droplet size significantly affects satiety: A pre-ingestive approach

Data and discussions contained within this chapter have been published within:

Lett, A.M., Norton, J.E. and Yeomans, M.R. (2016). Emulsion oil droplet size significantly affects satiety: A pre-ingestive approach**.** .*Appetite, 96*, 18-24

6.1. Abstract

Previous research has demonstrated that the manipulation of oil droplet size within oil-in-water emulsions significantly affects sensory characteristics, hedonics and expectations of food intake, independently of energy content. Smaller oil droplets enhanced perceived creaminess, increased Liking and generated greater expectations of satiation and satiety, indicating that creaminess is a satiety-relevant sensory cue within these systems. This paper extends these findings by investigating the effect of oil droplet size ($d_{4,3}$: 2 and 50 µm) on food intake and appetite. Male participants (n = 34 aged 18 – 37; BMI of 22.7 ± 1.6 kg/m^2; DEBQ restricted eating score of 1.8 ± 0.1.) completed two test days, where they visited the laboratory to consume a fixed-portion breakfast, returning three hours later for a “drink”, which was the emulsion preload containing either 2 or 50 µm oil droplets. This was followed 20 minutes later with an *ad libitum* pasta lunch. Participants consumed significantly less at the *ad libitum* lunch after the preload containing 2 µm oil droplets than after the 50 µm preload, with an average reduction of 12% (62.4 kcal). Despite the significant differences in intake, no significant differences in sensory characteristics were noted. The findings show that the impact that an emulsion has on satiety can be enhanced without producing significantly perceivable differences in sensory properties. Therefore, by introducing a processing step which results in a smaller droplets, emulsion based liquid food products can be produced that enhance satiety, allowing covert functional redesign. Future work should consider the mechanism responsible for this effect.

6.2. Introduction

Fat is the most energy dense macronutrient at 9 kcal per gram (Atwater and Woods, 1896) and consequently is of interest in the redesign of food products to tackle the "obesogenic" food environment. Reducing fat content within foods has been a commonly proposed method to reduce consumers' energy intake. However, this is typically detrimental to the food product's sensory properties (Norton, Moore and Fryer, 2007; Roller and Jones, 2001).

Increasing the functionality of the fat to reduce intake could be a novel alternative to produce inherently "healthier" fat based foods (Himaya *et al.,* 1997). Increasing a food product's impact on satiety may lead to a reduction in overall energy intake through inhibition of appetite after consumption (Chambers, McCrickerd and Yeomans, 2014; Hetherington *et al.,* 2013).

Designing food structures for functional benefits is a growing area of interest. Redesigning foods that are high in fat (such as emulsions) to impact on appetite has added importance because fat is considered to be the least satiating macronutrient (Blundell, Green, and Burley, 1994; Blundell and Macdiarmid, 1997; Blundell and Tremblay, 1995). Emulsions are common fat based food structures that are found within a variety of commercially available food products, such as sauces, condiments, spreads, dressings and desserts. Emulsions are formed by mixing two immiscible liquids, such as oil and water, so one liquid is dispersed within the other as droplets stabilised by an emulsifier.

Previous research considering emulsion structures has predominantly considered gastro-intestinal structuring, in an attempt to achieve satiety via post-ingestive and post-absorptive mechanisms, with emulsion oil droplet size and emulsifier type being the two main properties investigated (Armand *et al.,* 1999; Maljaars *et al.,* 2012; Mun, Decker and McClements, 2007; Golding and Wooster, 2010; Lundin, Golding and Wooster, 2008; Peters *et al.,* 2014;

Seimon *et al.*, 2009; Singh, Ye and Horne, 2009; van Aken *et al.*, 2011). However, structuring emulsions to achieve satiety via pre-ingestive approaches (i.e. considering sensory mechanisms) has recently been considered and highlighted as potentially effective (Lett et al., 2016). In that study, decreasing the oil droplet size within an oil-in-water emulsion model drink, increased creaminess, which in turn increased liking and expectations of satiation and satiety, independent of energy content (Lett et al., 2016). Creaminess within emulsions was therefore highlighted as a hedonic sensory cue, and a potential satiety-relevant sensory cue, which agrees with other findings that high-energy beverages are more satiating when creamy sensory characteristics are present (McCrickerd, Chambers and Yeomans, 2014; Yeomans and Chamber, 2011). The mechanism by which satiety-relevant sensory cues appear to work suggests that people learn to associate sensory characteristics with the subsequent experience of satiety post-consumption (Brunstrom, Shakeshaft and Scott-Samuel, 2008; Yeomans *et al*., 2014). As such, it is thought that creaminess, which is typically associated with high fat content (de Wijk, Rasing and Wilkinson, 2003; Frost and Janhoj, 2007), generates expectations of satiety typically achieved after the consumption of fat containing energy dense foods, with the intensity of creaminess being a predictive marker of energy content.

If the enhanced expectation of satiety through altering oil droplet size also impacts on the experience of post-ingestive satiety, this could confirm this type of restructuring as a valuable approach to product development. Early pre-ingestive satiety signals, such as sensory properties integrate with post-ingestive and post-absorptive signals (Blundell, Rogers, and Hill, 1987), and adjust digestive and absorptive mechanisms accordingly, at least partly through anticipatory physiological responses (Power and Schulkin, 2007; Smeets, Erkner and de Graaf, 2010).

The present study aimed to extend previous findings from Lett et al. (2015). We hypothesised that reducing the average oil droplet size of an oil-in-water emulsion will enhance satiety, through pre-ingestive sensory-mediated routes by increasing the perception of the identified satiety-relevant sensory cue, creaminess.

6.3. Materials and methodology

6.3.1. Design

A repeated-measures single-blind randomised cross-over design preload paradigm was used to investigate the satiating effects of two oil-in-water emulsion based drinks, varying in oil droplet size, but with equal energy content. Test meal intake and subjective ratings (Visual analogue scales: VAS) were used to assess food intake behaviour. Ethical approval for the study was obtained from the University of Birmingham ethics committee.

6.3.2. Participants

Thirty-four healthy male adults participated in the study. Sample size was determined on the basis of the effect size needed to find a difference in satiety between two emulsions with different average oil droplet sizes (2 and 50 µm). These emulsions were produced in a preliminary study in which oil droplet size of an emulsion beverage had been manipulated changing sensory properties (Lett et al., 2016). To estimate participant numbers, we examined the outcome of previous preload studies where a difference in creaminess, similar in size to that in our recent emulsion study, was associated with a significant reduction in intake at a similar test meal. One such study where a difference in creaminess was associated with reduced intake was Yeomans and Chambers (2011), where less was consumed after a preload with higher rated creaminess (achieved primarily by varying viscosity) than after an isoenergetic less creamy preload. Based on the intake data in that study, one-tailed significance (p <0.05, predicted reduction with more creamy preload) and power = 0.8, indicated that a sample of 34 would be required. All participants were staff or students at the University of Birmingham, who had expressed an interest in participating in a research study investigating "The effect of mood on appetite", as to mask any expectancy effects concerning the true nature of the investigation. Prospective participants were contacted by a recruitment email via an email database and were asked to reply if they were interested in participation and considered themselves to be a healthy, non-smoking, normal weight (BMI: 18.5 -25) male with no food allergies or intolerances. Females were excluded as they typically practice significantly higher levels of restricted eating and other eating behaviours than males (Arganini *et al.*, 2012; Fortes *et al.*, 2014; Wardle, 1987), and males who do not restrict their eating behaviour were chosen, as this cohort demonstrates the most accurate regulation of food intake (Rolls *et al.*, 1994). Respondents to the recruitment email were provided with an information sheet and enrolled in the study if they were still interested in participation. Prior to the start of a session, participants were screened for food allergies, smoking habits and current medical status via a health questionnaire, body mass index (BMI), calculated as kg/m^2 (with height and weight measurements being obtained with participants wearing light clothes and in a fasted state using a freestanding stadiometer (Seca 213, Birmingham, UK) and digital calibrated weighing scales (Seca 813, Birmingham, UK) and dietary restraint measured using the restraint scale from the Dutch eating behaviour questionnaire (DEBQ) (van Strien, *et al*.,1986). Potential participants were prevented from participating if they indicated any food allergies, history of smoking, had a BMI above 24.9 kg/m^2 or below 18.5 kg/m^2, were taking medication known to interfere with sensory perception or food intake or had a DEBQ restricted eating score of >2.4. One potential participant was prevented from participating, based on the recruitment criteria. Additionally, participants were given the opportunity to ask any questions about the study and its protocol to clarify issues or queries before the study began. The test cohort was made up of 34 men aged 18 - 37, with a mean BMI of 22.7 ± 1.6 Kg/m^2 and DEBQ restricted eating score of 1.8 ± 0.1. All participants gave written informed consent prior to participation.

6.3.3. Procedure

Participants attended 2 sessions on non-consecutive days. Study protocol was identical on each test day, with only the preload varying (See Fig. 6.1). Participants arrived at a scheduled date and time between 08.30 - 10.30 am, Monday to Friday. Participants arrived having consumed only water from 11.00pm the night before. All testing was carried out in an individual booth containing a PC computer running Sussex Ingestion Pattern Monitor (SIPM). SIPM was used to collect VAS scores of all mood and appetite questions throughout the study and preload sensory scores, and monitor food intake at lunch with a digital balance concealed by a placemat (Sartorius BP 4100). All VASs used within the study, collecting data on mood, appetite and preload sensory ratings were randomised differently for all participants. SIPM equipment and software were developed at the University of Sussex (Yeomans, 2000), based on a modification of the Universal Eating Monitor developed by Kissileff, Kilngsberg, and Van Italie (1980), and has been used extensively in studies of human appetite (Yeomans and Bertenshaw, 2008). After successful screening, the participant sat within an individual booth to begin the breakfast session and consumed the test breakfast (see Section 6.3.4.1) within 15 minutes. To begin participants completed a set of the mood and appetite questions. The mood ratings (Alert, Anxious, Calm, Clearheaded, Happy and Tired) and appetite ratings (Hunger and Fullness), were presented as 100-point computerised VASs anchored with "not at all [mood or appetite]" and "extremely [mood or appetite]". Mood questions were included as distracters and to be consistent with the premise that the study was investigating "The effect of mood on appetite". The participant was then instructed to return exactly 3 hours later for the preload session. During the inter breakfast-preload period participants were not allowed to participate in exercise or consume any food or drink, apart from a 250 ml bottle of still water, which was provided and had to be fully consumed upon their return. Upon the participants return, they began the preload session. Participants completed the standard mood and appetite questions and then were presented with 200 ml of one of the two preloads (see Section 6.3.4.3). 17 participants received the 2 µm droplet preload on their first session and the other 17 participants received the 50 µm droplet preload on their first session, with the other preload being consumed on the second session. SIPM instructed the participant to take a mouthful and then carry out a number of VAS to assess the samples sensory characteristics. The preload was evaluated for Thickness, Slipperiness, Smoothness, Creamy Mouthfeel, Overall Creaminess, Liking, expectation of Hunger in 1 hours' time (Satiety) and expectation of Fullness immediately (Satiation). Both questions determining expectations of food intake were in reference to if they consumed the full portion presented. Sensory VAS questions were headed "How [target rating] is the drink?" and end-anchored with "not at all [target rating]" (scored as zero) and "extremely [target rating]" (scored as 100); wording may have slightly differed to be grammatically correct. Upon completion of the sensory VAS questions, SIPM instructed the participant to consume the rest of the preload within 5 minutes, before another series of standard mood and appetite questions were presented to finish the preload session. Participants then remained within the laboratory until the lunch session. Results from our previous work showed that expectations of food intake are significantly different due to sensory differences between the emulsions. As such, a 20 minute delay between the preload being presented and the lunch session was used. This fits within the optimal time period for detecting oro-sensory effects on satiety (<30 minutes) (Livingstone *et al*., 2000), and allowed enough time for participants to comfortably consume the preload and complete all mood, appetite and sensory VASs. During the lunch session, participants first completed a set of standard mood and appetite ratings in the absence of any food cues (pre-lunch ratings). Next, 500 g white penne pasta with tomato and herb sauce (see Section 6.3.4.2) was served by an experimenter who explained that the participant could eat as little or as much as he liked. A hidden digital balance secured under a placemat and linked to SIPM, which recorded the weight of food being eaten. If the participant consumed 300 g of the lunch, an onscreen alert message prompted the participant to call the experimenter. The experimenter then served the participant another 500 g pasta in a new bowl, with the consumed bowl of pasta being removed; no limit was placed on the number of refills permitted. To reduce the influence of habit and portion-size effects on intake, participants were encouraged not to use the refill prompt as a cue to end the lunch session. When participants had confirmed that they had finished eating, the participants then completed a final set of standard mood and appetite ratings (post-lunch ratings) before the lunch session and test day was completed. On the final test day, the participants were given a £20 Amazon voucher as compensation for participating in the study.

