The influence of chewing and oral lubrication on satiety using hydrogels

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The candidate confirms that the work submitted is her own, except where work which has formed part of jointly authored publications has been included. The contribution of the candidate and the other authors to this work has been explicitly indicated below. The candidate confirms that appropriate credit has been given within the thesis where reference has been made to the work of others. Details of the jointly authored publications and contributions of each author are outlined on the next page.

Further details of the jointly authored publications in peer-reviewed journals, which chapters they appear in and the contributions of the candidate and the other authors are outlined below.

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The candidate developed the research question and search protocol, conducted the systematic review and, collected and organized the data for the meta-analysis.

The candidate received input from CN and LP to run the meta-analyses and the candidate analysed and interpreted the results. The candidate wrote the manuscript and MMH, SM and AS all contributed to proofreading and provided feedback for revisions of the manuscript.

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The candidate designed the experimental plan and developed the model hydrogels, and ran all the instrumental and sensory characterisation analyses.

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E. M. Krop, M. M. Hetherington, S. Miquel-Kergoat, A. Sarkar (2017). Influence of oral processing on appetite and food intake - A systematic review and meta-analysis. *The 12th Pangborn Sensory Science Symposium*, Providence, USA, 20-24 August 2017.

Emma Krop, Marion Hetherington, Melvin Holmes, Anwesha Sarkar (2018). Relating large deformation rheology and tribology to sensory properties of hydrocolloid gels. *The 17th Food Colloids Conference: Application of Soft Matter Concepts*, Leeds, UK, 18-11 April 2018.

E. M. Krop, M. M. Hetherington, S. Miquel, A. Sarkar (2018). Influence of both chewing and lubrication during oral processing of hydrogels on hunger and energy intake. *The Nutrition Society Summer Conference 2018: Getting the energy balance right*, Leeds, UK, 10-12 July 2018.

Emma Krop, Marion Hetherington, Chandani Nekitsing, Sophie Miquel, Luminita Postelnicu, Anwesha Sarkar (2018). A systematic review and meta-analysis investigating the effects of oral processing on hunger and energy intake. *The 8th European Conference on Sensory and Consumer Research (EuroSense)*, Verona, Italy, 2-5 September 2018.

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Industry day: Food Innovation for Delivering Nutrition, Health and Wellness, Leeds, UK, 18 January 2019.

E. M. Krop, M. M. Hetherington, A. Sarkar (2019). Effects of rheological and tribological properties of hydrogels compared to individuals' oro-physiological capabilities on oral processing behaviour. *The 20th Gums & Stabilisers for the Food Industry Conference*, San Sebastian, Spain, 11-14 June 2019.

Emma M. Krop, Marion M. Hetherington, Anwesha Sarkar (2019). The effect of hydrogel preload foods with different tribological properties on food intake. *International Symposium on Food Rheology and Structure (ISFRS 2019)*, Zurich, Switzerland, 17-20 July 2019.

E. M. Krop, M. M. Hetherington, A. Sarkar (2019). Oral processing: food texture matters more than consumers' eating capability in younger adults. *The 13th Pangborn Sensory Science Symposium*, Edinburgh, UK, 28 July - 1 August 2019.

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Abstract

Certain oral processing strategies, such as slow eating, high number of chews and hard food textures, have been linked to lowering food intake in a systematic review and meta-analysis. Although oral lubrication is an important aspect of oral processing, its effects on satiation remain unclear. Therefore, this thesis aimed to study the effects of both chewing and oral lubrication on snack intake by developing model foods (non-fat hydrogels) with different textural properties. The methodology used in this thesis ranged from instrumental techniques to human trials. Instrumental (texture analysis, rheology, tribology) and sensory evaluations (descriptive analysis, n=11) were used to characterise simple and mixed hydrogels with/without inhomogeneity using different concentrations and ratios of biopolymers. Viscosity and friction coefficients (μ) of the hydrogel-boli were characterized after simulated oral processing. Results demonstrated that gel fracture properties were directly correlated to the chewing-related sensory attributes, such as 'firm', 'elastic' and 'chewy' ($p < 0.05$). On the other hand, μ at orally relevant speeds (3-50 mm/s) was inversely correlated to 'pasty' of the gel bolus fluid where the large bolus fragments were filtered out. In addition, it was questioned whether the eating capabilities (EC) of young healthy consumers can be a determining factor in oral processing. Using quantitative frame-by-frame video analysis (n=28), it was demonstrated that number of chews and oral residence time were mainly dictated by food material properties rather than EC of young panellists. The effects of these novel hydrogels on subjective appetite and objective intake of a salty snack were measured in a preload, between-subjects design (n=55). Results showed that oral lubrication rather than chewing resulted in a reduced

snack intake after consuming a hydrogel preload ($p < 0.05$). In summary, manipulating oral lubrication is a promising new construct to reduce snack intake that merits future research in the oro-sensory satiety domain.

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List of abbreviations

ANOVA	Analysis Of Variance
BMI	Body Mass Index
BF	Bite Force
CaA	Calcium Alginate
CI	Confidence Interval
DA	sensory Descriptive Analysis
DE	Desire to Eat
DEBQ	Dutch Eating Behaviour Questionnaire
EC	Eating Capability
EHL	Elastohydrodynamic lubrication
EMG	Electromyography
FSTA	Food Science and Technology Abstracts
IOPI	Iowa Oral Performance Instrument
ιC	Iota-Carrageenan
κC	Kappa-Carrageenan
LBG	Locust Bean Gum
λC	Lambda-Carrageenan
MA	Meta-Analysis
ME model	Mixed Effects
M/F	Male/Female

MRI	Magnetic Resonance Imaging
MTM	Mini-Traction Machine
NA	Not Applicable/Available
NaA	Sodium Alginate
NW	Normal Weight, BMI of 18.5-24.9 kg/m^2
OB	Obese, BMI $\geq$ 30 kg/m^2
OTC	Optical Tribological Configuration
OW	Overweight, BMI of 25-29.9 kg/m^2
O/W	Oil-in-Water emulsion
PC	Principal Component
PCA	Principal Component Analysis
PDMS	Polydimethylsiloxane
PICOS	Population, Intervention, Comparison, Outcome, and Setting
PLS/PLSR	Partial Least Squares Regression
PRISMA	Preferred Reporting Items for Systematic Reviews and Meta-Analysis
PCTFE	Polychlorotrifluoroethylene
QDA	Quantitative Descriptive Analysis
RE model	Random Effects model
SD	Standard Deviation
SEM	Standard Error of Mean
SOP	Simulated Oral Processing
SRR	Systematic Research Review

SRR	Slide-to-Roll Ratio
TA	Texture Analysis
(T)CATA	(Temporal) Check-All-That-Apply
TDS	Temporal Dominance of Sensations
TFEQ	Three Factor Eating Questionnaire
TP	Tongue Pressure
UW	Underweight, BMI < 18.5 kg/m^2
VAS	Visual Analogue Scales
VMG	Vibromyography
WHO	World Health Organization

List of nomenclature

A	Area
D	Displacement
$d_{3,2}$	Particle size diameter
E	Young's modulus
E'	Reduced elastic modulus
F	Force
H	Height
n	Number
r	Radius
R	Correlation coefficient
R_a	Surface roughness
T	Temperature
t	Time
U	Entrainment speed
V	Velocity
W	Normal load
α	Surface contact
ε	Fracture strain
η	Viscosity
μ	Friction coefficient

ν	Poisson’s ratio
σ	Fracture stress
φ	Volume fraction

Chapter 1

General introduction

1.1. Introduction and overall research aim

Understanding the relationship between oral processing, texture perception and food satisfaction can help improve the design of food products tailored to the energy needs of specific consumers. The intake of food is essential human behaviour, providing the body with energy and nutrients to sustain life. The current trend of increased energy intake in excess of energy requirements has led to serious problems with overweight and obesity in the world population, particularly in urban settings (World Health Organization 2000). Overweight and obesity form major risk factors for a number of chronic diseases, such as diabetes, cardiovascular diseases and cancer, and impose a huge burden on health-care systems (World Health Organization 2016a). The food industry can play a significant role in promoting healthy diets by ensuring healthy and nutritious food options are available. Among many nutritional strategies proposed to reduce food intake, foods that inhibit appetite between meals (satiety) and increase the strength and duration of sensory, cognitive and post-ingestive signals that determine when a meal ends (satiation) have been on the radar of food industries (Chambers, McCrickerd and Yeomans 2015).

Previous research has indicated that certain oral processing strategies, such as more chews per bite and eating more slowly, result in lower food intake (Christen and Christen 1997). This would suggest that foods with textural properties that stimulate these oral processing strategies may be used to enhance satisfaction. Therefore, understanding the influence of the material as well as the sensorial texture properties of food on the oral processing behaviour is of great importance.

Oral processing is a dynamic process, with the food structure being broken down into a smaller particles and being mixed with saliva to form a cohesive food bolus that can be swallowed (Chen 2009; Chen and Stokes 2012). The food physical properties change drastically during this process, from first bite until swallowing, and consequently, oral processing plays a major role on the sensorial texture perception as well. Due to the continuously changing nature of oral processing, food properties transition from being rheology-dominated (*i.e.* bulk flow properties) to tribology-dominated (*i.e.* friction or lubrication due to interactions of food with the tongue and oral palate surfaces). In order to fully understand the textural changes during oral processing that eventually contribute to reduced food intake and their influence on texture perception, it is important to consider food properties both from a rheological and tribological point of view, with tribology experiments being a relatively new area in food research.

Thesis Aim: This PhD project aims to understand better the influence of oral processing on satiety and satiation, both from a rheological as well as a tribological perspective. The project involves a multidisciplinary approach due to the complexity of the parameters involved in oral processing, such as food science,

sensory science, psychology and mechanical engineering. Three main research areas were identified: (1) the material and sensory texture properties of model hydrogels, (2) their oral processing characteristics to study how such instrumental and sensory properties are realised during food consumption, and (3) the satiety/satiation responses resulting from foods with different chewing and oral lubrication properties (see **Figure 1.1**). By studying the instrumental characteristics of a wide range of single and mixed hydrocolloid model systems, soft solid hydrogels with different texture and oral lubrication properties can be designed. Instrumental analysis (rheology, aqueous tribology) can be used to describe the material properties and predict the sensory perception and eventually the oral processing behaviour of hydrogels. **Thesis hypothesis:** This thesis postulates that besides the chewing behaviour, the lubrication generated during oral processing by exogenously introduced hydrogels plays a role in reducing hunger and food intake.

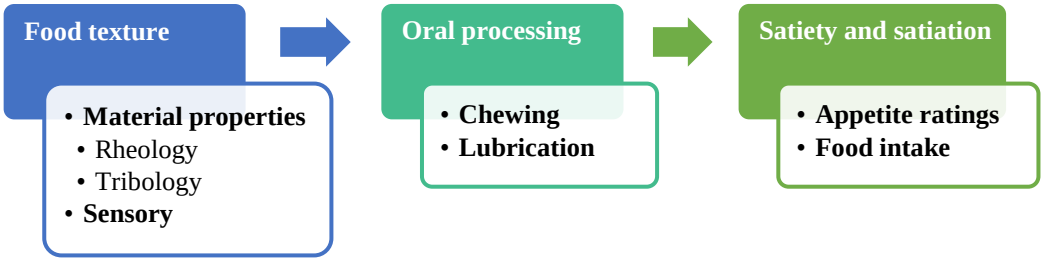

Figure 1.1. Schematic overview of the three main themes in this thesis.