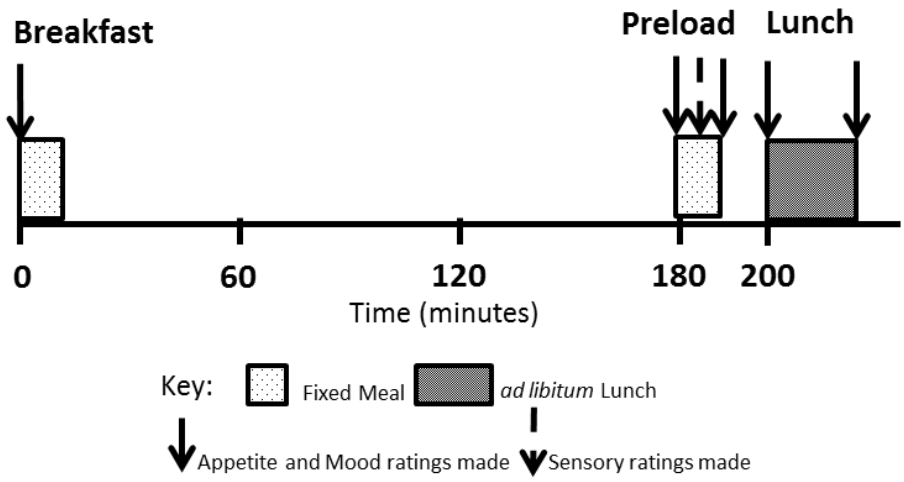


Fig. 6.1 Schematic representation of the timing of the fixed and test meals and the sets of appetite and mood ratings and sensory ratings on a test day.

6.3.4. Test Foods

6.3.4.1. Standard Breakfast

On the morning of each test day, participants consumed a breakfast of 60 g of a proprietary breakfast cereal (Crunchy Nut Cornflakes; Kellogg Co) plus 160 mL semi-skimmed milk (Tesco) and 200 mL orange juice (Tesco). The breakfast provided 420 kcal, 6.3 g fat, 10.8 g protein, and 79.1 g carbohydrate. The breakfast provided approximately 17% of a male adults daily average recommended energy intake.

6.3.4.2. Lunch

For the ad libitum lunch, each 500 g serving of pasta consisted of 300 g cooked weight of white pasta (Penne; Aldi) plus 200 g of a prepared pasta sauce (tomato and herb; Aldi) served hot. The pasta lunch was cooked on the test day as per packaging instructions. The test meal provided 96 Kcal energy (3.2 g protein; 19.5 g carbohydrate and 0.58 g fat) per 100 g.

6.3.4.3. Drink Preloads

The preload drinks were 200 ml of emulsions containing either 2 or 50 µm droplets; these were *No Flavour* versions of emulsion samples described in a previous study (Chapter 5). These emulsions were chosen as, based on our previous work, emulsions containing 2 µm droplets gained significantly greater ratings for Creaminess (p = 0.003) and Liking (p = 0.01) and resulted in reduced expectations of Hunger in 1 hours' time (Satiety) than the emulsion containing 50 µm droplets (p = 0.017). Rheological and lubrication properties of these systems have been investigated in other work and it was shown that 2 and 50 µm emulsions were also comparable in these properties (Chapter 3). Samples consisted of an oil-in-distilled water emulsion (1 wt. % sodium caseinate (Excellion EM7, DMV International, The Netherlands); 2 wt. % sucrose (Silverspoon granulated, British Sugar Plc, UK) and 15 wt. % sunflower oil (Tesco Plc, UK)). Emulsions were produced using two different methods dependent upon the required mean droplet size of the emulsion being produced: a high shear mixer (Silverson L5M, Silverson machines Ltd, UK) or a high-pressure homogeniser (GEA Niro Soavi Panda Plus 2000, GEA Niro Soavi, Italy). In a 600 ml beaker, 15 wt.% sunflower oil was added to 85 wt.% aqueous phase (1 wt.% NaCas, 2 wt.% sucrose, 97 wt.% distilled water solution). The whole sample was then emulsified for 5 minutes using the high shear mixer. Dependent on oil droplet size being produced the sample was subjected to a different rotational speed (rpm) and emulsor screen. 50 µm oil droplet samples were subject to high shear mixing at 2500 rpm with a 1.6 mm pore emulsor screen. 2 µm oil droplet samples were subject to high shear mixing at maximum rpm with a 0.8 mm pore emulsor screen to produce a pre-emulsion, the pre-emulsion was then homogenised at 100 Bar with 2 passes. All samples were produced in 400 g batches, under clean and hygienic conditions on the day of evaluation and stored under refrigerated conditions at 2-5 ^{o}C. The 200 ml emulsion preload provided approximately 282 kcal, 30 g fat, 2 g protein, and 4 g carbohydrate.

6.3.5. Data Analysis

The aim of the study was to investigate whether altering the oil droplet size of an emulsion altered subsequent food intake behaviour. Data were analysed using the Statistical Package for the Social Sciences (SPSS) version 21 (SPSS Inc., USA). VAS scores for hunger and fullness throughout the study are reported from baseline (pre-preload) data, and were analysed using 2-way ANOVA based on the three post-preload time points (immediately post-preload, pre-lunch and post-lunch) and two oil droplet sizes. Nutrient and energy composition of the breakfast and lunch was calculated using compositional data provided by the manufacturers. The energy density of the preload drink was calculated using Atwater factors (Atwater and Woods, 1896).

6.4. Results

6.4.1. Mood and appetite ratings

Protected contrasts of baseline evaluations of mood and appetite (hunger and fullness) ratings before preload ingestion (at breakfast and just before preload consumption) did not differ significantly, and so effects of preload oil droplet size on appetite were assessed using change from baseline data. As can be seen (Figure 6.2a), hunger decreased immediately after the preload, recovered prior to lunch and then fell markedly after lunch, reflected in an overall effect of rating time on hunger ($F(2,66) = 182.68$, $p<0.001$, ETA = 0.85), but the change in hunger was consistently lower after the preload with 2 µm than 50 µm oil droplet size ($F(1,33 = 9.66$, $p=0.004$, ETA = 0.23), with no significant time x droplet interaction ($F(2,66) = 1.04$, $p=0.36$, ETA = 0.03). Fullness ratings showed the reverse pattern over time to hunger (Figure 2b: effect of time $F(2,66) = 82.45$, $p<0.001$, ETA = 0.71), but there was no significant effect of droplet size ($F(1,33 = 0.07$, $p=0.80$, ETA = 0.01) or time x droplet interaction ($F(2,66) = 0.71$, $p=0.50$, ETA = 0.02).

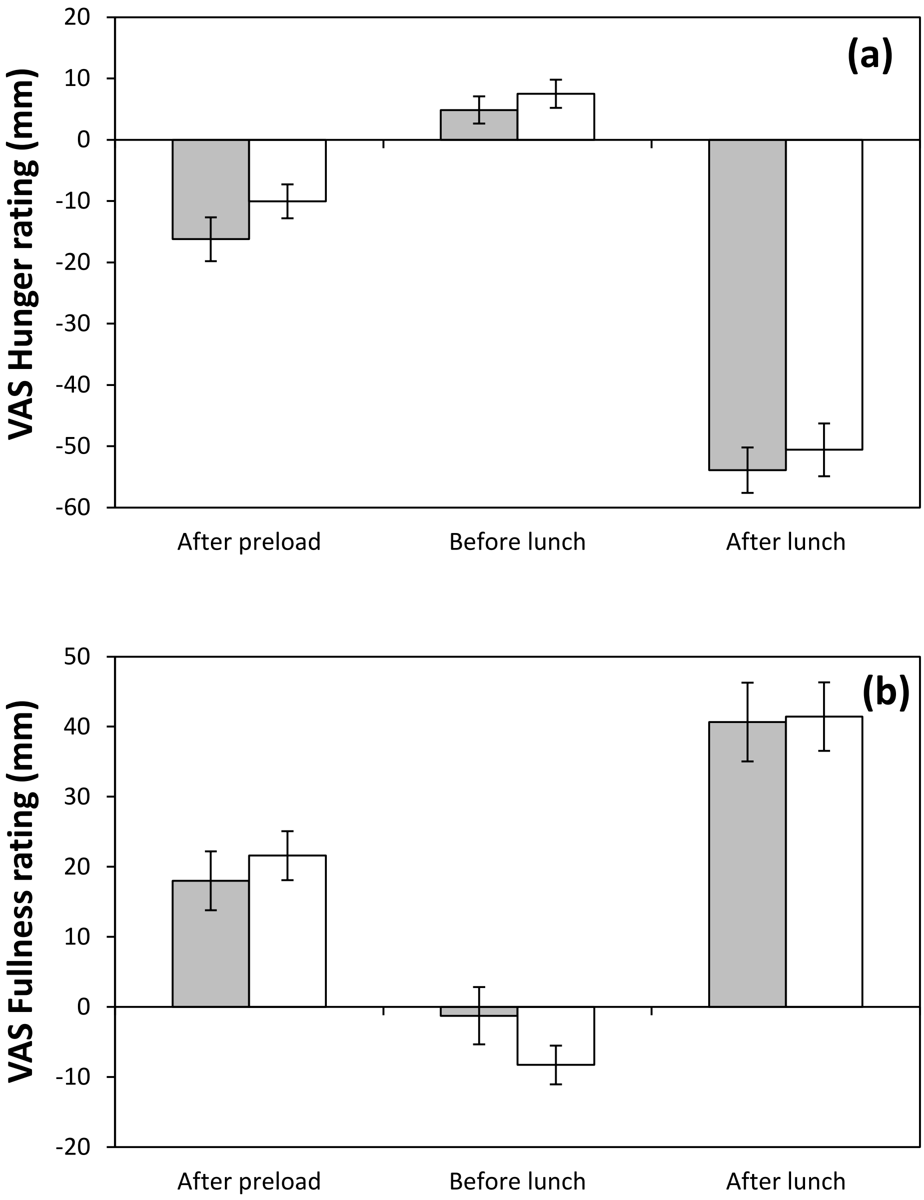


Fig. 6.2 Mean (± SEM) changes in VAS ratings (mm) calculated from baseline of ratings of Hunger (a) and Fullness (b) across the course of the test session for both 2 µm (filled bars) and 50 µm (open bars) emulsion preloads.

6.4.2. Preload sensory and hedonic ratings

There were no significant differences in the scores of any sensory attributes, hedonics and expectations of food intake for the 2 µm and 50 µm emulsion preloads (p >0.05: See Table 6.1). This finding contradicts previous results (Chapter 5: see Table 6.1) and is discussed further in section 6.5.

Table. 6.1 Mean (± SEM) of attribute ratings of 2 μm and 50 μm samples used in current study and Chapter 5 (N = 24). Filled cells represent significance (p < 0.05) between 2 μm and 50 μm.