In this chapter, the rationale behind the instrumental techniques and sensory analysis methodologies used in this thesis are explained, as discusses the rationale for using model foods. At the end, an outline of this PhD thesis is provided. This chapter is followed by a systematic review highlighting the gaps in literature in **Chapter 2** and the selection method for the hydrogel model systems in **Chapter 3**.

1.2. Rationale behind different techniques

For many years texture has been considered an overlooked food quality attribute, being the last of the key attributes (appearance, taste, aroma and texture) to gain wider research interest. However, the increased understanding of the importance of texture in food acceptance the last few decades has led to an increased use of both instrumental and sensory analysis methodologies, as well as the development of more universal equipment to measure texture and sensory methods that focus more on the dynamic aspects of texture perception (Bourne 2002b; Chen 2009). Due to the wide variety in food products and their different types of rheological and textural properties, there is a need for a wide variety of different methods to measure texture instrumentally. Although an adequate number of different methods exists nowadays, satisfactory measurements for certain product groups is as of yet unavailable or needs to be improved further (Bourne 2002a). The theoretical and technical aspects of the main instrumental and sensory characterisation tools used in this thesis are discussed below, as well as the techniques used to quantify oral processing and satiety/satiation.

1.2.1. Rheology

Rheology is the study of flow and deformation of materials under applied forces. Rheological properties of food can be measured either at small or large deformations.

1.2.1.1. Large deformation rheology

The large deformation measurements have found to be more relevant in oral processing studies, as food is subjected to large deformations during consumption;

the physical and chemical bonds between atoms and molecules in soft solids, such as gels, are weakened and eventually destroyed. Force measuring instruments, such as a Texture Analyser, are the most common for texture evaluation and several different tests can be used to apply large deformations. Examples of these force tests are puncture tests, (uniaxial) compression tests, bending and snapping tests (*e.g.* 3-point), tension or torsion tests, or compression-extrusion tests for bulk measurements (Bourne 2002a).

The plate-to-plate uniaxial compression is a commonly used method to test large deformation properties in soft solids. A constant force is applied to the test sample along a straight axis, and the sample is allowed to deform freely in the other directions. The applied force over time is measured, and a fracture stress (σ) and fracture strain (ε) can be used to describe the deformation behaviour. The stress is defined as the force applied per unit area of food responsible for the deformation, and the strain represents the deformation per unit length of the food as caused by the applied force (**Figure 1.2**).

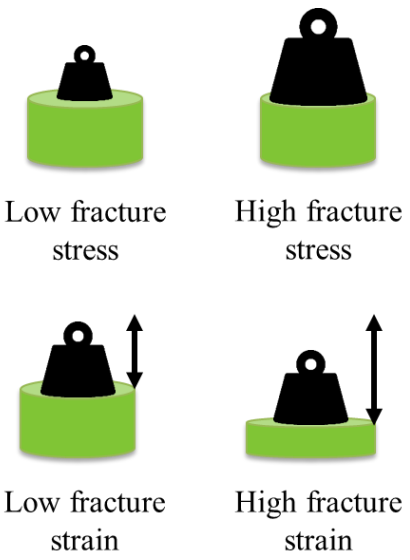

Figure 1.2. Schematic representation of fracture stress and fracture strain.

Thus, using these definitions, the true stress and true strain can be obtained by Equations 1.1 and 1.2:

$$True\ stress\ (Pa) = \frac{F_t(N)}{A_t\ (m^2)} \tag{1.1}$$

$$True\ strain\ (dimensionless) = \ln\left[\frac{H_t(m)}{H_0(mm)/1000}\right] \tag{1.2}$$

where F_t is the applied force at a given time and A_t the cross-sectional area of the sample at a given time. This area can be determined using Eq. 1.3:

$$A_t\ (m^2) = \frac{A_0(mm^2)/1000000 \times H_0(mm)/1000}{H_t\ (m)} \tag{1.3}$$

with A_0 the original cross-sectional area of the sample, H_0 the original height of the test sample and H_t the height of the sample at a given time, obtained from Eqs. 1.4 and 1.5:

$$A_0 = \pi r^2 \tag{1.4}$$

$$H_t\ (m) = \frac{H_0\ (mm) - D_t(mm)}{1000} \tag{1.5}$$

with r the radius of the sample and D_t the displacement of the sample at a given time. Thus, the stress-strain curve can be drawn from the force-time curve

(see **Figure 1.3**). The fracture properties can be determined at the maximum point of the stress-strain curve. The fracture energy is determined as the area under the curve up to the fracture point, and the Young's modulus is the initial slope of the force displacement (Peleg 1984). These fracture properties under large deformation have been linked to the sensory perception of food textures, with fracture stress often being related to hardness and fracture strain with brittleness (Foegeding *et al.* 2011).

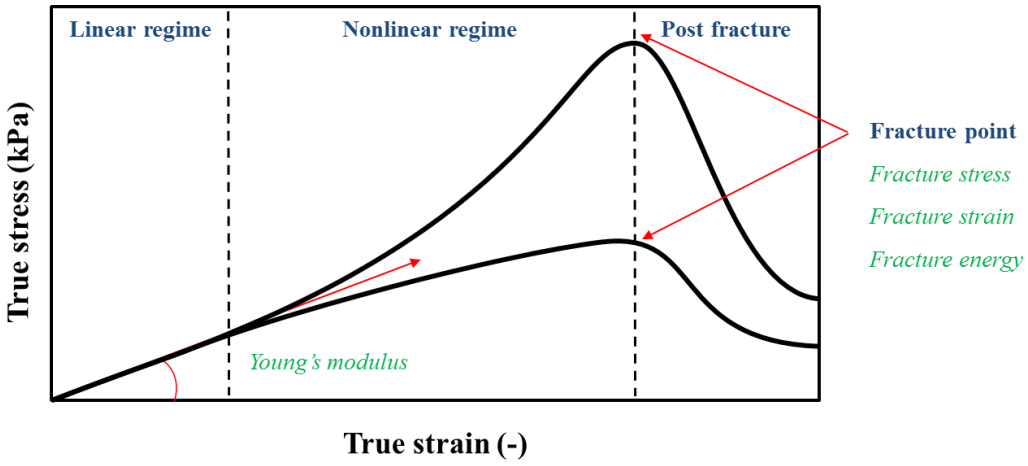

Figure 1.3. Representation of typical stress strain curve of solid materials under large deformation, explaining the fracture behaviour (Koç *et al.* 2013).

In literature it has been described that lubrication, or lack of lubrication, of the contact surface between the food and the mouth affects the force required to reach a certain degree of compression, with a non-lubricated surface requiring a higher force than a lubricated surface for the same degree of compression (Culioli and Sherman 1976). Also, a cylindrically shaped food sample compresses in different ways depending on whether lubricant is present or not. However, during food compression in the mouth, the cusps of the molars act as anchors to hold the food in place and prevent lateral movement. Comparison tests have shown that the

anchoring effect of molars prevailed over the lubricating effects provided by saliva. Therefore, the surface should be lubricated in compression tests trying to replicate true rheological data, but in tests trying to see what happens in the mouth the surface should not be lubricated (Brennan and Bourne 1994).

In this thesis, compression tests to characterise the model foods were used (**Chapter 4**), as well as puncture tests to mimic first bite oral processing conditions (**Chapters 3 and 5**). As we were mostly interested in initial food properties, no saliva was added during testing.

1.2.1.1. Flow behaviour and apparent viscosity

The flow behaviour of liquid foods is commonly characterised by viscosity measurements, where the sample is subjected to a shearing deformation. Depending on the flowing behaviour, liquids can be classified as Newtonian or non-Newtonian. For ideal Newtonian fluids, the viscosity can be determined by the slope of shear stress by shear rate, and will be independent of shear rate and shearing time at a specific temperature (van Vliet 2014). Typically, samples with higher molecular weight will not have a linear relationship between shear stress and shear rate, and are classified as non-Newtonian liquids. Non-Newtonian liquids can display shear thinning or shear thickening behaviour, as shown in **Figure 1.4**. Products thickened by polysaccharides are typically shear thinning. Under applied shear, the polymers will disentangle in the direction of the flow, leading to a viscosity decrease with increasing shear rate. Shear thickening behaviour is much less common, and mainly occurs in concentrated dispersions. With increasing shear rates, the molecules will form new clusters thus increasing the viscosity. Viscosity dependent on the shear rate is termed apparent viscosity.

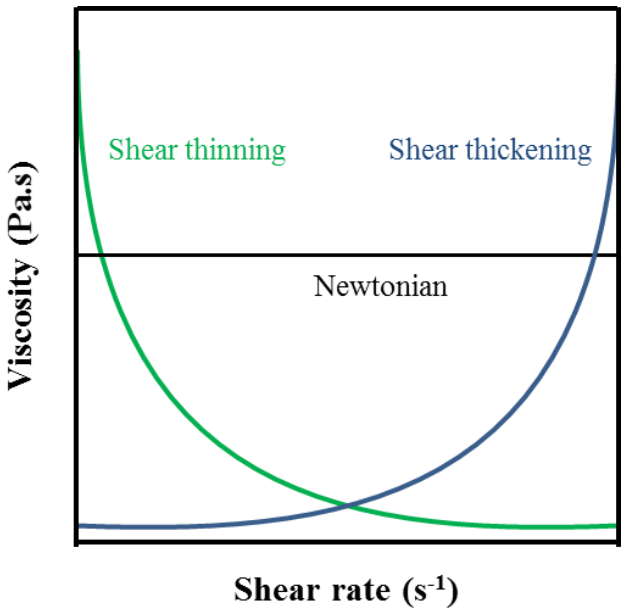

Figure 1.4. Typical flow-curves for Newtonian and non-Newtonian fluids (van Vliet 2014).

In this thesis, the apparent viscosity of simulated hydrogel bolus samples was studied. Using a stress-controlled rheometer, the so-called flow curves or viscosity curves can be obtained (**Chapter 4**). Rheological measurements can be conducted using three different type of geometries: plate on plate (two parallel plates), cone on plate or a concentric cylinder (*i.e.* cup and bob) geometry. Due to the heterogeneous nature of bolus samples with gel particles, a plate-plate geometry was used where the gap could be widened in order to accommodate the gel particles of various sizes in the different samples.

1.2.2. Tribology

Most studies on oral processing focus mainly on the mechanical response (via compression tests) and bulk rheology (via viscosity measurements) of the food (Prakash, Tan and Chen 2013). However, as demonstrated in **Figure 1.5**, food undergoes continuous structural changes during oral processing, such as reduction

of food particle size, release of nutrients, addition of saliva to increase the moisture content for the formation of a cohesive bolus and formation of an oral coating in the mouth after swallowing of the bolus (Chen 2009; Foegeding *et al.* 2011; Stokes, Boehm and Baier 2013).

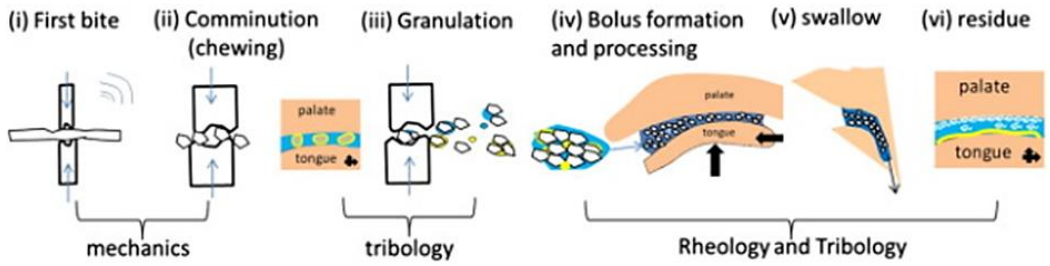

Figure 1.5. Oral processing, going from rheology-dominant to tribology-dominant, adapted from Stokes, Boehm and Baier (2013).

The rheological properties of the food structure before consumption are not sufficient to describe some of the observed differences in flow behaviour and the surface-related sensory texture and mouthfeel attributes (Chen and Stokes 2012; Selway and Stokes 2013). Instead these attributes can be explored further by studying the frictional responses of the food-saliva mixture between the oral-contact surfaces (Selway and Stokes 2013; Stokes 2012a). Tribology, although well-established in mechanical engineering and material science as a technique for measuring these frictional properties, is a relatively new technique in food and related soft matter sciences. The main goal of applying tribological measurements in food science literature has been to replicate oral processing conditions, with the two interacting surfaces representing the tongue and upper-palate. Materials for these interacting surfaces vary from rubber, nylon, and modified PDMS (polydimethylsiloxane) to animal tissue such as a pig's tongue (Selway and Stokes 2013; Stokes 2012a; van Vliet *et al.* 2009).

Figure 1.6 shows a schematic representation of a mini-traction machine (MTM), which is a general purpose instrument used in this thesis (**Chapter 4**) for measuring the frictional properties of lubricated and non-lubricated contacts under a wide range of rolling and sliding conditions. The ball is loaded against the face of the disc, and the ball and disc are turned independently to create a mixed rolling/sliding contact. The frictional force between the ball and disc, the turning speed of the ball and the disc, the applied load and the lubricant and the pot temperature are recorded by the tribometer. To mimic body temperature, tests have been performed at 37 °C.

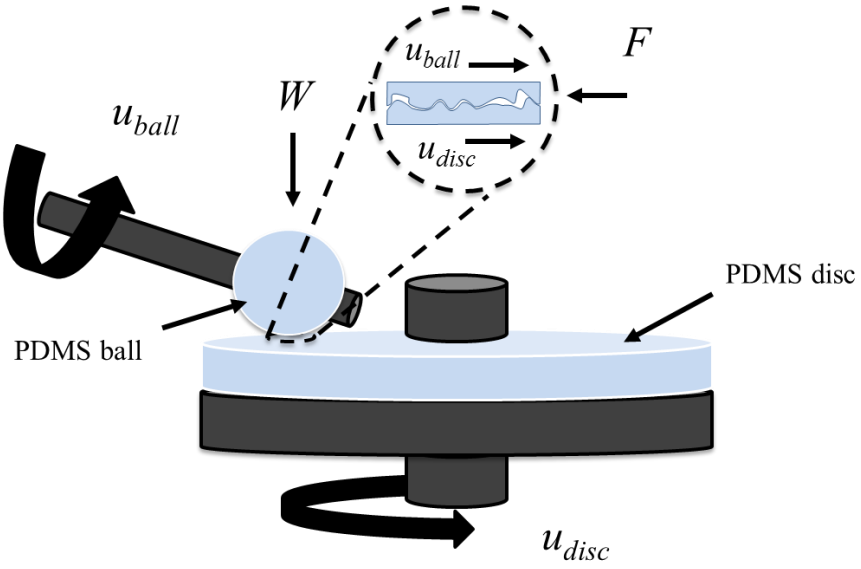

Figure 1.6. Schematic representation of a tribometer set-up, with W the applied contact load, u_{ball} and u_{disc} the ball and disc speed, respectively, and F the friction force.

As an alternative to the hard specimens made from steel commonly used in mechanical engineering, soft contact ball and disc surfaces can be used. Among the soft contact materials, polydimethylsiloxane (PDMS) elastomers are well-characterised and more suitable for measuring the tribological behaviour in

conditions representative of in-mouth oral processing than hard specimens (Selway and Stokes 2013; Sarkar *et al.* 2019). PDMS tribopairs (see **Figure 1.7**) are prepared by mixing two base elastomers and allowing them to form cross-links, resulting in certain physical and chemical properties. The oral surfaces are intrinsically hydrophobic, however, due to the continuous presence of saliva in the mouth, become rather hydrophilic (Sarkar *et al.* 2019). The PDMS surfaces on the other hand are also hydrophobic, and therefore were coated in artificial saliva before the measurement in this thesis (**Chapter 4**) in an attempt to make them more hydrophilic and thus better mimic the conditions in the mouth.

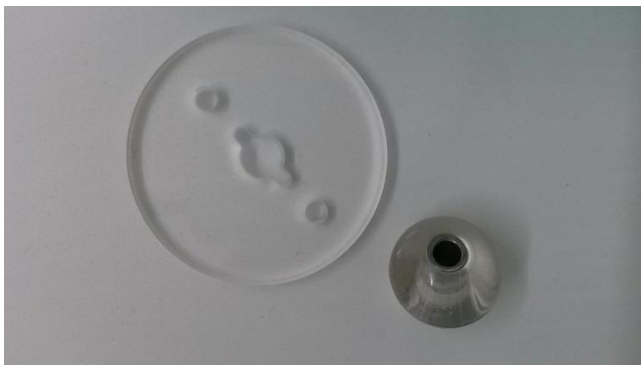

Figure 1.7. Commercial PDMS ball and disc tribology specimens.

Using these contact surfaces, the friction behaviour can be presented via the construction of a Stribeck curve. In this curve, the coefficient of friction is plotted against a controlling parameter, such as the entrainment speed. Often a measure of the film thickness or the sample viscosity is included (Stokes 2012a). A typical Stribeck curve, as shown in **Figure 1.8**, can be divided into three distinctive regimes: the boundary, mixed and hydrodynamic lubrication regimes (Chen and Stokes 2012; Joyner, Pernell and Daubert 2014). In the boundary regime, a constant, relatively high friction can be observed at low sliding speeds. This means that the sample thickness is lower compared to the roughness of the surface, and

the ball and disc are in direct asperity contact. With increasing sliding speeds, the friction will eventually start to decrease, shifting to the mixed regime. The sample thickness will start to increase with increasing sliding speeds, starting to separate the contact surfaces from each other and thus lowering the coefficient of friction. After the surfaces are completely separated, the minimum friction is reached and the curve will move to the hydrodynamic regime (Stokes 2012a; Joyner, Pernell and Daubert 2014). This typical Stribeck curve is mostly seen for Newtonian liquids and with contact surfaces made from steel, so with non-Newtonian samples and soft contact specimens the shape of the curve might differ (Joyner, Pernell and Daubert 2014).

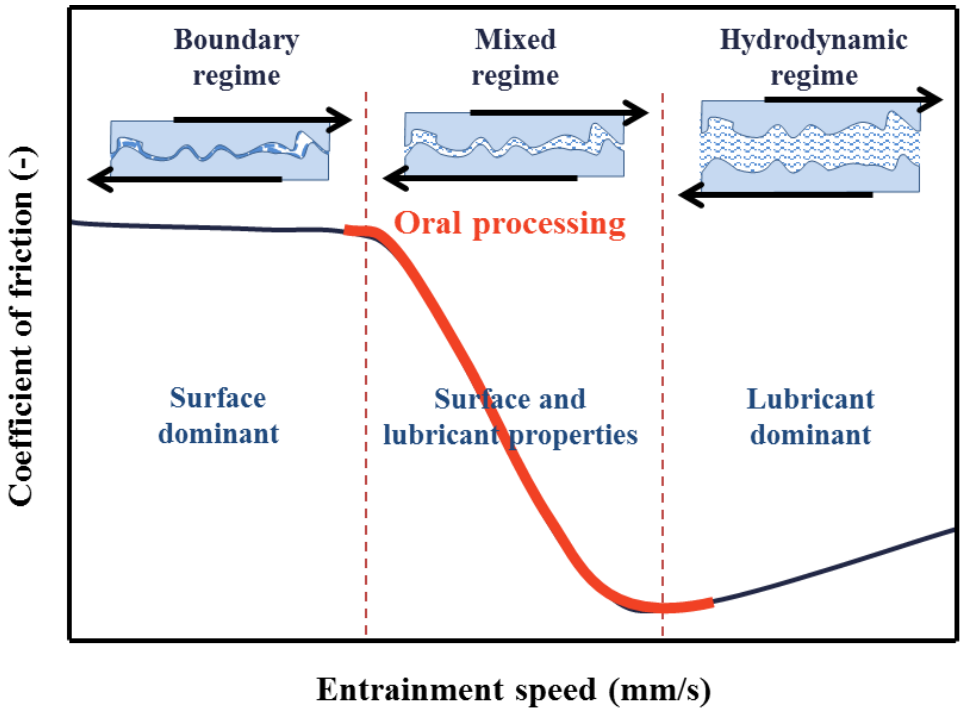

Figure 1.8. Typical Stribeck curve, adapted from (Bongaerts, Fourtouni and Stokes 2007; Stokes 2012b; Liu 2016).

Using the high shear rheology viscosity, a Stribeck master curve can be modelled for Newtonian liquids (de Vicente, Stokes and Spikes 2005; Bongaerts, Fourtouni and Stokes 2007). By multiplying the entrainment speed with the viscosity of the lubricant and plotting that against the friction coefficient, a baseline curve can be generated against which the Stribeck curves of other samples can be compared. In the mixed and hydrodynamic lubrication regimes, the friction coefficient mainly depends on the bulk viscosity properties. However, in the boundary regime the friction coefficient also depends on the adhesion properties of the lubricant. Therefore, the master curve will fit less well in the boundary regime. Additionally, for non-Newtonian fluids the master curve will look slightly different. By using the product of entrainment speed and the dynamic viscosity, the mixed lubrication effects of non-Newtonian lubricants can be compared (de Vicente, Stokes and Spikes 2006).

1.2.3. Sensory perception

Texture plays a major role in consumer liking and acceptance of food (Szczesniak 2002). In addition to appearance, taste and aroma, texture is a key sensory attribute in food perception. Texture perception, or *haptaesthesis*, is a dynamic process and involves a complex interplay between the structural properties of the food, the oral processing characteristics and integration of the stimuli as perceived by the senses leading to a conscious perception of the sensory attributes (Wilkinson, Dijksterhuis and Minekus 2000). Unlike for taste and aroma, no specific receptors exist for texture perception. Despite the developments in instrumental texture analysis methodologies, instruments can only measure certain physical properties of the food which must then be interpreted in terms of sensory perception (Szczesniak

2002). Human assessment remains the most accurate method to determine the textural properties of a food. Since texture consists of a number of different physical sensations, elicited by the food structure, it is considered a multi-parameter attribute and better referred to as a group of properties (Bourne 2002b; Szczesniak 2002). Texture properties are mainly perceived through the senses of touch and kinesthesis (the perception of muscle movement), during food preparation and handling as well as during oral processing in the oral cavity. In addition, textural properties may be assessed by the senses of vision and hearing, however, this thesis will focus on the attributes perceived in the mouth by the senses of touch and kinesthesis.