Attribute	When Rated	2 μm	50 μm
Thickness	Current Study	43.4 ± 3.1	47 ± 3.6
	Chapter 5	40.2 ± 3.4	32.8 ± 2.9
Creamy Mouthfeel	Current Study	58.8 ± 2.9	60.3 ±3.2
	Chapter 5	58.7 ± 3.7	44.6 ± 3.6
Creaminess	Current Study	56.9 ± 3.2	59.4 ± 3.4
	Chapter 5	59.2 ± 4.1	43 ± 3.5
Slipperiness	Current Study	61.6 ± 2.8	62.3 ± 2.4
	Chapter 5	58 ± 3.7	58.1 ± 3
Smoothness	Current Study	65.9 ± 2.9	68.2 ± 2.8
	Chapter 5	63.4 ± 2.9	60.1 ± 3.8
Liking	Current Study	40.9 ± 3.6	39.4 ± 3.6
	Chapter 5	47.8 ± 3.4	40.4 ± 3.7
Expected Fullness	Current Study	54.3 ± 3.3	52.8 ± 4
	Chapter 5	61.1 ± 3.7	50.8 ± 4
Expected Hunger	Current Study	62.6 ± 4.2	62.9 ± 4
	Chapter 5	44.9 ± 5.1	57.4 ± 4

6.4.3. Lunch Intake

Total lunch intake was significantly different dependent on oil droplet size preload consumed, with participants consuming significantly less after consumption of the 2 μm preload (p = 0.027). Total consumption was 67.7g or 62.4 Kcal less, which is a 12.3 % reduction in total food intake (g) and a 12.2 % reduction in energy intake (Kcal) (see Fig. 6.3a and b). No significant effect of preload session order on intake, for both droplet sizes (p >0.05), was also shown, highlighting participant fatigue of the protocol did not factor in ratings or *ad libitum* food intake.

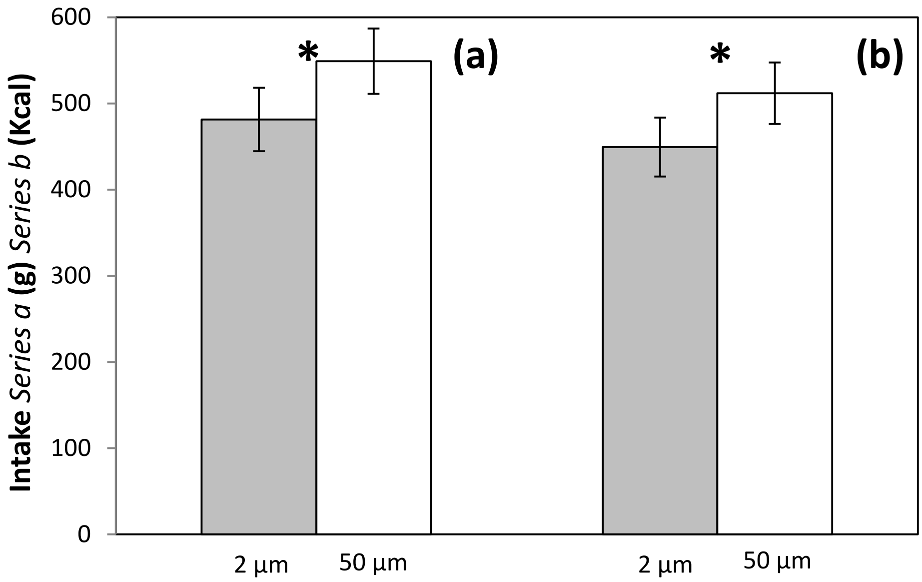


Fig. 6.3 Mean overall intake at the test meal (± SEM) in grams (a) and kilocalories (b). Filled bars represent preloads containing 2 µm droplets and Open bars represent preloads containing 50 µm. * represents significance at $p < 0.05$.

6.5. Discussion

The main finding from this study was that by decreasing the oil droplet size of an oil-in-water emulsion, the degree to which an emulsion impacts on satiety can be significantly increased, independent of energy content. Participants consumed 12.2 % (Kcal) less at the test meal after consuming an oil-in-water emulsion preload containing 2 µm droplets, than they did following consumption of a preload containing 50 µm oil droplets (See Fig.6.3a and b).

Earlier work (Chapter 5) looked to identify satiety-relevant oro-sensory cues within model oil-in-water emulsions, with the intention of designing emulsion structures to promote these cues, therefore increasing an emulsion based food or beverages capacity to generate satiety. Using the same model emulsion systems as used within this study, the authors showed that on decreasing the oil droplet size of the emulsion, Creaminess perception significantly increases (see Table 6.1). Reducing oil droplet size also significantly increased hedonic appeal, in addition to significantly decreasing expectations of Hunger in 1 hours' time (an indication of satiety). As such, it is thought that, Creaminess is a potential satiety-relevant oro-sensory cue.

Our current work has shown that although expectations of food intake behaviour have been successfully realised in actual eating behaviour (See Fig. 6.3), the mechanism mediating the effect has not been identified, as ratings of Creaminess, or any other attribute, for the two preloads were not significantly different (see Table 6.1). Therefore, our findings do not fully agree with our hypothesis. Given that Chapter 5 identified potential satiety-relevant sensory cues within these systems, and that the current studies protocol was designed to maximise the influence of potential sensory effects of the preload on subsequent food intake (Blundell, 2010; Livingstone *et al*., 2000), it is unusual that a significant difference in satiety was identified (See Fig. 6.3), but no significant differences in sensory perception were found.

Other studies have also shown differences between sensory properties of preloads in the "pilot" sensory study, but not in the "main" preload study, despite similarities in the studies cohort (Chambers, Ells and Yeomans, 2013; McCrickerd, Chambers and Yeomans, 2014; Yeomans and Chambers, 2011). Consequently, it seems sensible to suggest that the difference in protocol between this and our earlier study (Lett et al., 2016), is the reason for the change in sensory results. The protocol in Lett et al. (2015) promoted sample assessment in a more analytical manner. Firstly, participants were recruited to participate in a "sensory analysis of emulsions" study, so would have approached the study consciously seeking sensory differences between samples. Although samples were unidentifiable and randomly ordered, the methodology used would not have controlled for the cross-comparison of sensory attributes between samples, as samples were analysed in a sequential manner in one session. Secondly, all sensory attributes investigated were defined via a description reference and not at the discretion of the individual participant, as was the case within the current protocol. Our previous study (Lett et al., 2016) also used 100mm paper-based VAS scale, compared to the use of 100-point computerised VAS using SIPM here. Although no published study has explicitly compared manual VAS ratings and computerised based VAS on SIPM, studies have shown VAS scores change, even subtlety, dependent on the protocol for collecting VAS based data used (Brunger *et al.*, 2015). Within the current study, participants were recruited to participant in a study investigating “mood and appetite”, and so attention was not drawn specifically to the preload’s sensory properties. Furthermore, and importantly, although preloads were also unidentifiable and randomly ordered, they were assessed for sensory attributes at least 48 hours apart, with participants’ practicing free-living behaviour between test days. The method would therefore have hindered participant’s ability to draw cross-comparisons between sensory attributes of the preloads as seen with the sensory protocol of the previous study. Consequently, results presented by Lett et al. (2015) would be expected to highlight more pronounced sensory differences between samples because of the comparative nature of the rating task used in the earlier study. Nevertheless, given participants consumed commercially available foods at customary meal times, with at least a 48 hour free-living period in-between test days, our current studies protocol is more replicable of “real world” behaviour. As no significant differences in sensory properties between 2 µm and 50 µm emulsion preloads were identified within this study (see Table 6.1), findings indicate that satiety can be significantly enhanced without producing significantly perceivable differences in sensory properties. Therefore, using the same formulation, by introducing a processing step which results in a smaller average droplet size (for example, higher shear/pressure processing), emulsion based liquid food products can be produced with enhanced effects on satiety, but with a very similar sensory profile as the original product, allowing functional redesign unbeknown to the consumer.

A methodological issue with studies investigating satiety is the considerable overlap of physiological and cognitive factors in satiety development (Livingstone *et al.*, 2000). The mechanism in which oil droplet size changes satiety can, therefore, not be characterised simply according to one factor of the “satiety cascade” (Blundell, Rodgers and Hill, 1987), especially as a lack of clarity exists concerning the primary mechanism of our main finding (See Fig. 6.3).

To the best of our knowledge this is the first paper to consider an emulsion structuring approach for pre-ingesitve mediated satiety. Previous work considering emulsion oil droplet size as a design mechanism for satiety has only considered gastrointestinal structuring (Golding and Wooster, 2010; Lundin, Golding and Wooster, 2008; Singh, Ye and Horne, 2009). Emulsion gastric structuring for satiety being an approach that uses physiological understanding to structure and design emulsions, to ensure, under gastric conditions, the emulsion behaves or structures in a certain way, which is favourable in enhancing satiety. Although gastric colloidal behaviour is largely governed by emulsifier type (Mun, Decker and McClements, 2007; van Aken *et al.,* 2011), oil droplet size has been shown to effect digestive and absorptive behaviours, which would impact on satiety through post-ingestive and post-absorptive effects and feedback mechanisms. For example, a considerably greater rate of lipolysis (and therefore plasma triglyceride concentration and CCK release) is observed with smaller oil droplet sizes, due to the greater interfacial area available for digestive lipase binding. This behaviour has been observed within *in vitro* (Armand *et al.,* 1992; Peters *et al.,* 2014) and *in vivo* studies (Armand *et al.,* 1999; Borel *et al,* 1994; Maljaars *et al.,* 2012; Peters *et al.,* 2014; Seimon *et al.,* 2009). However, it should be understood that the pre-prandial oil droplet size may change substantially through all digestive mechanisms prior to gastric or intestinal entrance (van Aken, Vingerhoeds and de Hoog, 2007). Apart from Peter's work (2014), all previous work mentioned has bypassed oral processing, via infusion of the emulsion to specific sites of the gastrointestinal tract. However, in reality there is no disassociation between sensory and gastric influences on satiety. In fact, even before the arrival of food to the gut, sensory and cognitive signals, generated by the visual and sensory aspects of a food, are influencing food intake behaviour. These early pre-ingestive satiety signals integrate with post-ingestive and post-absorptive signals to determine the overall satiating capacity of a food, by influencing physiologically readiness for effective digestion, absorption and metabolism, through mechanisms such as endocrine response and gastric/intestinal secretions and motility. Cassady, Considine and Mattes (2012) demonstrated the importance of pre-ingestive sensory and cognitive information on physiological satiety responses as ratings of hunger were lower, gastric emptying was slower, insulin and GLP-1 release increased, ghrelin decreased and subsequent *ad libitum* food intake was lower when participants believed a beverage preload would gel in their stomach, even though it did not. This was also reflected in the subjective comments made by participants after the consumption of the preloads. Additionally, studies designed to bypass pre-ingestive signals have demonstrated weaker satiety responses than studies also considering sensory and cognitive influences (Cecil *et al*, 1998; Cecil, Francis and Read, 1998; Lavin *et al.*, 2002). This evidence highlights the importance of pre-ingestive sensory signals in subsequent satiety response through interaction with subsequent satiety mechanisms (anticipatory physiological regulation interactions). Overall, this suggests that although no difference in sensory properties is observed between preloads within this study (see Table. 6.1), the difference in satiety (See Fig. 6.3) suggests not to be exclusively physiologically mediated. Therefore the sensory results from the previous study should be considered (Lett et al., 2016), especially as the only variable was droplet size.