The texture profile method (Brandt, Skinner and Coleman 1963) is one of the few sensory methods focussed solely on texture assessment of food. However, other techniques such as Quantitative Descriptive Analysis (QDA®) (Stone *et al.* 1974) and Spectrum™ descriptive analysis (Muñoz and Civille 1998) can be modified to focus solely on the texture attributes. Although these methods acknowledge the dynamic nature of texture perception, assessors are required to integrate the attribute ratings over time to provide single intensity values (Cliff and Heymann 1993). Therefore, the evolution of texture attributes during oral processing, from first bite to swallowing, is not included in these measurements.

Oral texture properties are commonly divided into three sub-groups: 1) those related to the food mechanical properties, 2) the geometrical properties, and 3) attributes related to the moisture and fat content (Szczesniak 1963). Mechanical attributes, such as hardness, fracturability (brittleness) and chewiness, can be easily linked to the chewing aspects of oral processing, whereas geometrical and

moisture/fat-related attributes are more associated with the friction/lubrication in the mouth. Mouthfeel attributes, such as creamy, smooth, watery, sticky and astringency, seem to be connected to oral lubrication.

In this thesis, descriptive sensory analysis was used to provide a complete quantitative picture of the texture attributes in the designed model foods (**Chapter 4**). Sensory attributes were specifically selected with an eye on their connection to oral lubrication. Attributes were rated on a 100 mm unstructured line scale, labelled from "not at all" to "very" at opposing ends. Descriptive analysis allows the investigation of the impact of changing structural elements on specific sensory attributes, as well as a way to study the relationship between different sensory attributes and instrumentally measured properties.

1.2.4. Oral processing

Food oral processing is a complex dynamic process in which the food is manipulated in the mouth from first bite until swallowing to form a food bolus that is suitable for further digestion and the uptake of energy and nutrients to sustain life. As a result of this dynamic process, texture perception will depend on the continuously changing physical properties of the food product (Bourne 1975; Chen 2009). This led Hutchings and Lillford (1988) to come up with a new approach in which oral processing is considered a combination of reduction of particle size (degree of structure) and an increase of moisture content (degree of lubrication) over time. They outlined the breakdown path of different types of food using this approach, where the degree of structure and degree of lubrication are manipulated over time until the swallowing threshold is reached (see **Figure 1.9**). The initial structure of the food determines the oral processes needed before swallowing, *e.g.*

liquid foods will require little processing and will be transported through the mouth relatively quickly, whereas semi-solids may be processed by tongue compressions and solids require chewing to break the food down into smaller particles before swallowing can occur (Pascua, Koç and Foegeding 2013). The moisture content may be increased due to water released from the food matrix upon breakdown or by the addition of saliva in the mouth.

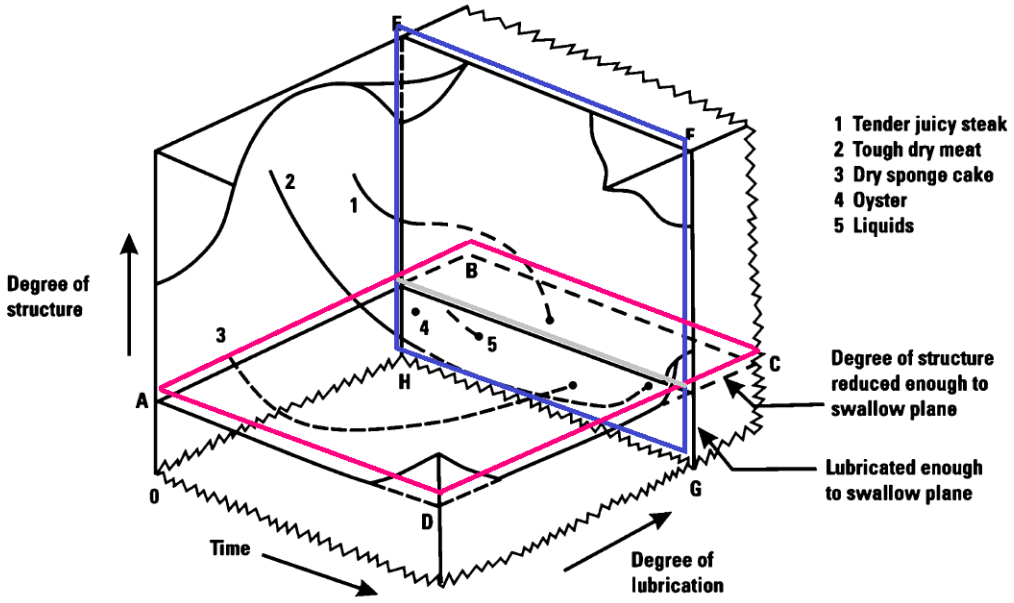

Figure 1.9. Schematic model of the in-mouth breakdown path during oral processing, adapted from Hutchings and Lillford (1988).

Oral processing may be divided in several stages, including grip, first bite, fracture, particle size reduction, transportation and swallowing (Lucas *et al.* 2002), and is regulated by sensory feedback to the central nervous system as the physical properties of the food are modified (van der Bilt *et al.* 1995; de Wijk *et al.* 2008). There are various different methods with which the oral processing behaviour has been investigated, such as electromyography (EMG) or vibromyography (VMG)

to measure muscle activity, real-time magnetic resonance imaging (MRI), jaw-tracking, sirognathography (magnetic tracking of jaw movements), videofluography, or by less invasive techniques such as self-tracking, direct observation or video recording (Chen 2009; Pascua, Koç and Foegeding 2013; de Wijk *et al.* 2008; Çakir *et al.* 2012; Gonzalez Espinosa and Chen 2012). Video recording is considered a relatively easy, yet accurate technique and allows for the analysis of a large number of participants in relatively short time (Hennequin *et al.* 2005; Wilson *et al.* 2016), and therefore was used in this thesis to analyse the oral processing behaviour for different model foods (**Chapters 5 and 6**). Using specialised behavioural observation software, videos can be coded for specific parameters to facilitate accurate analysis, see **Figure 1.10** (Forde *et al.* 2013).

Figure 1.10. Oral processing video analysis, example of the coding scheme with participant 28 drinking a liquid tea sample and participant 36 eating a soft solid hydrogel sample.

1.2.4.1. Saliva

An important aspect of oral processing of solid and semi-solid foods is the incorporation of saliva to form a swallowable food bolus. Saliva is a complex biological fluid that consists of mainly water (~99.5%), various enzymes (α-amylase, lysozyme, lingual lipase, etc.) and proteins, (~0.3%), small organic compounds and inorganic salts (Sarkar, Goh and Singh 2009). The key protein component in human saliva is highly glycosylated mucin, which mainly contributes to the lubrication and shear-thinning properties of saliva (Schipper, Silletti and Vingerhoeds 2007). Mucins cover the oral mucosa and either form an immobile salivary pellicle on the epithelial cells or a mobile salivary film (Laguna and Sarkar 2017; Xu, Laguna and Sarkar 2019). Different types of mucins have been identified in humans, and are specific to its mobility function in the mucosal salivary pellicle (Morzel *et al.* 2014).

The incorporation of saliva in the food bolus over time has a major effect on the sensory texture perception (Funami *et al.* 2012; Hutchings and Lillford 1988). Therefore, the mechanical and friction properties might change significantly due to the interactions between food and salivary components, such as mucins and salts. Saliva is a biological fluid that might contain pathogens. For this reason, any work on bolus samples containing real saliva could pose a risk to the person handling the samples. Due to chemical and enzymatic reactions, the properties of saliva and a food bolus containing saliva will change over time. Therefore, analysis of the samples should happen within a very short time frame. To minimise any risk, instead of using real human saliva, artificial saliva was prepared and used for the instrumental measurements in this thesis (**Chapters 4, 5 and 6**). For ethical

reasons, porcine gastric mucin was used as a close representative of human salivary mucins (Sarkar, Goh and Singh 2009).

1.2.4.2. Eating capabilities

Besides differences in oral processing due to the physical properties of food, oral processing may vary due to oro-physiological differences between individuals. Factors, such as age, gender, volume of the oral cavity, muscle strength or even the time of day the food is being consumed, can have an influence on the oral processing behaviour (Chen 2009; Pereira and van der Bilt 2016; Lassauzay *et al.* 2000). Though mainly relevant in studies looking at individuals with reduced oral capabilities, such as the elderly or patients with dysphagia (a swallowing disorder), differences in oral processing for healthy participants have been noted in many studies (Chen 2009; Woda *et al.* 2006). A number of techniques can be to measure the oro-physiological properties involved in oral processing, such as bite and tongue capabilities.

There are several oro-facial muscles that are relevant in oral processing, such as the left and right masseter and temporalis muscles and the medial pterygoid (Chen 2009; Foegeding *et al.* 2011). These muscles perform a collaborative function to make the open and closed jaw movements necessary for oral processing. The thickness of the muscles has an effect of bite force and may differ depending on the individual, gender and age (Pereira 2012). A simple, cost-effective device to measure bite force was designed by Flanagan *et al.* (2012). A very thin flexible force sensor sandwiched between two adhesive silicone discs can be used to measure changes in electronic resistance under load (Flanagan *et al.* 2012). By placing the sensor between the teeth and biting down at full capacity, the maximum

bite force can be recorded. As shown in **Figure 1.11**, the sensors are connected to a multi-meter to detect the changes in resistance, and can then be converted to force (N) using calibration data (Flanagan *et al.* 2012; Laguna *et al.* 2015).

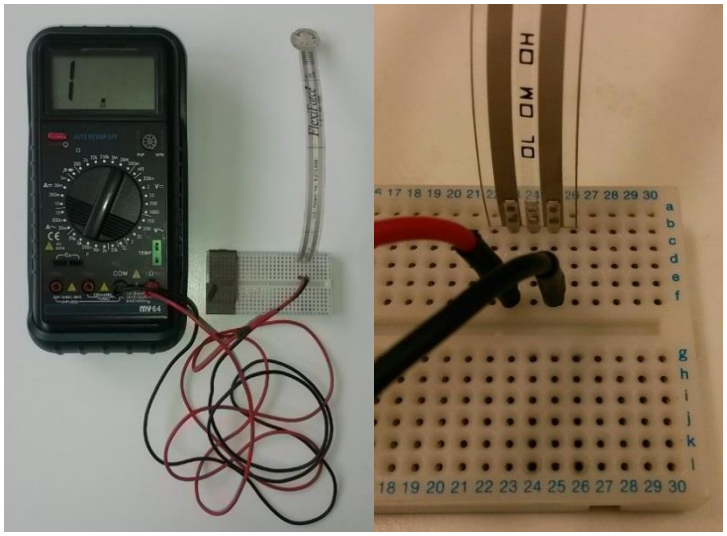

Figure 1.11. Multi-meter and flexisensor with silicon disc for bite force measurements, connected through a bread board.

The tongue plays a major role in oral processing due to its ability to deform and its flexibility to move in all directions in a highly coordinated manner. It consists of a large bundle of muscles and forms a major part of the oral cavity (Chen 2009; Pereira 2012). The effectiveness of the tongue may be determined by the strength of the muscle to facilitate bolus movement through the mouth (Laguna, Sarkar and Chen 2015; Alsanei and Chen 2014). The Iowa Oral Performance Instrument (IOPI) is a medical device used for measuring the maximum tongue pressure that can be expressed (**Figure 1.12**). By compressing a disposable tongue bulb linked to a pressure transducer between the tongue and the upper hard palate, the tongue pressure can be recorded.

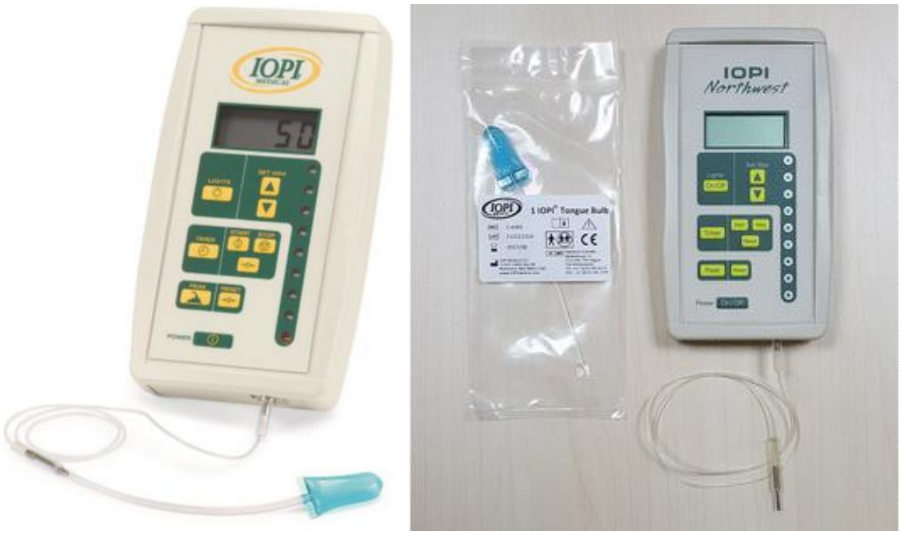

Figure 1.12. The Iowa Oral Performance Instrument (IOPI) and tongue bulb used for tongue pressure measurements.

The tongue pressure and bite force may give an indication of the forces used during oral processing. In this thesis, the eating capabilities of healthy participants were quantified by measuring the maximum tongue pressure and maximum bite force, and are described in **Chapter 5**.

1.2.5. Satiety studies

The concept of human appetite has become an increasingly important research topic in the last couple of decades. With the world's increasing overweight and obesity related health problems, the need for studies on the expression of appetite and energy intake regulation is becoming more urgent (World Health Organization 2016a). An adult with a healthy body weight is characterised by a normal body mass index (BMI) between 18.5 and 24.9 kg/m^2. BMI is a simple index of weight divided by a person's square height. Below 18.5 kg/m^2 a person is considered underweight, with a BMI of 25 kg/m^2 or above one is considered overweight, and 30 kg/m^2 or higher is the threshold for adult obesity (World Health Organization

2016b). Overweight and obesity tend to be the result of food intake in excess of energy requirements (World Health Organization 2016b; Blundell *et al.* 2009). Healthy regulation of appetite and food intake involve both the decision to select a variety of food products and their consumption in appropriate quantities (Blundell *et al.* 2009).

The "Satiety Cascade" is a multifactorial framework, shown in **Figure 1.13**, used to capture the various processes, attributes of food, and related psychological and behavioural states involved in the regulation of satiation and satiety (Blundell, Rogers and Hill 1987; Blundell *et al.* 2010; Mela 2006). Satiation is defined as the short-term processes leading to the termination of an eating event, and encompasses all the sensations and psychological influences perceived during the meal itself (Blundell *et al.* 2009). As satiation is said to control the meal size, it is usually represented by the objective measure of *ad libitum* food intake, *i.e.* the amount of food selected freely in that moment, measured in weight or energy content (Blundell *et al.* 2010). Additionally, the subjective appetite sensations, such as hunger, fullness, desire to eat and thirst, can be monitored using visual analogue scales (VAS). This is a reproducible method for detecting small effects when there is no impact on food intake (Stratton *et al.* 1998; Stubbs *et al.* 2000). VAS typically consists of an unmarked 100 mm horizontal line that is anchored at either end with the two extreme states (*e.g.* minimum and maximum hunger), although alternative versions, such as 5 to 10-point Likert scales, 150 mm line scales or bipolar scales, exist (Merrill *et al.* 2002; Blundell *et al.* 2009). Satiation may be influenced by a number of dietary factors, such as portion size, energy density of the presented food, and the sensory properties and perceived palatability (Blundell *et al.* 2009),

as well as non-dietary factors, such as stress, mindfulness during eating, memory, social facilitation, food availability and price *etc.*.

Satiety, on the other hand, refers to the suppression of appetite and inhibition of further eating after a meal has ended due to the cognitive and physiological effects involved in the digestion of food (Blundell *et al.* 2009). Satiety may be measured objectively by the time until the next eating occasion, but meal times are highly conditioned in most people and are determined to a high degree by convenience (de Castro 1987; de Castro and Elmore 1988). However, more commonly satiety is characterised by subjective feelings of appetite at pre-specified time intervals after a meal. Thus, satiety is more influenced by the actual amount of energy consumed and the macronutrient content, rather than the weight of the food. Due to experience over time, people learn to estimate the satiating effects of many different foods. This learned expected satiety, for example for certain textures, may determine food intake at subsequent meals regardless of the actual energy density of the food (Blundell *et al.* 2010).

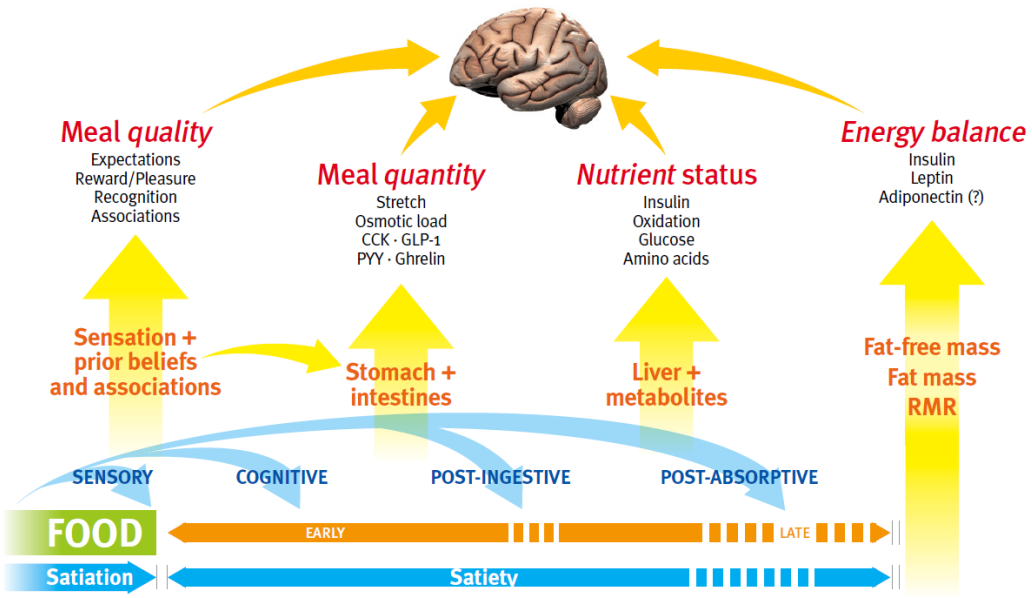

Figure 1.13. Satiety cascade, adapted from Blundell *et al.* (2010).

Due to the multifaceted nature of appetite regulation, a study on the short-term satiation and satiety effects needs to be designed under carefully controlled cognitive, sensory and environmental circumstances where hunger levels between participants are standardised prior to the start of the study (Blundell *et al.* 2009; Robinson *et al.* 2018; Hetherington and Rolls 2018; Meule 2018).

1.3. Rationale for using biopolymer model gels

Food products generally consist of rather complex structures, whether occurring naturally or manufactured by industry. While taste and aroma (flavour) can be modified, it is more difficult to change food texture. Therefore, model foods with simplified structures may offer a means to manipulate specific aspects of food texture (Funami 2011). Biopolymer gels from polysaccharides or proteins have been characterised extensively in the literature, *e.g.* the effects of composition (such as type of polymer, concentrations, combinations, addition of salts *etc.*) and processing (temperature, applied shear, pH) on the structural and rheological properties, as well as the sensory texture properties (Foegeding 2007; Funami *et al.* 2012; Stieger and van de Velde 2013). The hydrogel selection process for this thesis is discussed further in **Chapter 3**.

1.4. Thesis outline

This thesis starts with a systematic review of the effects of oral processing related to satiety and satiation, and continues with experimental studies on different hydrogels for selection and characterisation using instrumental and sensory techniques, the measurement of the oral processing characteristics of these hydrogels and eating capabilities of participants, up to the investigation of the

satiating properties of these hydrogels varying in chewing and oral lubrication properties. The outline of this thesis is highlighted in **Figure 1.14.**

Chapter 2 provides the context for this thesis by presenting a systematic research review and meta-analysis of the literature on oral processing, including chewing and oral lubrication, related to appetite ratings and food intake. The aim of this chapter is to set the scene for subsequent research by identifying gaps in current knowledge. The content of this chapter is published in the peer-reviewed journal '*Appetite*'.

Chapter 3 discusses a preliminary study of the fracture properties of a wide range of single hydrogels, mixed hydrogels and hydrogels with texture complexity. Based on these fracture properties, different hydrogels were then selected for further study.

In **Chapter 4** the selected hydrogels are characterised in detail using different instrumental and sensory methods. The fracture properties will be used to link to chewing-related sensory attributes, whereas the apparent viscosity and friction properties of simulated bolus hydrogels might relate to lubrication-related sensory attributes. For the first time, tribology measurements of non-fat containing food products are explored and their relevance to sensory perception is determined. The results of this chapter are published in the peer-reviewed journal '*Food Hydrocolloids*'.

The aim of **Chapter 5** is to disaggregate the effects of the food material properties and the eating capabilities of the individual eating the food on their oral processing behaviour. The correlation data of this chapter have been submitted for publication in the peer-reviewed journal '*Journal of Texture Studies*'.