Having demonstrated a clear effect of manipulated droplet size on the behavioural expression of satiety, a key question is how this effect was achieved, and there are a number of possible explanations which would be valuable for future work to consider. One possibility is that the subtle differences in orosensory experience of the emulsions (which were clearly evident in our earlier study but less evident from the ratings made in the present study) differentially effect cephalic phase responses (Smeets, Erkner and de Graff, 2010) , so altering the degree to which the gut was primed to respond to the ingested nutrients. To test this, future studies should examine how the pattern of release of key hormones implicated in cephalic phase responses (e.g. insulin and pancreatic polypeptide) and in broader satiety responses (e.g. CCK, PYY, GLP1) differ depending on emulsion droplet size. Additionally, extensional work should look to assess whether such satiety responses are reflected with repeated consumption of these preloads. Such findings would be important in understating whether participants modify their satiety response, as a result of a learning effect between the ingested energy content and preparatory cognitive and sensory influences. This would highlight the effectiveness of the microstructural approach used within this study in the longer term, and may highlight whether sensory differences between preloads are detectable, if a modified satiety response occurs. In a formulation context, whether droplet size still manipulates satiety at different fat concentrations would also be of interest, particularly, lower fat concentrations as energy dense foods, which taste satiating, can promote appetite through rebound hunger mechanisms (Chambers, McCrickerd and Yeomans, 2015). Nevertheless, if successful, this would allow integration of effective satiety approaches into lower fat foods. Finally, to begin creating an integrated approach, in microstructural engineering efforts for satiety, investigating the difference between the consumed and the oral/gastric/intestinal oil droplet size would be beneficial, as anticipatory physiological regulation responses and gastric structuring approaches can begin to be combined.

6.6. Conclusions

The present study has shown that smaller droplets within an emulsion preload result in a significant reduction in food intake at a subsequent *ad libitum* meal, independent of formulation change, energy content and perceivable changes in sensory characteristics. This outcome suggests that emulsion based liquid food products can be produced to impact upon satiety, but with the same sensory properties as the original product. Future studies should look to further understand the relationship between emulsion droplet size in relation to satiety and the application of these results in commercially available food systems.

Chapter 7. Conclusions and future recommendations

Taking a unique multidisciplinary approach, combining understanding of food engineering, sensory science, nutrition and psychology, the objective of this thesis was to advance the understanding of microstructural engineering approaches to produce emulsion based food products, which have maintained or improved sensory and hedonic qualities, but that promote functional benefits of modified food intake behaviour. Oil droplet size was investigated in regards to its effect on emulsion physical, sensory, hedonic and satiating properties. Specifically, the work consisted of investigating the:

- Inter-relation between emulsion microstructure, modified by oil droplet size, physical properties, viscosity and friction, and sensory outcomes, particularly Creaminess and its sensory constitutes (Chapter 3 and 4).

- Inter-relation between emulsion microstructure, modified by oil droplet size, sensory outcomes, particularly Creaminess and expected and actual food intake behaviour (Chapter 5 and 6).

This thesis is a reflection of pioneering attempts to apply chemical engineering principles through multidisciplinary approaches to modify consumer eating behaviour. Although the purpose of food research is for the benefit of the end user, the consumer, often food research centred around disciplines such as chemical engineering neglect its final purpose in the design of food microstructure for foods with functionality. The results presented in this thesis highlight the value of combining disciplines and encourages the adoption of this approach. The main conclusions from each results chapter of this thesis are summarised in the following sections, followed by an overall thesis.

7.1. ***Chapter 3*** – Influence of oil droplet size on the tribological properties of oil-in-water emulsion systems and the relation to sensory perception

7.1.1. Conclusions

- The oil droplet size investigated was shown to be important in regards to both sensory perception and tribological behaviour, and extends previous understanding only considering oil droplet sizes between 0.2 - 6 µm. This approach additionally highlighted why previously investigated droplet size ranges may have only shown minor non-significant differences in sensory perception.

- Increasing oil droplet size resulted in a negative linear relationship with friction coefficient. The relationship was concluded to be a result of distinct lubrication mechanisms. With increasing droplet size, greater deformability and droplet break-up, for droplets greater than 20 µm was observed, which was concluded to be driving the linear decrease in friction coefficient. The emulsion with 0.2 µm sized droplets, did not fit the trend, this was concluded to be a result of 0.2 µm droplets unique ability to be entrained within surface asperities, as a hysteresis was observed, with lower friction coefficient on reducing entrainment speed sweeps, than on increasing sweeps.

- Oil droplet size had an effect on the perception of Smoothness, Slipperiness, Oiliness and Creaminess. The mechanism in which this is driven and to what degree was investigated in chapter 4 and 5.

- Friction was shown to be involved in the perception of certain attributes (Oiliness and Slipperiness), further demonstrating the value of tribological techniques in understanding the physical make-up of sensory attributes. Limitations in only using these techniques were highlighted, with certain attributes not being fully explained by the physical properties measured.

7.2. ***Chapter 4*** – Iso-viscous and Iso-friction oil-in-water emulsions: The contribution of viscosity and friction in sensory perception

7.2.1. Conclusions

- To the best of our knowledge, this is the first work to consider the design and use of iso-friction emulsions, with all similar prior work only using iso-viscous systems. Through the use of these novel systems, new insights were gained into the influences involved in certain sensory attributes within emulsion systems, and previous understanding surrounding certain sensory attributes was confirmed.

- Results confirmed that Thickness is a sensory interpretation of viscosity.

- Smoothness perception was concluded to be mediated by the behaviour of oil droplets during oral processing, which varied as a function of droplet size.

- Creaminess perception was shown to be influenced by a combined influence of viscosity and droplet behaviour, the sensory interpretation of these two physical variables being Thickness and Smoothness. This is in agreement with Kokini’s Creaminess predication equation that showed that Creaminess is a combination of these two independent sensory variables (Thickness and Smoothness).

7.3. Chapter 5- Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour

7.3.1. Conclusions

- Decreasing oil droplet size significantly increased flavour intensity and perception of Thickness and Creaminess. Smoothness also significantly decreased with increasing droplet size, apart from droplets of 50 µm indicating a polynomial trend. The conclusions of how these behaviours occurred can be taken from conclusions within Chapter 4, however caution should be taken in its interpretation as Chapter 4 samples additionally contained a hydrocolloid.

- Decreasing oil droplet size also significantly increased Liking (hedonic properties) and expectations of Expected Filling (satiation) and decreased Expected Hunger in 1 hours' time (Satiety). For, hedonic properties and both expectations of food intake, this relationship was shown to be largely a result of their strong correlations with Creaminess, which was hypothesised to be driven by the association between Creaminess and energy content. This finding in expectations was further investigated in actual eating behaviour in chapter 6.

7.4. Chapter 6 - Emulsion oil droplet size significantly effects satiety: A pre-ingestive approach

7.4.1. Conclusions

- Participants consumed significantly less at the *ad libitum* lunch after the preload containing 2 µm oil droplets than after the 50 µm preload, with an average reduction of 12%. This result reflects the expectations of satiety (Chapter 5) which formed the hypothesis.

- However, despite the significant differences in *ad libitum* intake, no significant differences in sensory characteristics were noted, despite previous data suggesting that there would be a difference in perception (Chapter 5). This was concluded to be a result of the difference in the protocols used as the protocol in chapter 5, promoted sample assessment in a more analytical manner, with samples consumed sequentially in one session, using sensory attributes defined via a description reference. Whereas within chapter 6, samples were assessed for sensory attributes at least 48 hours apart, with participants' practicing free-living behaviour between test days, this method would therefore have hindered participant's ability to draw cross-comparisons between sensory attributes of the preloads unlike the sensory protocol used in chapter 5. Consequently, sensory results presented in chapter 5 would be expected to highlight more pronounced sensory differences between samples because of the comparative nature of the rating task used.

- The potential has been realised, within these model systems, to structure emulsions to promote satiety via pre-ingestive routes, as it was shown the impact that an emulsion has on satiety can be enhanced without producing significantly perceivable differences in sensory properties. Therefore, by introducing a processing step which results in a smaller droplets, emulsion based liquid food products can be produced that enhance satiety, allowing functional redesign unbeknown to the consumer. Additionally to the best of our knowledge, this is the first body of work to consider microstructural engineering approaches to enhance satiety via pre-ingestive routes. As significant results were shown, the findings of this work have important future implications as they highlight the invaluable potential that "engineering" satiety via pre-ingestive routes could offer.

7.5. Overall Conclusion of Thesis

The overarching aim of the work presented within this thesis, was to use a modifiable microstructural property (Oil droplet size) to investigate approaches in which oil-in-water emulsion based foods can be made more satiating yet still provide desirable sensory characteristics. Essentially, this aim was achieved.

The main finding of the work highlighted oil droplet size significantly affects hedonics, food intake behaviour expectations, actual food intake behaviour and the perception of numerous sensory attributes.

It was shown the perception of Creaminess was a strong hedonic indicator, but interestingly it was also shown to significantly induce greater expectations of satiety and satiation. Structurally, Creaminess significantly increased with decreasing oil droplet size. Results from studies assessing the sensory properties of emulsions, strongly suggested that the mechanism in which oil droplet size modified Creaminess was through altered texture and mouthfeel. Instrumental characterisation of the emulsion samples indicated that this was a result of a combined influence of viscosity and droplet behaviour during oral processing, the sensory interpretation of these two physical variables being Thickness and Smoothness, reflecting Kokini's Creaminess predication equation.

In relation to eating behaviour, through a preload study using the Sussex ingestion pattern monitor, expectations of satiety and satiation were shown to be reflected in actual food intake behaviour, with smaller oil droplets inducing a significant 12% reduction in food intake at a *ad libitum* meal. As such, throughout this thesis the potential of emulsion structure on synergistically increasing both the satiety and hedonics of an emulsion based foods was realised.

7.6. Future recommendations

This sections highlights areas for further research, based on the conclusions developed from this thesis.

- **Extend the understanding of oil droplet behaviour in lubrication through droplet dynamics**

As within Chapter 3 it was concluded that factors governing droplet dynamics were responsible for the mechanism in which droplets of different sizes lubricate, investigating these factors would be an area to consider for further investigation, to further understand how droplets, and particularly droplet size, can be used to alter lubrication capacity of an emulsion system. This would not only be of interest to the food industry for the development of desired sensory profiles, but numerous other industries in which emulsions are, or could be, used as lubricants. Key droplet dynamic factors of interest would be: The viscosity ratio of the two immiscible phases, achievable through the use of a continuous and/or oil phase of different viscosities, Droplet features such as interfacial tension, achievable by changing the emulsifier or surfactant type, concentration or interfacial behaviour, the flow type or flow conditions, achievable through tribometer type or configuration and the shear applied. Stone (1994) provides a comprehensive review of such droplet dynamic factors which could be further explored.

- **Investigations to further understand oil droplet behaviour within the mouth**

Although a small number of studies have investigated oil droplet behaviour under oral conditions, using both *in vivo* and *in vitro* techniques, understanding is still extremely limited. Although challenging, new studies designed to understand emulsion destabilisation behaviour, during oral processing, in a time-dependent manner and its effect on sensory perception, would be extremely valuable in emulsion design for desired sensory profiles. The importance of this is highlighted by limitations in understanding in both Chapter 3 and especially Chapter 4. Approaches could use a combination of instrumental techniques conformed to oral conditions, such as through the use of oral surfaces, saliva, temperature control and imitated oral movement, and *in vivo* time-dependent techniques such as integrating temporal dominance sensation with emulsion “spit out” analysis, for example analysis of type and degree of emulsion destabilisation, viscosity, saliva dilution. When such techniques are established, as destabilisation of emulsions within the mouth is predominantly a result of flocculation and coalescence, droplet interfacial properties suggest to be a sensible starting point for emulsion microstructural development for specific sensory profiles.