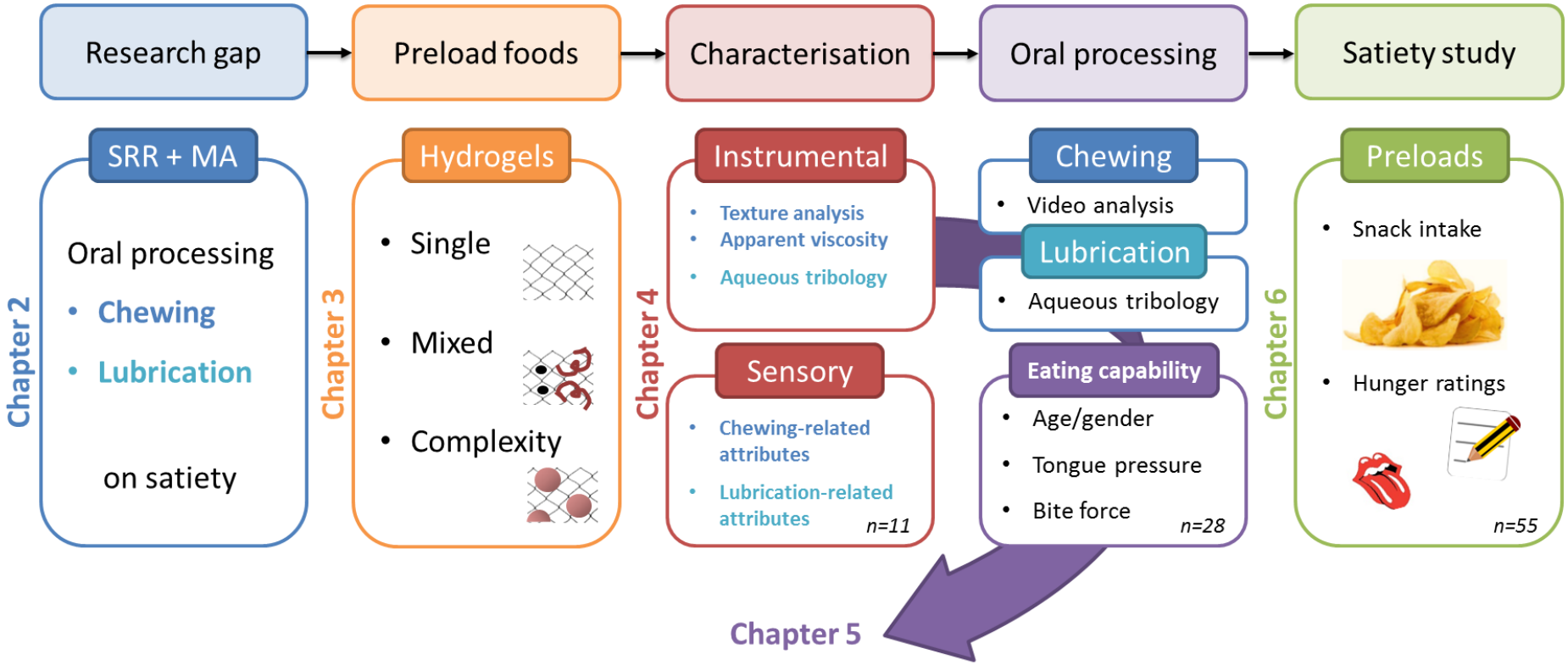

Figure 1.14. Schematic overview of the experimental approach employed in this thesis and the associated research chapters: linking the instrumental and sensory properties of hydrogels to satiety through chewing and oral lubrication.

Chapter 6 shows the effects of hydrogel preload foods differing in chewing and lubrication properties on the intake of a salty snack. For the first time, instrumental and sensory parameters for lubrication will be linked to measures of satiation and satiety. The effects of the hydrogel preloads on snack intake have been published in the peer-reviewed journal '*Food Quality and Preference*'.

The final chapter, **Chapter 7**, includes a general summary and discussion of the main findings of this PhD project. In addition, the implications of the results along with recommendations for future research are included.

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Chapter 2

Influence of oral processing on appetite and food intake - A systematic review and meta-analysis[a]

Abstract

Food delivers energy, nutrients and a pleasurable experience. Slow eating and prolonged oro-sensory exposure to food during consumption can enhance the processes that promote satiation. This systematic review and meta-analysis investigated the effects of oral processing on subjective measures of appetite (hunger, desire to eat) and objectively measured food intake. The aim was to investigate the influence of oral processing characteristics, specifically "chewing" and "lubrication", on "appetite" and "food intake". A literature search of six databases (Cochrane library, PubMed, Medline, Food Science and Technology Abstracts, Web of Science, Scopus), yielded 12161 articles which were reduced to a set of 40 articles using pre-specified inclusion and exclusion criteria. A further two articles were excluded from the meta-analysis due to missing relevant data. From the remaining 38 papers, detailing 40 unique studies with 70 subgroups, raw data were extracted for meta-analysis (food intake n = 65, hunger n = 22 and desire

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to eat ratings n = 15) and analysed using random effects modelling. Oral processing parameters, such as number of chews, eating rate and texture manipulation, appeared to influence food intake markedly but appetite ratings to a lesser extent. Meta-analysis confirmed a significant effect of the direct and indirect aspects of oral processing that were related to chewing on both self-reported hunger (-0.20 effect size, 95% confidence interval CI: -0.30, -0.11), and food intake (-0.28 effect size, 95% CI: -0.36, -0.19). Although lubrication is an important aspect of oral processing, few studies on its effects on appetite have been conducted. Future experiments using standardized approaches should provide a clearer understanding of the role of oral processing, including both chewing and lubrication, in promoting satiety.

2.1. Introduction

Food intake is a motivated behaviour essential to survival by providing energy and nutrients to the body. However, chronic energy intake in excess of requirements leads to a positive energy balance, and in the long term, contributes to obesity (World Health Organization 2000). For the first time in human history, the proportion of the population that is obese (body mass index, BMI $\geq$ 30 kg/m^2) and overweight (BMI of 25 to < 30 kg/m^2) has surpassed the proportion of adults who are underweight (BMI < 18.5 kg/m^2). The WHO (2016) estimates that about 1.9 billion adults globally have overweight, of whom > 30% have obesity (World Health Organization 2016). Consumers are encouraged to eat less and move more (Hill 2006) and food manufacturers have been working to reformulate foods to reduce their energy content whilst maintaining or improving satisfaction for

example, by increasing oral processing to enhance satiation and satiety (Hetherington *et al.* 2013).

While the terms “satiation” and “satiety” are often used synonymously in the literature, they encompass different components of the satiety cascade. Satiation is defined as the processes leading to meal termination, and therefore includes all events taking place during the course of the eating occurrence and controls meal size (Blundell *et al.* 2009). On the other hand, satiety is described as the inhibition of further eating as well as the suppression of feelings of hunger (Blundell *et al.* 2009; Blundell *et al.* 2010). Satiety has an influence on the time between two meals during which hunger, which has been suppressed, then begins to increase until the next eating occurrence. Constructs such as hunger and desire to eat represent approach behaviours indicative of appetite or readiness to eat (Stubbs *et al.* 2000). During sham feeding studies in humans, chewing fails to reduce hunger and desire to eat (subjective appetite) but produces sensory specific satiety and decreases food intake (Nolan and Hetherington 2009). Therefore, in examining the effects of oral processing it is important to attend to behavioural markers of both appetite and satiation.

During food consumption, food is processed in the mouth from first bite to swallowing, primarily involving reduction in the particle size driven by “chewing”, and the incorporation of saliva to form a swallowable bolus through “oral lubrication” (Chen 2009; Chen and Stokes 2012; Sarkar and Singh 2012; Sarkar, Ye and Singh 2017). Depending on the nature of food and its oral interactions, the length or intensity of the oro-sensory exposure (*i.e.* oral residence time) may vary (Ferriday *et al.* 2016; Forde *et al.* 2013; Laguna and Sarkar 2016; Viskaal-van

Dongen, Kok and de Graaf 2011). For instance, in previous studies food manipulations to influence oral processing indirectly have involved the comparison of solid versus liquid forms of food, variations in viscosity or texture, or flavour intensities. The more direct influence of chewing on appetite ratings and food intake has been studied by varying the number of chews of a target food, and examining chewing gum interventions (Hogenkamp and Schiöth 2013; Robinson *et al.* 2014; Miquel-Kergoat *et al.* 2015). However, it is recognized that altering chewing in this way also varies oral residence time, eating rate, muscle fatigue and other oral processing attributes. Therefore, the effects of chewing in isolation is rarely studied due to the interrelated nature of these variables.

Lubrication is an important aspect of oral processing in addition to chewing *per se* (Laguna and Sarkar 2017; Laguna *et al.* 2017; Stokes, Boehm and Baier 2013). In-mouth lubrication may depend on the type of food consumed, its interactions with saliva and with the oral surfaces (*e.g.* tongue, teeth, oral palate). The mechanical properties of food can be evaluated using rheological measurements, such as viscosity, small and large deformation rheology. However, rheological measurements do not account for changes that occur in the food during the later stages of oral processing, such as the incorporation of saliva. Furthermore, the rheology of food during oral processing is not static; it is a highly dynamic process and the textural properties change continuously when the food is exposed to the oral cavity and becomes largely tribology-dominant, *i.e.* lubrication or friction dependent (Stokes, Boehm and Baier 2013). To that end, the lubricating effects arising from the incorporation of saliva can be measured using tribological measurements (Laguna and Sarkar 2017), a technique introduced relatively recently in food science. Although oral lubrication is an integral part of oral

processing, to date this has not been reviewed systematically with reference to satiety.

The main aim of this systematic review and meta-analysis was to understand the impact of oral processing, including both chewing and lubrication, on appetite and food intake. It was hypothesized that the enhancement of both chewing and lubrication during oral processing will affect appetite sensations, and reduce food intake. The main dependent variables included were: 1) subjective ratings of hunger and desire to eat as markers of appetite and readiness to eat, and 2) objective measures of energy intake following manipulation of food as a marker of satiation and meal termination. This review aimed to provide insights into potential oral processing manipulation strategies that could ultimately be applied to design foods offering enhanced satisfaction and satiety (Hetherington *et al.* 2013).

2.2. Materials and methods

The 2009 PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analysis) guidelines were used for reporting this systematic review (Liberati *et al.* 2009). The search strategy and inclusion criteria were specified in advance and documented in a protocol. This protocol was registered with the International prospective register of systematic reviews PROSPERO, registration number: CRD42016034019.

2.2.1. Search strategy

A systematic review attempts to collate all empirical evidence that fits pre-specified eligibility criteria to answer a particular research question. The research question of this systematic review was formulated using PICOS (Population, Intervention,

Comparison, Outcome, and Setting). The population was defined as healthy people with a healthy oral status that would not interfere with normal chewing and/or oral lubrication. The intervention was considered to be any manipulation directly or indirectly affecting oral processing characteristics, such as eating rate, oral residence time and number of chews, and where the comparison would involve two extreme conditions (see **Table 2.1**). For the outcomes, measures related to subjective appetite (hunger, desire to eat) and/or objectively measured food intake, as a consequence of manipulating oral processing, were included. The setting mostly involved a laboratory environment, but other settings were not excluded.

Table 2.1. Oral processing parameters as compared across studies.

Parameter[i]	**"Reduced" oral processing**	**"Enhanced" oral processing**
Bite size (5-15g)	Large	Small
Eating rate	Fast	Slow
Number of chews (10-40 chews)	Low	High
Oral residence time (3-30s)	Short	Long
Texture	Liquid (soft foods)	Semi-solid (hard foods)
Texture complexity	Low	High
Chewing gum	No gum	Gum

[i] In brackets: the lowest and highest values of the different oral processing parameters that were used in the different studies. For instance in the study by Cassady et al. 2009, the lowest number of chews was 10, whereas the lowest number of chews by Li et al. 2011 was 15 number of chews (for both the highest number of chews was 40 per mouthful).

A comprehensive literature search was conducted using six different online databases, including Cochrane Library, OVID Medline, PubMed, OVID Food Science and Technology Abstracts (FSTA), Web of Science (Thomson Reuters) and Scopus (Elsevier). The last search was run on 12 May 2017. Additional studies

were identified using the reference lists of the articles found in the search. Only articles published in English were included in this systematic review and no time limit was set. A broad range of search terms were used to increase the chance of locating all relevant literature. Three combined searches were performed in the six selected databases, linking chewing to satiety, lubrication to satiety and tribological measurements to satiety (this is related to lubrication, but extra search key words were added at a later stage). The search terms related to chewing were: ["oral processing" OR chewing OR mastication OR "structural breakdown" OR "food breakdown" OR "food destruction" OR "chewing cycle"]. The lubrication related search terms were: ["oral processing" OR "oral behavio*r" OR lubrication OR saliva OR "artificial saliva" OR "oral coating" OR "oral exposure" OR tongue]. For satiety the following search terms were used: [satiety OR satiation OR "expected satiety" OR "food intake" OR appetite OR hunger OR fullness OR "sensory specific satiety" OR "energy intake" OR "food behavio*r" OR "eating behavio*r"]. The selected key words for the added tribological variable were: [tribology OR tribometer OR thin-film rheology OR soft tribology OR tribol*].

The search in Scopus was limited to publications where the search terms appear in the title, abstract or keywords. No additional limitations were set for the other databases. The search strategy was validated by checking that a number of pre-selected relevant articles were indeed retrieved in at least one of the databases. The pre-selection was made during the orientation phase of literature research, focusing on more general articles based on the research topic, as well as articles found in previous related systematic review by Miquel-Kergoat *et al.* (2015). The citations of all found articles were exported to the reference software Endnote X7 for further processing.

2.2.2. Study selection

Only original research reports of human studies were included in this systematic review. The study selection phase was executed by first author EK. A summary of the selection procedure (PRISMA four-phase flow diagram) is given in **Figure 2.1**. The initial 12161 identified articles were reduced to 5825 after duplicates were removed. The remaining articles were screened for relevance based on their title. An additional 5505 studies were excluded based on the PICOS criteria. Research reports involving animal studies (2043), or medical studies on patients with certain diseases or disorders, studies with children, the elderly or participants of whom it was suspected that normal chewing was hindered (1762) were excluded. Additionally, articles not addressing the topic of interest were excluded (5464), as well as studies published in any other language than English (458). Some articles were excluded for multiple reasons, therefore the total number of articles is lower than the sum.

The remaining 320 articles were screened for their abstracts, resulting in the exclusion of an additional 241 articles (219 based on their topic, 17 were review papers without original data and 12 were meeting and conference abstracts, as well as posters presentation abstracts, and one was a data-set). The remaining number for the next screening step was n = 100, including an additional 21 articles that were identified through supplementary approaches. For example, the PRISMA statement for reporting systematic reviews (item 7 in Liberati *et al.* (2009)) advocates hand searches of the reference lists from screened articles so that relevant papers are not omitted.

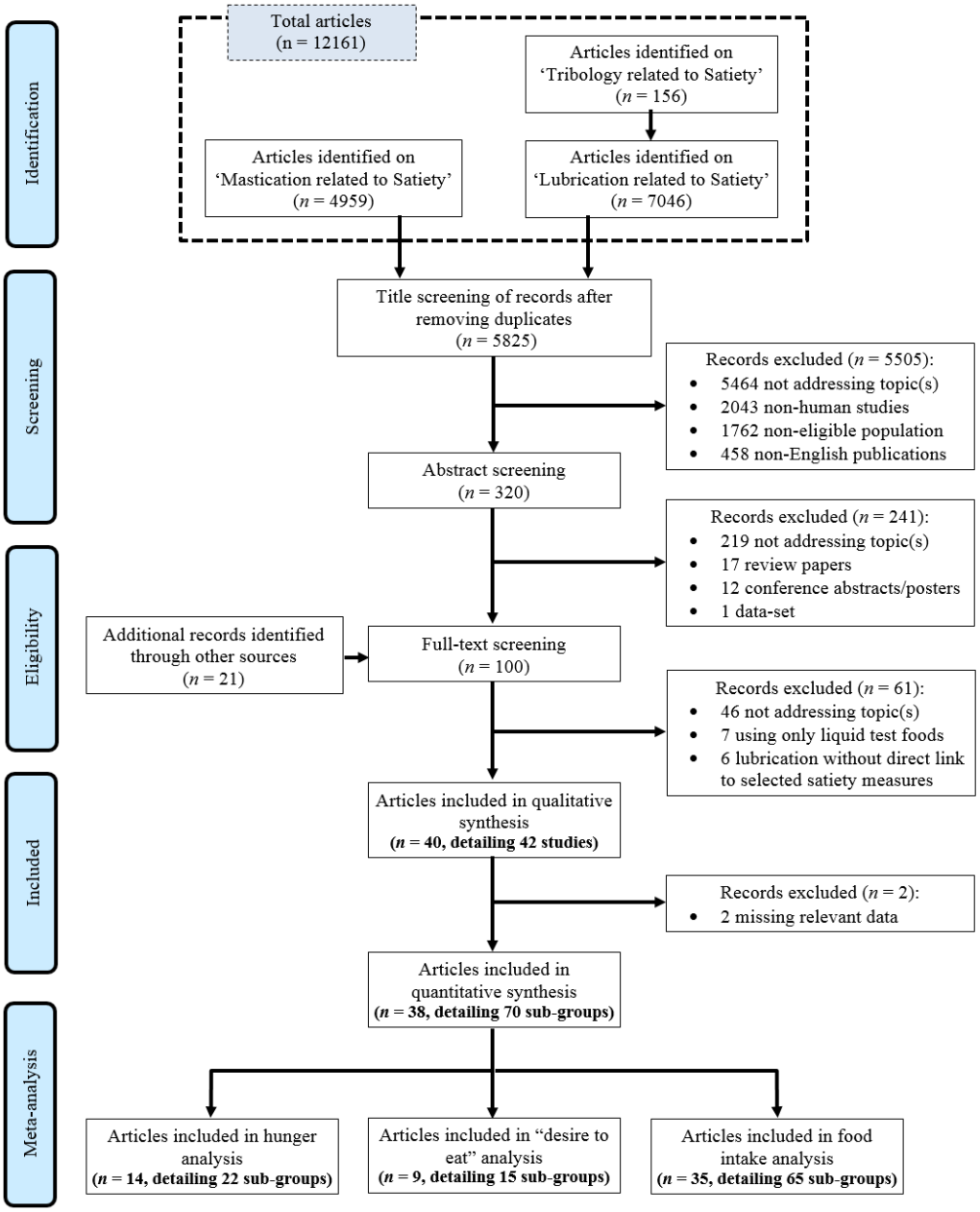

Figure 2.1. PRISMA flow-chart of the study selection procedure.

Finally, after assessing the full-text of these articles, another 61 articles were excluded for one or more reasons. Articles not addressing the topic of interest or studies aiming at validating new devices or methods (n = 46), articles where the two extreme oral processing characteristics were achieved by comparing two liquid products of for example differing viscosity (n = 7) and studies focusing on lubrication related parameters without direct measures of satiety/satiation (n = 6)

were eliminated, leading to a set of 40 articles. Two of those articles reported two independent studies (de Wijk *et al.* 2008; Zijlstra *et al.* 2008), bringing the total number of studies for qualitative synthesis to 42.

The quality assessment tool developed and validated by Moore (2012) was used to assess the quality of the included studies. Additionally, these 42 studies were critically appraised for risk of bias at both the study level and outcome levels. The quality and accuracy of a sample (~35%) of the extracted data was checked by authors MH and AS.

2.2.3. Study characteristics

Relevant information, such as study design, participant age, body mass index (BMI) status and gender ratio, as well as study outcomes on appetite ratings and food intake measures, was extracted from the 42 included studies. The key study characteristics are given in **Table 2.5**. In addition, means and standard deviations of the two most extreme outcome measures were extracted for the meta-analysis by author EK, as well as their statistical significance (p-values). The corresponding authors of more recent articles, where the values of interest were measured but not actually reported, were contacted with a data request. In the case of 9 articles (10 studies) data was received and incorporated into the current systematic research review (Cassady *et al.* 2009; Higgs and Jones 2013; Hogenkamp *et al.* 2010; Hogenkamp *et al.* 2012a; Hogenkamp *et al.* 2012b; Smit *et al.* 2011; Zijlstra *et al.* 2008, Study 1 and 2; Zijlstra *et al.* 2009b; Zijlstra *et al.* 2010) and in the case of the study by Ferriday *et al.* (2016) additional data was made publicly available online (Bosworth 2015).

All studies selected for qualitative synthesis were well-controlled experiments, in which participants were randomly assigned to experimental conditions. Of the 42 studies, all but two were laboratory based (Zijlstra *et al.* 2008, Study 1; Zijlstra *et al.* 2010) and all but two had a within subjects design (Hogenkamp *et al.* 2010; Higgs and Jones 2013). In only 10 of the studies, a power calculation was used to determine the number of participants needed to find a meaningful significant difference (Ferriday *et al.* 2016; Forde *et al.* 2013; Hogenkamp *et al.* 2012a; Lasschuijt *et al.* 2017; Martens *et al.* 2011; Martin *et al.* 2007; McCrickerd *et al.* 2017; Zhang, Leidy and Vardhanabhuti 2015; Zhu and Hollis 2014; Zhu, Hsu and Hollis 2013).

The total number of participants of all 40 studies included in the quantitative synthesis was 1711, arising from studies with samples varying from 9 to 120 participants, and involved mainly young adults (mean 25.1 years). Ideally studies should have an equal ratio of men to women, however for a number of studies more women than men were included, with six studies using more than 70% women (Bolhuis *et al.* 2014; Hetherington and Regan 2011; Higgs and Jones 2013; Hogenkamp *et al.* 2012a; Weijzen *et al.* 2008; Zijlstra *et al.* 2011). On the other hand, five studies included only males (Bolhuis *et al.* 2011; Labouré *et al.* 2002; Li *et al.* 2011; Martens *et al.* 2011; Zhu, Hsu and Hollis 2013), whereas only four studies included just females (Andrade, Greene and Melanson 2008; Komai *et al.* 2016; Park *et al.* 2016; Spiegel *et al.* 1993). Weight status varied across studies, with 20 studies specifically selecting participants within a healthy BMI range, five studies selecting people from specific weight groups to control for the influence of weight status whereas the remaining 15 studies did not specifically select or control for BMI. From those studies, there were two that also included participants with a

BMI higher than 25 (Julis and Mattes 2007; Martin *et al.* 2007). In most studies (29 out of 40), participants with any dietary restriction or dramatic weight change were specifically excluded as well as those who reported high levels of dietary restraint (27 out of 40) as assessed by either the Dutch Eating Behaviour Questionnaire (DEBQ) (van Strien *et al.* 1986) or the Three Factor Eating Questionnaire (TFEQ) (Stunkard and Messick 1985). None of the studies were double blinded, however in 22 studies the participants were distracted from the true aim through the use of a cover story.