- **Application of findings into food products and more complex model systems for further understanding and development**

As results from this thesis have realised the potential to structure emulsions to promote satiety via pre-ingestive routes, such findings should look to be applied within commercially available food products, in order to be beneficial in reducing the obesogenic food environment. Due to the nature of the model emulsion systems used throughout Chapter 5 and 6, results are thought to be transferable with immediate application to oil based condiments, such as salad dressings, due to the similarity of these systems with the model systems used throughout this thesis. Further development would be need in order to apply results to beverages, such as smoothies and shakes, and sauces. More complex food structures, such as soft-solids like yoghurt, would need significant further development and validation to ensure such results are still reflected in semi-solid foods. Chapter 4, has already identified methods to increase the complexity of the model systems used, in order to investigate and apply findings in other food models i.e. introduction of hydrocolloid to aqueous phase.

- **Extend understanding of microstructural approaches to enhance pre-ingestive mediated satiety**

Research, carried out in a similar manner to chapter 5 and 6, should now be conducted to investigate other microstructural properties which can be used to mediate satiety via pre-ingestive routes. This approach is also not limited to emulsions, as it seems sensible to suggest such an approach would work in any system whose microstructure is modifiable e.g. gels, foams. As certain studies have already investigated the effect of microstructure on the perception of numerous sensory attributes, such work could be simply extended to see whether said microstructural properties effect satiety via pre-ingestive routes.

- **Integrate microstructural approaches enhancing satiety via both pre-ingestive and post-ingestive routes**

Future studies should consider creating an integrated approach to "engineering" satiety, combining anticipatory physiological regulation responses (Pre-ingestive) and gastric structuring approaches (Post-ingestive). Although this is the first study to realise to potential of microstructural engineering approaches for pre-ingestive mediated satiety, if work of this nature is continued, research can look to integrate pre-ingestive approaches with the already established microstructural engineering approaches which mediate post-ingestive satiety, such as lipolysis, gelation and intra-gastric structuring. Theoretically this could produce very satiating food products, through, for example, creating an emulsion with small oil droplets, which tastes Creamy and Thick, increasing anticipatory physiological regulation responses, but also has a favourable intra-gastric structure and lipolysis rate.

References

Akhtar, M., Murray, B.S., & Dickinson, E. (2006). Perception of creaminess of model oil-in-water dairy emulsions: Influence of the shear-thinning nature of a viscosity-controlling hydrocolloid. *Food Hydrocolloids, 20,* 839-847

Akhtar, M., Stenzel, J., Murray, B.S. & Dickinson, E. (2005). Factors affecting the perception of creaminess of oil-in-water emulsions. *Food Hydrocolloids, 19,* 521-526

Atwater, W.O. & Woods, C.D. (1896). Chemical composition of American food material. *FAO*. 47pp

Arganini, C., Saba, A., Comitato, R., Virgili, F. & Turrini, A. (2012). Gender Differences in food choice and dietary intake in modern western societies. In: J., Maddock, ed. 2012. *Public health - Social and behavioral health.* P86-88

Armand, M., Borel, P., Ythier, P., Dutot, G., Melin, C., Senft, M., Lafont, H. & Lairon, D. (1992). Effects of droplet size, triacylglycerol composition, and calcium on the hydrolysis of complex emulsions by pancreatic lipase - an in vitro study. *Journal of Nutritional Biochemistry, 7 (3),* 333-341

Armand, M., Pasquier, B., Andre, M., Borel, P., Senft, M., Peyrot, J., Salducci, J., Portugal, H., Jaussan, V. & Lairon, D. (1999). Digestion and absorption of 2 fat emulsions with different droplet sizes in the human digestive tract. *American Journal of Clinical Nutrition, 70,* 1096–1106

Bateman, D.N. (1982). Effects of meal temperature and volume on the emptying of liquid from the human stomach. *Journal of Physiology, 331,* 461–467

Bellamy, M., Godinot, N., Mischler, S., Martin, N. & Hartmann, C. (2009). Influence of emulsion composition on lubrication capacity and texture perception. *International Journal of Food Science and Technology, 44,* 1939-1949.

Bellisle, F. (2008). Functional foods and the satiety cascade. *Nutrition Bulletin, 33,* 8-14

Bertenshaw, E.J., Lluch, A., & Yeomans, M.R. (2013). Perceived thickness and creaminess modulates the short-term satiating effects of high-protein drinks. *British Journal of Nutrition, 110,* 578-586

Bilman, E.M., Kleef, E.V., Mela, D.J., Hulshof, T. & van Trijp, H.C.M. (2012). Consumer understanding, interpretation and perceived levels of personal responsibility in relation to satiety-related claims. *Appetite, 59* (3), 912-920

Binks, B. P. (1998). Modern aspects of emulsion science (1st ed.). The Royal Society of Chemistry.

Birch, L.L. & Deysher, M. (1985). Conditioned and unconditioned caloric compensation: evidence for self-regulation of food intake in young children. *Learning and Motivation, 16,* 341–355

Blundell, J. (1999). The control of appetite: basic concepts and practical implications. *Schweiz Med Wochenschr,* 129, 182–188

Blundell, J. (2010). Making claims: Functional foods for managing appetite and weight. *Nature Reviews Endocrinology, 6,* 53-56

Blundell, J. E., Green, S. & Burley, V. (1994). Carbohydrates and human appetite. *American Journal of Clinical Nutrition, 59(3),* 728-734.

Blundell, J. E. & Macdiarmid, J. I. (1997). Fat as a risk factor for overconsumption: satiation, satiety, and patterns of eating. *Journal of the American Dietetic Association, 97(7),* 63-69.

Blundell, J. & Tremblay, A. (1995). Appetite control and energy (fuel) balance. *Nutrition Research Reviews, 8(1),* 225-242

Blundell, J., Rogers, P. & Hill, A. (1987). Evaluating the satiating power of foods: implications for acceptance and consumption. In J. Colms, D. A. Booth, R. M. Pangborn, & O. Raunhardt (Eds.), *Food acceptance and nutrition* (pp. 205-219).

Booth, D.A., Mather, P. & Fuller, J. (1982). Starch content of ordinary foods associatively conditions human appetite and satiation, indexed by intake and pleasantness of starch-paired flavours. *Appetite, 3,* 163–184

Booth, D.A., Lee, M. & McAleavey, C. (1976). Acquired sensory control of satiation in man. *British Journal of Psychology, 67,* 137–147

Borel, P., Armand, M., Pasquier, B., Senft, M., Dutot, G., Melin, C., Lafont, H. & Lairon, D. (1994). Digestion and absorption of tube-feeding emulsions with different droplet sizes and compositions in the rat. *Journal of Parenteral and Enteral Nutrition, 18,* 534–543

Brunstrom, J.M., Collingwood, J., & Rogers, P.J. (2010). Perceived volume, expected satiation, and the energy content of self-selected meals. *Appetite, 55,* (1), 25–29

Brunstrom, J.M., Shakeshaft, N.G. & Scott-Samuel, N.E. (2008). Measuring 'Expected Satiety' in a range of common foods using a method of constant stimuli. *Appetite, 51,* 604-614

Cassady, B. A., Considine, R. V. & Mattes, R. D. (2012). Beverage consumption, appetite, and energy intake: what did you expect? *American Journal of Clinical Nutrition, 95(3),* 587-593

Cecil, J., Castiglione, K., French, S., Francis, J. & Read, N. (1998). Effects of intragastric infusions of fat and carbohydrate on appetite ratings and food intake from a test meal. *Appetite, 30(1),* 65-77

Cecil, J. E., Francis, J. & Read, N.W. (1998). Relative contributions of intestinal, gastric, oro-sensory influences and information to changes in appetite induced by the same liquid meal. *Appetite, 31(3),* 377-390

Chambers, L., Ells, H. & Yeomans, M. R. (2013). Can the satiating power of a high energy beverage be improved by manipulating sensory characteristics and label information?. *Food Quality and Preference, 28,* 271-278

Chambers, L., McCrickerd, K. & Yeomans, M.R. (2015) Optimising foods for satiety. *Trends in Food science and technology, 41,* 149-160

Chen, J. (2009). Food oral processing: A review. *Food Hydrocolloids, 23,* 1–15

Chen, J. (2014). Food oral processing: Some important underpinning principles of eating and sensory perception. *Food Structure, 1,* 91 -105

Chen, J. & Eaton, L. (2012). Multimodal mechanisms of food creaminess sensation. *Food and Function, 12, (3),* 1265-1270.

Chen, J. & Engelen, L. (2012), Food oral processing. Oxford: Wiley-Blackwell.

Chen, J. & Stokes, J. (2012). Rheology and tribology: Two distinctive regimes of food texture sensation. *Trends in Food Science and Technology, 25, (1),* 4-12.

Chojnicka-Paszun, A., de Jongh, H.H.J. & de Kruif, C.G. (2012). Sensory perception and lubrication properties of milk: Influence of fat content. *International Dairy Journal, 26,* 15–22.

Christensen, C.M. (1979). Oral perception of solution viscosity. *Journal of Texture Studies, 10,* 153-164.

Christensen, C.M. & Casper, L.M. (1987). Oral and Nonoral Perception of Solution Viscosity. *Journal of Food Science, 52, (2),* 445-447.

Chung, C. & McClements, D.J. (2014). Structure–function relationships in food emulsions: Improving food quality and sensory perception. *Food Structure,* 1, 106-126.

Cliff, M. & Noble, A.C. (1990). Time-intensity evaluation of sweetness and fruitiness and their interaction in a model solution. *Journal of Food Science, 55,* 450-454

Cosgrove, T. (2010). Colloid science principles, methods and applications. (2^{nd} ed.). CRC Press

Crum, A.J., Corbin, W.R., Brownell, K.D. & Salovey, P. (2011). Mind over milkshakes: Mindsets, not just nutrients, determine ghrelin response. *Health Psychology, 30(4),* 424–429

Cussler, E. L., Kokini, J.L., Weinheimer, R. L. & Moskowitz, H. R. (1979). Food texture in the mouth. *Food Technology, 33, (10),* 89–92.

Cutler, A. N., Morris, E.R. & Taylor, L.J. (1983). Oral perception of viscosity in fluid foods and model systems. *Journal of Texture Studies, 14(4),* 377–395

Daget, N., Joerg, M. & Bourne, M. (1987). Creamy perception I: In model dessert creams. *Journal of Texture Studies, 18,* 367 -388

Davidson, T.L. & Swithers, S.E. (2004). A Pavlovian approach to the problem of obesity. *International Journal of Obesity, 28*(7), 933-935

Davis, J.D. & Smith, G.P. (2009). The conditioned satiating effect of orosensory stimuli. *Physiology & Behavior*, *97*, 293–303

de Graaf, C. (2012). Texture and satiation: The role of oro-sensory exposure time. *Physiology and Behaviour, 107(4),* 496-501

de Graaf, C., de Jong, L.S. & Lambers, A.C. (1999). Palatability affects satiation but not satiety. *Physiology and Behaviour, 66(4),* 681 - 688

de Graaf, C. & Kok, F.J. (2010). Slow food, fast food and the control of food intake. *Nature Reviews Endocrinology, 6*, 290-293

de Hoog, E.H.A., Prinz, J.F., Huntjens, L., Dresselhuis, D.M. & van Aken, G.A. (2006). Lubrication of oral surfaces by food emulsions: the importance of surface characteristics. *Journal of Food Science, 71, (7),* 337-341.

de Vicente, J., Stokes, J.R. & Spikes, H.A. (2006). Soft lubrication of model hydro-colloids. *Food Hydrocolloids, 20(4),* 483-491.

de Wijk, R., Janssen, A.M. & Prinz, J.F. (2011). Oral movements and the perception of semi-solid foods. *Physiology and Behaviour*, *104, (3),* 423-428.

de Wijk, R. & Prinz, J. (2005). The role of friction in perceived oral texture. *Food Quality and Preference, 16(2),* 121-129

de Wijk, R. & Prinz, J. (2007). Fatty versus creamy sensations for custard desserts, white sauces, and mayonnaises. *Food Quality and Preference, 18,* 641-650.

de Wijk, R.A., Rasing, F. & Wilkinson, C.L. (2003). Texture of semi-solids: Sensory flavour-texture interactions for custard desserts. *Journal of Texture Studies*, *34*, 131-146

Dhar, R. & Wertenbroch, K. (2000). Consumer choice between hedonic and utilitarian goods. *Journal of Marketing research, vol XXXVII,* 60-71

Dhillion, J., Running, C.A., Tucker, R.M. & Mattes, R.D. (2015). Effects of food form on appetite and energy balance. *Food Quality and Preference*

Dickinson, E. (1989). A model of a concentrated dispersion exhibiting bridging flocculation and depletion flocculation. *Journal of Colloid and Interface Science, 132* (1), 274-278.

Dickinson, E. (1992). An Introduction to Food Colloids, Oxford University Press, Oxford.

Dickinson, E. (1999). Caseins in emulsions: interfacial properties and interactions. *International Dairy Journal*, *9*(3-6), 305–312.

Dickinson, E. (2001). Milk protein interfacial layers and the relationship to emulsion stability and rheology. *Colloids and Surfaces B:Bioinerfaces, 20,* 197-210.

Dickinson, E. (2010). Flocculation of protein-stabilized oil-in-water emulsions. *Colloids and Surfaces. B, Biointerfaces*, *81*(1), 130–40.

Dongowski G. (1997). Effect of pH on the in vitro interactions between bile acids and pectin. *Z Lebensm Unters Forsch A*, 205, 185–192

Dresselhuis, D.M., de Hoog, E.H.A., Cohen Stuart, M.A. & van Aken, G.A. (2008a). Application of oral tissue in tribological measurements in an emulsion perception context. *Food Hydrocolloids, 22,* 323–335.

Dresselhuis, D.M., de Hoog, E.H.A., Cohen Stuart, M.A., Vingerhoeds, M.H. & van Aken, G.A. (2008b). The occurrence of in-mouth coalescence of emulsion droplets in relation to perception of fat. *Food Hydrocolloids, 22,* 1170–1183.

Dresselhuis, D.M., Klok, H.J., Stuart, M.A.C., de Vries, R.J., van Aken, G.A. & de Hoog, E.H.A. (2007). Tribology of O/W emulsions under mouth-like conditions: Determinants of friction. *Food Biophysics, 2,* 158–171.

Drewnowski, A. (1990). The new fat replacements - A strategy for reducing fat consumption. *Postgraduate Medicine, 87(6),* 111-121

Drewnowski, A. (1998). Energy density, palatability and satiety: Implications for weight control. *Nutrition reviews, 56(12),* 347 – 353

Drewnowski, A. & Specter, S. (2004). Poverty and obesity: The role of energy density and energy costs. *American Journal of Clinical Nutrition. 79,* 6–16.

Edgar, W.M. (1990). Saliva and dental health. *British Dental Journal, 169(3–4),* 96–98

Elmore, J.R., Heymann, H., Johnson, J., & Hewett, J.E. (1999). Preference mapping: Relating acceptance of creaminess to a descriptive sensory map of a semi-solid. *Food Quality and Preference, 10,* 465–475

Erni, P., Fischer, P. & Windhab, E. J. (2005). Deformation of single emulsion drops covered with a viscoelastic adsorbed protein layer in simple shear flow. *Applied physics letter,* Volume 87.

Ertekin, C. & Aydogdu, I. (2003) Neurophysiology of Swallowing. *Clinical Neurophysiology, 114, (12),* 2226-2244.

Evans, J.D. (1996). Straightforward statistics for the behavioural sciences. Brooks/Cole Publishing. Pacific Grove, CA.

Feldman, M. & Richardson, C.T. (1986). Role of Thought, Sight, Smell, and Taste of Food in the Cephalic Phase of Gastric-Acid Secretion in Humans. *Gastroenterology, 90*(2), 428-433

Floury, J., Legrand, J., & Desrumaux, A. (2004). Analysis of a new type of high pressure homogeniser. Part B. Study of droplet break-up and recoalescence phenomena. *Chemical Engineering Science, 59,* 1285–1294

Foltz, M., Maljaars, J., Schuring, E. A., van derWal, R. J., Boer, T., Duchateau, G. S., *et al.* (2009). Intragastric layering of lipids delays lipid absorption and increases plasma CCK but has minor effects on gastric emptying and appetite. *American Journal of Physiology – Gastrointestinal and Liver Physiology, 296* (5), 982-991

Fortes, L.D.S., Kakeshita, I.S., Almeida, S.S., Gomes, A.R. & Ferreira, M.E.C. (2014). Eating behaviours in youths: A comparison between female and male athletes and non-athletes. *Scandinavian Journal of Medicine and Science in Sports, 24* (1), 62-68.

Foster, K.D., Grigor, J.M.V., Cheong, J.N., Yoo, M.J.Y., Bronlund, J. & Morgenstern, M. P. (2011). The role of oral processing in dynamic sensory perception. *Journal of Food Science, 76*, 49-61.

Frank, R.A. & Byram, J. (1988). Taste-smell interactions are tastant and odorant dependent. *Chemical Senses, 13,* 445-455

Frank, R.A., Ducheny, K. & Mize, S.J.S. (1989). Strawberry odor, but not red color, enhances the sweetness of sucrose solutions. *Chemical Senses, 14,* 371-377

Frost, M.B. & Janhoj, T. (2007). Understanding creaminess. *International Dairy Journal, 17,* 1298-1311

Gabriele, A., Spyropoulos, F., & Norton, I. T. (2010). A conceptual model for fluid gel lubrication. *Soft Matter, 6(17),* 4205-4213.

Garrec, D.A., Frasch-Melnik, S., Henry, J.V.L., Spyropoulos, F. & Norton, I.T. (2012). Designing colloidal structures for micro and macro nutrient content and release in foods. *Faraday Discussions, 158,* 37-49.

Garrec, D.A. & Norton, I.T. (2013). Kappa carrageenan fluid gel material properties. Part 2: Tribology. *Food Hydrocolloids, 33 (1),* 160-167.

Golding, M. & Wooster, T.J. (2010). The influence of emulsion structure and stability on lipid digestion. *Current Opinion in Colloid & Interface Science, 15,* 90-101

Golding, M., Wooster, T.J., Day, L., Xu, M., Lundin, L., Keogh, J. & Clifton, P. (2011). Impact of gastric structuring on the lipolysis of emulsified lipids. *Soft Matter, 7,* 3513–3523

Guinard, J.X. & Mazzucchelli, R. (1996). The sensory perception of texture and mouthfeel. *Trends in Food Science & Technology, 7,* 213–219

Gunstone, F.D., Harwood, J.L. & Dijkstra, A.J. (2007). The Lipid Handbook (3rd ed.). CRC Press

Hamrock, B. J. (1994). Fundamentals of fluid film lubrication. London: McGraw-Hill.

Heath, M.R. (2002). The oral management of food: The bases of oral success and for understanding the sensations that drive us to eat. *Food Quality and Preference, 13,* 453–461

Hetherington, M. M., Cunningham, K., Dye, L., Gibson, E. L., Gregersen, N. T., Halford, J. C. G., Lawton, C.L., Lluch, A., Mela, D.J. & van Trijp, H.C.M. (2013). Potential benefits of satiety to the consumer: scientific considerations. *Nutrition Research Reviews, 26(1),* 22-38.

Himaya, A., Fantino, M., Antoine, J.M., Brondel, L. & Louis-Sylvestre, J. (1997). Satiety power of dietary fat: a new appraisal. *American Journal of Clinical Nutrition, 65,* 1410-1408

Hoad, C.L., Rayment, P., Spiller, R.C., Marciani, L., Alonso, B.D., Traynor, C. & Gowland, P.A. (2004). In vivo imaging of intragastric gelation and its effect on satiety in humans. *Journal of Nutrition, 134*(9), 2293-2300

Hogenkamp, P. S., Brunstrom, J.M., Stafleu, A., Mars, M. & de Graaf, C. (2012). Expected satiation after repeated consumption of low- or high-energy-dense soup. *British Journal of Nutrition, 108*(1), 182–190

Hogenkamp, P.S., Stafleu, A., Mars, M., Brunstrom, J.M. & de Graaf, C. (2011). Texture, not flavour, determines expected satiation of dairy products. *Appetite, 57(3),* 635–641

Holt, S.H.A., Brand Miller, J.C., Petocz, P. & Farmakalidis, E. (1995). A satiety index of common foods. *European Journal of Clinical Nutrition, 49,* 675-690

Hougardy H, Dahl W and Grabke H. J (2012): Steel, 5. Microstructures, Properties and Adjustement of Properties. In: *Ullmann's encyclopedia of industrial chemistry.* s.l.:Wiley, 271-334.

Humphrey, S.P. & Williamson, R.T. (2001). A review of saliva: Normal composition, flow and function. *Journal of Prosthetic Dentistry, 85,* 162–169

Hutchings, J.B. & Lillford, P.J. (1988). The perception of food texture – The philosophy of the break down path. *Journal of Texture Studies,19,* 103-115.

Ilg J, Rupitsch S. J, Sutor A and Lerch R (2012): Determination of Dynamic Material Properties of Silicone Rubber Using One-point Measurments and Finite Element Simulations. *IEEE transactions on instrumentation and measurement* 61(11): 3031-3038

Jenkins, G.N. (1978). The physiology and biochemistry of the mouth (4th ed.). Oxford: Blackwell.

Jequier, E. (1984). Energy expenditure in obesity. *Clinics in Endocrinology and Metabolism, 13,* 563-580

Jowitt, R. (1974). The terminology of food texture. *Journal of Texture Studies, 5,* 351–358

Keogh, J.B., Wooster, T.J., Golding, M., Day, L., Otto, B. & Clifton, P.M. (2011). Slowly and rapidly digested fat emulsions are equally satiating but their triglycerides are differentially absorbed and metabolized in human. *The Journal of Nutrition,* 141-145 (5), 809-815

Kilcast, D. & Clegg, S. (2002). Sensory perception of creaminess and its relationship with food structure. *Food Quality and Preference, 13,* 609-623

Kissileff, H. R., Kilngsberg, G. & Van Italie, T.B. (1980). Universal eating monitor for continuous recording of solid or liquid consumption in man. *American Journal of Physiology, 238,* R14–R22.

Kilcast, D. & Clegg, S. (2002). Sensory perception of creaminess and its relationship with food structure. *Food Quality and Preference, 13,* 609-623.

Kokini, J.L. (1987). The physical basis of liquid food texture and texture-taste interactions. *Journal of Food Engineering, 6, (1),* 51-81.

Kokini, J.L. & Cussler, E. L. (1983). Predicting the texture of liquid and melting semi-solid foods. *Journal of Food Science, 48,* 1221-1225.

Kokini, J.L., Kadane, J. & Cussler, E. L. (1977). Liquid texture perceived in the mouth. *Journal of Texture Studies, 8,* 195-218.

Kokini, J.L., Poole, M., Mason, P., Miller, S. & Stier, E. F. (1984). Identification of key textural attributes of fluids and semi-solid foods using regression analysis. *Journal of Food Science, 49,* 47–51.

Koseki, M., Kitabatake, N., Doi, E., Yasuno, T., Ogino, S., Kazama, M., *et al.* (1987). Binding of taurocholate by pectin in the presence of calcium ions. *Journal of Food Science,* 52:1744–1745

Laughton, W.B. & Powley, T.L. (1987). Localization of efferent function in the dorsal motor nucleus of the vagus. *American Journal of Physiology, 252* (1 Pt 2), R13–R25

Lavin, J., French, S., Ruxton, C. & Read, N. (2002). An investigation of the role of oro-sensory stimulation in sugar satiety. *International Journal of Obesity, 26(3),* 384-388

Ledeboer, M., Masclee, A.A., Biemond, I. & Lamers, C.B. (1999). Differences in cholecystokinin release and gallbladder contraction between emulsified and nonemulsified long-chain triglycerides. *Journal of Parenteral and Enteral Nutrition, 23* (4), 203-206.

Lee, L. L., Hancocks, R. D., Noble, I. & Norton, I. T. (2014). Production of water-in-oil nanoemulsions using high pressure homogenisation: A study on droplet break-up. *Journal of Food engineering, 131,* 33-37.

Lenfant, F., Loret, C., Pineau, N., Hartmann, C. & Martin, N (2009). Perception of oral food breakdown. The concept of sensory trajectory. *Appetite, 52,* 659–667.

Le Magnen. J (1982) Neurobiology of Feeding and Nutrition. Academic Press, San Diego

Lett, A.M., Norton, J.E. and Yeomans, M.R. (2016). Emulsion oil droplet size significantly affects satiety: A pre-ingestive approach**.** .*Appetite, 96*, 18-24

Lett, A.M., Yeomans, M.R., Norton, I.T. and Norton, J.E. (2016). Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour. *Food Quality and Preference, 47*, 148-155

Levine, M.J., Reddy, M.S., Tabak, L.A., Loomis, R.E., Bergey, E.J., Jones, P.C., Cohen, R.E., Stinson, M.W. and Al-Hashimi, I. (1987). Structural aspects of salivary glycoproteins. *Journal of Dental Researches, 66,* 436–441

Livingstone, B.E., Robson, P.J., Welch, R.W., Burns, A.A., Burrows, M.S. & McCormack, C. (2000). Methodological issues in the assessment of satiety. *Scandinavian Journal of Nutrition, 44,* 98-103

Ludin, L., Golding, M. & Wooster, T.J. (2008). Understanding food structure and function in developing food for appetite control. *Nutrition & Dietetics, 65(3),* 79-85.

Mattes, R.D. & Rothacker, D. (2001). Beverage viscosity is inversely related to postprandial hunger in humans. *Physiology and Behaviour, 74(4-5),* 551-557

Maljaars, P., van derWal, R., Haddeman, E., Peters, H., Beindorff, C. & Masclee, A. (2008). Effect of droplet size of a fat emulsion delivered in the small intestine on satiety and food intake in healthy volunteers. *European Journal of Gastroenterology & Hepatology, 20* (7),

Maljaars, P.W.J., van der Wal, R.J.P, Wiersma, T., Peters, H.P.F., Haddeman, E. & Masclee, A.A.M. (2012). The effect of lipid droplet size on satiety and peptide secretion is intestinal site-specific. *Clinical Nutrition, 31,* 535–542

Malone, M.E., Appelqvist, I.A.M. & Norton, I.T. (2003). Oral behaviour of food hydrocolloids and emulsions. Part 2. Taste and aroma release. *Food Hydrocolloids, 17(6),* 775 - 784

Marciani, L., Faulks, R., Wickham, M.S.J., Bush, D., Pick, B., Wright, J., *et al.* (2009). Effect of intragastric acid stability of fat emulsions on gastric emptying, plasma lipid profile and postprandial satiety. *British Journal of Nutrition, 101*, 919-928.

Marciani, L., Wickham, M. S., Bush, D., Faulks, R.,Wright, J., Fillery-Travis, A. J., *et al.* (2006). Magnetic resonance imaging of the behaviour of oil-in-water emulsions in the gastric lumen of man. *British Journal of Nutrition, 95*,331-339.

Marciani, L., Wickham, M., Singh, G., Bush, D., Pick, B., Cox, E., *et al.* (2007). Enhancement of intragastric acid stability of a fat emulsion meal delays gastric emptying and increases cholecystokinin release and gallbladder contraction. *American Journal of Physiology, 292*, 1607-1613

McClements, D. J. (1998). Food Emulsions: Principles, Practices, and Techniques (1st ed.). CRC Press.

McClements, D. J. (2005). Food Emulsions: Principles, Practices, and Techniques (2nd ed.). CRC Press.

McClements, D. J. (2007). Critical review of techniques and methodologies for characterization of emulsion stability. *Critical Reviews in Food Science and Nutrition, 47*(7), 611-649.

McClements, D. J. (2009). *Biopolymers in Food Emulsions*. *Modern Biopolymer Science* (1st ed). Elsevier Inc.

McCrickerd, K., Chambers, L., Brunstrom, J.M. & Yeomans, M.R. (2012). Subtle changes in the flavour and texture of a drink enhance expectations of satiety. *Flavour, 1(20)*

McCrickerd, K., Chambers, L. & Yeomans, M.R. (2014). Does modifying the thick texture and creamy flavour of a drink change portion size selection and intake? *Appetite, 73*, 114-120

McCrickerd, K., Chambers, L., & Yeomans, M.R. (2014). Fluid or Fuel? The context of consuming a beverage is important for satiety. *PLoS ONE, 9(6)*

McCrickerd, K., Lensing, N. & Yeomans, M.R. (2015). The impact of food and beverage characteristics on expectations of satiation, satiety and thirst. *Food quality and preference, 44*, 130-138

Medicis, S. W. & Hiiemae, K. H. (1998). Natural bite sizes for common foods. *Journal of Dental Research, Special Issue A, 77,* 295

Mela, D.J. (1988). Sensory assessment of fat content in fluid dairy products. *Appetite*, *10,* 37-44.

Mela, D.J., Langley, K.R. & Martin, A. (1994). Sensory assessment of fat content: Effect of emulsion and subject characteristics. *Appetite, 22,* 67-81

Meullenet, J.F., Lyon, B.G., Carpenter, J.A. & Lyon, C.E. (1998). Relationship between sensory and instrumental texture profile attributes. *Journal of Sensory Studies, 13,* 77-93

Mills, T. (2011). Devlopment of in-vitro mouth methods for studying oral phenomena.. *PhD Thesis*. The University of Birmingham

Mills, T., Norton, I.T. & Bakalis, S. (2013). Development of tribology equipment to study dynamic processes. *Journal of Food Engineering, 114,* 384-390

Mills, T., Koay, A. & Norton, I.T (2013). Fluid gel lubrication as a function of solvent quality. *Food Hydrocolloids, 32,* 172-177

Moore, P.B., Langley, K., Wilde, P.J., Fillery-Travis, A. & Mela, D.J. (1998). Effect of emulsifier type of sensory properties of oil-in-water emulsions. *Journal of Science, Food and Agriculture, 76,* 469-476

Morris, E.R., Richardson, R.K. & Taylor, L.J. (1984). Correlation of the perceived texture of random coil polysaccharide solutions with objective parameters. *Carbohydrate Polymers, 4,* 175-191.

Morris, E.R. & Taylor, L.J. (1982). Oral perception of fluid viscosity. *Progress in Food and Nutrition Science, 6,* 285-296.

Muller-Riemenschneider, F., Reinhold, T., Berghofer, A., & Willich, S. N. (2008). Health-economic burden of obesity in Europe. *European Journal of Epidemiology. 23,* 499–509

Mun, S., Decker, E.A. & McClements, D.J. (2007). Influence of droplet characteristics on the formation of oil-in-water emulsions stabilized by surfactant-chitosan layers. *Food Research International, 40,* 770–781

Norton, I., Moore, S. & Fryer, P. (2007). Understanding food structuring and breakdown: Engineering approaches to obesity. *Obesity Reviews, 8,* 83–88

Ogawa, S., Decker, E.A. & McClements, D.J. (2003). Influence of environmental conditions on the stability of oil in water emulsions containing droplets stabilized by lecithin–chitosan membranes. *Journal of Agriculture and Food Chemistry, 51*, 5522–5527

Ostwald, W. (1910). Beiträge zur Kenntnis der Emulsionen. *Zeitschrift Für Chemie Und Industrie Der Kolloide*, *6*(2), 103–109.

Pascua, Y., Koc, H. & Foegeding, E.A. (2013). Food structure: roles of mechanical properties and oral processing in determining sensory texture of soft materials. *Current Opinion in Colloid & Interface Science, 18,* 324-333.

Perrigue, M., Carter, B., Roberts, S.A. & Drewnowski, A.(2010). A Low-Calorie Beverage Supplemented with Low-Viscosity Pectin Reduces Energy Intake at a Subsequent Meal. *Journal of Food Science, 75*(9), 300-305

Peters, H.P.F., Bouwens, E.C.M., Schuring, E.A.H., Haddeman, E., Velikov, K.P. & Melnikov, S.M. (2014). The effect of submicron fat droplets in a drink on satiety, food intake, and cholecystokinin in healthy volunteers. *European Journal of Nutrition, 53 (3),* 723-729

Picciano, M.F. (1998). Human milk: Nutritional aspects of a dynamic food. *Biology of the Neonate*, *74*(2), 84–93

Pickering, S. U. (1907). CXCVI.Emulsions. *Journal of the Chemical Society, Transactions*, *91*, 2001.

Pichot, R. (2010). Stability and Characterisation of Emulsions in the presence of Colloidal Particles and Surfactants. *PhD Thesis*. The University of Birmingham.

Pickering, S. U. (1907). CXCVI.Emulsions. *Journal of the Chemical Society, Transactions*, *91*, 2001.

Piqueras-Fiszman, B. & Spence, C. (2012). The weight of the container influences expected satiety, perceived density, and subsequent expected fullness. *Appetite*, *58* (2), 559–562

Pomerleau, J., Mckee, M., Lobstein, T. & Knai, C. (2003). The burden of disease attributable to nutrition in Europe. *Public Health Nutrition. 6*, 453–461

Power, M. L. & Schulkin, J. (2008). Anticipatory physiological regulation in feeding biology: cephalic phase responses. *Appetite, 50(2-3),* 194-206

Prentice, A.M., & Jebb, S.A. (2003). Fast foods, energy density and obesity. A possible mechanistic link. *Obesity Reviews, 4,* 187–194

Quemada, D. & Berli, C. (2002). Energy of interactions in colloids and its implications in rheological modelling. *Advances in Colloid and Interface Science, 98,* 51-85.

Richardson-Harman, N.J., Stevens, R., Walker, S., Gamble, J., Miller, M., Wong, M. & McPhearson, A. (2000). Mapping consumer perceptions of creaminess and liking for liquid dairy products. *Food Quality and Preference, 11,* 239 -246

Rodin, J. (1985). Insulin levels, hunger, and food intake - an example of feedback loops in body-weight regulation. *Health Psychology, 4*(1), 1-24

Roller, S. & Jones, S.A. (eds). *Handbook of Fat Replacers.* CRC Press: Boca Raton, 2001.

Rolls, B.J., Kim-Harris, S., Fischman, M.W., Foltin, R.W., Moran, T.H. & Stoner, S.A. (1994). Satiety after preloads with different amounts of fat and carbohydrate: implications for obesity. *American Journal of Clinical Nutrition, 60* (4), 476-487

Rolls, E.T. (2007). Understanding the mechanisms of food intake and obesity. *Obesity reviews, 8,* (1), 67-72

Sakata, T., Yoshimatsu, H., Masaki, T. & Tsuda, K. (2003). Anti-obesity actions of mastication driven by histamine neurons in rats. *Experimental Biology and Medicine, 228,* 1106–1110

Sarkar A, Goh, K.K.T. & Singh, H. (2009). Colloidal stability and interactions of milk-protein- stabilized emulsions in an artificial saliva. *Food Hydrocolloids, 23,* 1270–1278

Scott, C.L. and Downey, R.G. (2007). Types of food aversions: animal, vegetable, and texture. Journal of Psychology, 141(2), 127–134

Seimon, R.V., Wooster, T., Otto, B., Golding, M., Day, L., Little, T.J., Horowitz, M., Clifton, P.M. & Feinle-Bisset, C. (2009). The droplet size of intraduodenal fat emulsions influences antropyloroduodenal motility, hormone release, and appetite in healthy males. *American Journal of Clinical Nutrition, 89,* 1729–1736

Selway, N. & Stokes, J.R. (2013). Insights into the dynamics of oral lubrication and mouthfeel using soft tribology: Differentiating semi-fluid foods with similar rheology. *Food Research International, 54,* 423-431

Shama, F. & Sherman, P. (1973a). Identification of stimuli controlling the sensory evaluation of viscosity. II Oral methods. *Journal of Texture Studies, 4,* 111-118

Shama, F. & Sherman, P. (1973b). Variation in stimuli associated with oral evaluation of the viscosities of glucose solutions. *Journal of Texture Studies, 4,* 254-262

Silletti, E., Vingerhoeds, M.H., Norde, W. & van Aken, G.A. (2007). The role of electrostatics in saliva-induced emulsion flocculation. *Food Hydrocolloids, 21,* (4), 596-606

Singh, H., Ye, A. & Horne, D. (2009). Structuring food emulsions in the gastrointestinal tract to modify lipid digestion. *Progress in Lipid Research, 48(2),* 92-100

Smeets, P. A. M., Erkner, A. & De Graaf, C. (2010). Cephalic phase responses and appetite. *Nutrition Reviews, 68(11),* 643-655

Smith, C.H., Logemann, J.A., Burghardt, W.R., Carrell, T.D. and Zecker, S.G. (1997). Oral sensory discrimination of fluid viscosity. *Dysphagia, 73,* 68-73.

Smith, C.H., Logemann, J.A., Burghardt, W.R., Zecker, S.G. and Rademaker, A.W. (2007). Oral and oropharyngeal perceptions of fluid viscosity across the age span. *Dysphagia, 21, (4),* 209-217.

Somasundaran, P. (2006). Encyclopaedia of Surface and Colloid Science (1st ed.). CRC Press

Sonne, A., Busch-Stockfisch, M., Wiess, J. & Hinrichs, J. (2014). Improved mapping of in-mouth creaminess of semi-solid dairy products by combining rheology, particle size and tribology data. *LWT – Food Science and Technology, 59,* 342-347

Spikes, H.A. (1997). Mixed Lubrication – An Overview. *Lubrication Science, 9,* 221-253

Stanley, N. I. & Taylor, L. J. (1993). Rheological basis of oral characteristics of fluid and semi-solid foods: A review. *Acta Psychologica, 84,* 79–92.

Stone, H.A. (1994). Dynamics of drop deformation and breakup in viscous fluids. *Annual Review of Fluid Mechanics, 26,* 65-102.

Stokes, J.R., Boehm, M.W. & Baier, S.K. (2013). Oral processing, texture and mouthfeel: From rheology to tribology and beyond. *Current opinion in Colloid & Interface Science, 18, (4),* 349-359.

Szczesniak, A.S. (1963). Classification of textural characteristics. *Journal of Food Science, 28,* 385–389

Szczesniak, A. S. (1987). Relationship of texture to food acceptance and nutrition. In: J. Sohns, D. A. Booth, R. M. Pangbom and O. Raunhardt, *Food acceptance and nutrition*. New York: Academic Press

Tadros, T., Izquierdo, P., Esquena, J., & Solans, C. (2004). Formation and stability of nano-emulsions. *Advances in Colloid and Interface Science, 108–109*(0), 303–318.

Tarrega, A. & Costell, E. (2006). Effect of inulin addition on rheological and sensory properties of fat-free starch-based dairy desserts. *International Dairy Journal, 16, (9),* 1104-1112.

Taylor, G.I. (1934). The formation of emulsions in definable fields of flow. *Proc. R. Soc. London, Series A,* 146, 501-523.

Taylor, P. (1998). Ostwald ripening in emulsions. *Advances in Colloid and Interface Science, 75*, 107-163.

Taylor, A.J. & Linforth, R.S.T. (2010). Food Flavour Technology. (2nd ed.). Blackwell publishing

Wood, F.W. (1968). Psychophysical studies on the consistency of liquid foods. *S.C.I Monograph, 27,* 40-49.

Wardle, J. (1987). Eating style a validation eating behaviour study of the Dutch eating behaviour questionnaire in normal subjects and women with eating disorders. *Journal of Psychosomatic Research, 31 (2),* 161-169.

Weenen, H., Jellema, R.H., & de Wijk, R. A. (2005). Sensory sub- attributes of creamy mouthfeel in commercial mayonnaises, custard desserts and sauces. *Food Quality and Preference, 16*, 163–170.

Valentin, D., Chrea, C. & Nguyen, D.H. (2006). Taste–Odour interaction in sweet taste perception. In: *W. J. Spillane (Ed.), Optimising sweet taste in foods*. Woodhead Publishing Limited, 66–84

van Aken, G.A. Bomhof, E., Zoet, F.D., Verbeek, M. & Oosterveld, A. (2011) Differences in in vitro gastric behaviour between homogenized milk and emulsion stabilised by Tween80, whey protein, or whey protein and caseinate. *Food Hydrocolloids, 25 (4),* 781-788

van Aken, G.A. Vingerhoeds, M.H. & de Hoog, E.H.A. (2007) Food colloids under oral conditions. *Current Opinion in Colloid & Interface Science, 12 (4-5),* 251-262

van Aken, G.A., Vingerhoeds, M.H. & de Wijk, R.A. (2011). Textural perception of liquid emulsions: Role of oil content, oil viscosity and emulsion viscosity. *Food Hydrocolloids, 20,* 789-796 van Keelf, E., van Trijp, J.C.M., van den Borne, J.J.G.C. & Zondervan, C. (2012). Successful Development of Satiety Enhancing Food Products: Towards a Multidisciplinary Agenda of Research Challenges. *Critical Reviews in Food Science and Nutrition 52(7),* 611-628

van Ruth, S.M., King, C. & Giannouli, P. (2002). Influence of lipid fraction, emulsifier fraction, and mean particle diameter of oil-in-water emulsions on the release of 20 aroma compounds. *Journal of Agricultural and Food Chemistry, 50,* 2365– 2371.

van Strien, T., Frijters, J.E.R., Bergers, G.P.A. & Defares, P.B. (1986) The Dutch eating behaviour questionnaire (DEBQ) for assessment of restrained, emotional and external eating behaviour. *International Journal of Eating Disorders, 5(2),* 295-315

Vega, C. and Mercade-Prieto, R. (2011). Culinary Biophysics: on the nature of the 6xºc egg. *Food Biophysics, 6, (1),* 152-159.

Vinarov, Z., Tcholakova, S., Damyanova, B., Atanasov, Y., Denkov, N.D., Stoyanov, S.D. *et al* (2012). Effects of emulsifier charge and concentration on pancreatic lipolysis: 2. Interplay of emulsifiers and biles. *Langmuir,* 28, 12140-12150

Vincent, J.F.V. & Lillford, P.J. (1991). Feeding and the Texture of Food. The University Press, Cambridge, UK.

Vingerhoeds, M.H., Blijdenstein, T.B.J., Zoet, F.D. & van Aken, G.A. (2005). Emulsion flocculation induced by saliva and mucin. *Food Hydrocolloids, 19, (5),* 915-922.

Vingerhoeds, M.H., de Wijk, R.A., Zoet, F.D., Nixdorf, R.R. & van Aken, G.A. (2008). How emulsion composition and structure affect sensory perception of low-viscosity model emulsions. *Food Hydrocolloids, 22,* 631-646

Vingerhoeds, M.H., Silletti, E., de Groot, J., Schipper, R.G. & van Aken, G.A. (2009). Relating the effect of saliva-induced emulsion flocculation on rheological properties and retention on the tongue surface with sensory perception. *Food Hydrocolloids, 23, (3),* 773-785.

van Kleef, E., Van Trijp, J.C.M., Van Den Borne, J.J.G.C. & Zondervan, C. (2012). Successful development of satiety enhancing food products: Towards a multidisciplinary agenda of research challenges. *Critical Reviews in Food Science and Nutrition, 52(7),* 611-628

van Strien, T., Frijters, J.E.R., Bergers, G.P.A. & Defares, P.B (1986) The Dutch eating behaviour questionnaire (DEBQ) for assessment of restrained, emotional and external eating behaviour. *International Journal of Eating Disorders, 5(2),* 295-315

Walstra, P. (1993). Principles of emulsion formation. *Chemical Engineering Science*, *48*(2), 333–349.

Wardle, J. (1987). Eating style a validation eating behavior study of the Dutch eating behavior questionnaire in normal subjects and women with eating disorders. *Journal of Psychosomatic Research, 31 (2),* 161-169

Weenen, H., Jellema, R.H., & de Wijk, R. A. (2005). Sensory sub- attributes of creamy mouthfeel in commercial mayonnaises, custard desserts and sauces. *Food Quality and Preference, 16,* 163–170.

Weijzen, P. L., Smeets, P. A. & de Graaf, C. (2009). Sip size of orangeade: Effects on intake and sensory-specific satiation. *The British Journal of Nutrition, 102*(7), 1091–1097

Wickham, M., Garrood, M., Leney, J., Wilson, P.D.G. & Fillery-Travis, A (1998). Modification of a phospholipid stabilized emulsion interface by bile salt: effect on pancreatic lipase activity. *Journal of Lipid Research, 39,* 623–632

Wijlens, A.G.M., Erkner, A., Alexander, E., Mars, M., Smeets, P.A.M. & de Graaf, C. (2012). Effects of oral and gastric stimulation on appetite and energy intake. *Obesity, 20,* 2226–2232

Wooley, S.C. & Wooley, O.W. (1973). Salivation to the sight and thought of food: A new measure of appetite. *Psychosomatic Medicine, 35,* 136–142

Wood, F.W. (1968). Psychophysical studies on the consistency of liquid foods. *S.C.I Monograph, 27,* 40-49

World Health Organisation, 2015. *Obesity and Overweight.* [online] Available at http://www.who.int/mediacentre/factsheets/fs311/en/ [Accessed 16th June 2015].

Yeomans, M.R. & Bertenshaw, E.J. (2008). Appetite and food intake: a human experimental perspective. In R.B.S. Harris & R.D. Mattes (Eds.), *Appetite and food intake: behavioral and physiological considerations* (pp. 61–80). Boca Raton: Taylor and Francis.

Yeomans, M.R., Gould, N.J., Leitch, M. & Mobini S. (2009). Effects of energy density and portion- size on development of acquired flavour liking and learned satiety. *Appetite, 52,* 469–78

Yeomans, M.R. & Chambers, L. (2011). Satiety-relevant sensory qualities enhance the satiating effects of mixed carbohydrate-protein preloads. *American Journal of Clinical Nutrition, 94(6),* 1410–1417

Yeomans, M.R., McCrickerd, K., Brunstrom, J.M., & Chambers, L. (2014). Effects of repeated consumption on sensory-enhanced satiety. *British Journal of Nutrition,* 111*(6),* 1137-1144

Yeomans, M.R., Weinberg, L. & James, S. (2005). Effects of palatability and learned satiety on energy density influences on breakfast intake in humans. *Physiology and Behavior, 86,* 487–499

Zhang, J., Xu, S. & Li, W. (2012). High shear mixers: A review of typical applications and studies on power draw, flow pattern, energy dissipation and transfer properties. *Chemical Engineering ad Processing: Process Intensification, 57-58*, 25-41.

Zhu, Y., Hsu, W.H. & Hollis, J.H. (2013). The Impact of Food Viscosity on Eating Rate, Subjective Appetite, Glycemic Response and Gastric Emptying Rate. *Plos One*, *8*, (6), e67482.

Zijlstra, N., de Wijk, R.A., Mars, M., Stafleu, A. & de Graaf, C. (2009a). Effect of bite size and oral processing time of a semi-solid food on satiation. *American Journal of Clinical Nutrition, 90*, 269–75

Zijlstra, N., Mars, M., de Wijk, R.A., Westerterp-Plantenga, M.S., Holst, J.J. & de Graaf, C. (2009b). Effect of viscosity on appetite and gastro-intestinal hormones. *Physiology and Behaviour, 97(1),* 68–75