In all studies, the researchers intended to vary only one characteristic of oral processing. However manipulating one characteristic inevitably had an effect on other characteristics (*i.e.* a higher eating rate might directly shorten the oral residence time). In 16 studies a test food was given with manipulated texture, such as liquid versus semi-solid food, and in two studies a texture complexity component was added. In six studies the number of chews per bite was manipulated, in three studies the oral residence time was directly influenced, and in five studies participants were instructed to eat at a specific chewing rate. Another three studies were included where the bite size was changed, and the final six studies looked at the influence of chewing gum on satiety and food intake during a later meal. For the purpose of the meta-analysis, the minimum and maximum oral processing characteristics were compared to one another (see **Table 2.1**). The maximum values were set as the commonly recommended values for reducing food intake and controlling appetite, such as small bites, high number of chews and long oral residence time (Christen and Christen 1997; Smit *et al.* 2011). In addition to the 26 studies that directly compared two oral processing parameters, the remaining 14 studies examined other intermediate oral processing conditions that were not

considered in this systematic review. However, in the case of the study by Zijlstra *et al.* (2009b) more separate conditions were considered in the meta-analysis; *i.e.* conditions comparing different oral residence times after ingestion of free-choice boluses of liquid food (which the authors called “bites”) as well as small and large boluses delivered with a peristaltic pump.

In the second search for papers linking lubrication or tribological parameters of food to satiety measures, a relatively small number of studies were found which had a comparable study design. Only six studies emerged investigating a link between a lubrication parameter and satiety. These papers are discussed separately and were not included in the meta-analysis, since most did not examine any direct satiety measure, or they measured expected satiety.

2.2.4. Meta-analysis

For the purpose of the meta-analysis, an additional two articles were excluded because appropriate data on a number of outcome measures were missing (Forde *et al.* 2013; Zandian *et al.* 2009). The remaining 38 articles, detailing 40 studies, were further divided in 70 subgroups (See **Figure 2.1**), as some studies provided more than one unique comparison group. Rather than combining these groups (study as unit of analysis), we entered each subgroup separately into the meta-analysis (subgroup within study as unit of analysis). These subgroups included the same experiment repeated with different test foods, indicated by Product A, B *etc.*, such as Labouré et al. Part A studying soups and Part B looking at rusks (Labouré *et al.* 2002), as well as studies with different participant groups, indicated by Group A, B *etc.*, such as Martin et al. Group A with all males and Group B with all females (Martin *et al.* 2007).

Table 2.2. Participant data of studies included in the meta-analyses.

Study ID and reference	*n* 1	*n* 2	Male	Female	Mean Age ± SD	Mean BMI ± SD
1 Andrade, Greene and Melanson (2008)	30		0	30	22.9 ± 7.1	22.1 ± 2.9
2 Bolhuis *et al.* (2011), Part A + B	55		55	0	22.0 ± 3.0	22.0 ± 2.0
4 Bolhuis *et al.* (2014), Step 1 + 2	50		11	39	24.0 ± 2.0	21.0 ± 2.0
6 Cassady *et al.* (2009)	13		8	5	24.0 ± 6.5	23.1 ± 1.4
7 Ferriday *et al.* (2016), Step 1 + 2 + 3 + 4	24		12	12	22.8 ± 3.8	21.8 ± 2.6
11 Hetherington and Boyland (2007), Part A + B	60		20	40	21.7 ± 4.0	22.7 ± 3.4
13 Hetherington and Regan (2011)	60		7	53	32.3 ± 10.7	26.2 ± 4.0
14 Higgs and Jones (2013)	14	13[i]	7	34	20.6 ± 8.8	21.0 ± 8.2
15 Hogenkamp *et al.* (2010), Part A + B	34	35[ii]	36	33	22.0 ± 3.0	21.6 ± 1.7
17 Hogenkamp *et al.* (2012a), Part A + B	53		12	41	21.0 ± 2.9	21.8 ± 2.0
19 Hogenkamp *et al.* (2012b), Part A + B + C + D + E + F	81[iii]		9	18	21.0 ± 2.4	22.2 ± 1.6
25 Julis and Mattes (2007)	47		29	18	24.0 ± 6.3	28.3 ± 2.6
26 Komai *et al.* (2016)	10		0	10	20.6 ± 1.9	20.0 ± 1.3
27 Labouré *et al.* (2002), Product A + B	12		12	0	21.5 ± 2.1	22.3 ± 2.1
29 Larsen *et al.* (2016), Step 1 + 2	26					
31 Lasschuijt *et al.* (2017), Part A + B	58		14	44	23.0 ± 9.0	22.0 ± 2.0
33 Lavin *et al.* (2002)	20		10	10		23.7 ± 3.1
34 Li *et al.* (2011), Group A	16		16	0	20.8 ± 0.8	20.1 ± 2.0
35 Li *et al.* (2011), Group B	14		14	0	20.4 ± 0.7	30.1 ± 3.0
36 Martens *et al.* (2011)	10		10	0	21.1 ± 3.9	22.4 ± 1.2
37 Martin *et al.* (2007), Group A	22		22	0	32.0 ± 11.8	30.9 ± 2.6
38 Martin *et al.* (2007), Group B	26		0	26	29.6 ± 8.8	29.4 ± 2.9
39 Mattes and Considine (2013), Group A	30		15	15	25.7 ± 8.4	21.2 ± 1.3
40 Mattes and Considine (2013), Group B	30		15	15	26.5 ± 8.4	32.7 ± 1.6
41 McCrickerd *et al.* (2017)	58		28	30	24.6 ± 4.5	22.1 ± 3.0
43 Mourao *et al.* (2007), Product A	40		20	20	23.2 ± 5.0	26.2 ± 1.5
44 Mourao *et al.* (2007), Product B	40		20	20	25.4 ± 7.5	26.3 ± 1.7
45 Mourao *et al.* (2007), Product C	40		20	20	24.8 ± 4.9	27.1 ± 1.6
46 Park *et al.* (2016), Group A	25		0	25	26.0 ± 8.0	22.0 ± 2.0
47 Park *et al.* (2016), Group B	25		0	25	36.0 ± 13.0	33.0 ± 3.0
48 Smit *et al.* (2011)	11		4	7		27.2 ± 6.4
49 Spiegel *et al.* (1993), Product A + B	18		0	18	28.8 ± 9.8	26.8 ± 7.2
51 Swoboda and Temple (2013)	44		21	23	31.1 ± 11.5	26.2 ± 5.2
52 Tang *et al.* (2016), Step 1 + 2	38		22	16	25.2 ± 3.4	
54 Weijzen *et al.* (2008)	59		5	54	28.4	22.3
55 de Wijk *et al.* (2008), Study 1	9		4	5		24.4
56 de Wijk *et al.* (2008), Study 2	10		6	4		25.3
57 Zhang, Leidy and Vardhanabhuti (2015)	12					
58 Zhu and Hollis (2014)	47		24	23	23.5 ± 6.4	28.0 ± 6.1
59 Zhu, Hsu and Hollis (2013)	21		21	0	24.0 ± 4.6	24.8 ± 2.7

Study ID and reference	*n* 1	*n* 2	Male	Female	Mean Age ± SD	Mean BMI ± SD
60 Zijlstra *et al.* (2011), Group A	27		6	21	36.0 ± 14.0	21.8 ± 1.6
61 Zijlstra *et al.* (2011), Group B	27		6	21	36.0 ± 14.0	30.5 ± 5.7
62 Zijlstra *et al.* (2010), Product A + B + C	106		45	61	24.0 ± 7.0	21.8 ±1.7
65 Zijlstra *et al.* (2008), Study 1	108		36	72	26.0 ± 7.0	22.7 ± 2.4
66 Zijlstra *et al.* (2008), Study 2	49		14	35	24.0 ± 6.0	22.2 ± 2.3
67 Zijlstra *et al.* (2009a)	32		12	20	22.0 ± 2.0	21.9 ± 2.2
68 Zijlstra *et al.* (2009b), Condition 1 + 2 + 3	22		8	14	21.0 ± 2.0	21.9 ± 1.5

[i] Between subjects design
[ii] Between subjects design
[iii] Within subjects design; 27 participants * 3 meals per day = 81 observations

Some subgroups were indicated with Step 1, 2 *etc*., such as Bolhuis et al. Step 1 for *ad libitum* course one: lunch, and Bolhuis et al. Step 2 for *ad libitum* course 2: dinner (Bolhuis *et al.* 2014), as well as Part A, B *etc.* to indicate different subgroups that did not necessarily have an effect on oral processing for example different energy density products or different test days as extra replicates. The participants' characteristics of all individual subgroups can be found in **Table 2.2**.

The meta-analysis was conducted on three outcome measures: subjective appetite ratings of hunger and desire to eat and objective measures of food intake (see **Tables 2.3 and 2.4**). Despite the importance of standardizing hunger levels before the oral processing manipulation, only seven studies provided a standard or preload meal (Bolhuis *et al.* 2011; Lasschuijt *et al.* 2017; Mourao *et al.* 2007; Zhang, Leidy and Vardhanabhuti 2015; Zijlstra *et al.* 2008, Study 1 and 2; Zijlstra *et al.* 2010). The oral processing intervention consisted of a fixed amount of food or was an *ad libitum* meal where food intake was measured. In some studies *ad libitum* intake was permitted during the oral processing intervention, and in others there was a fixed amount of food consumed. In one study *ad libitum* intake was measured twice, once during the oral processing intervention and again at the test meal (Bolhuis *et al.* 2014). Appetite ratings were measured at baseline on arrival in the lab and/or directly after the standard meal. Measurements were repeated directly after the oral processing intervention, and in some cases at 30 minute or hourly intervals after for a specific period of time.

Table 2.3. Meta-analysis data on appetite ratings.

Study ID and reference	Category	*n* 1	*n* 2	Test food type	Fasting time (h)	Hunger units	Mean Hunger[i] ± SD	Mean Hunger[ii] ± SD	Hunger p-value	DE[iii] units	Mean DE[i] ± SD	Mean DE[ii] ± SD	DE p-value
6 Cassady *et al.* (2009)	Number of chews	13		Solid	8	mm	-12.5 ± 15.7	-22.0 ± 20.5	<0.05				
7 Ferriday *et al.* (2016), Step 1	Eating rate	24		Meal	3	mm	-28.9 ± 23.4	-28.9 ± 25.3	<0.05				
9 Ferriday *et al.* (2016), Step 3	Eating rate	24		Meal	3	mm	-27.2 ± 31.0	-35.5 ± 24.3	<0.05				
14 Higgs and Jones (2013)	Chewing duration	14	13[iv]	Solid	2	mm	-55.7 ± 18.1	-45.1 ± 17.6	>0.1	mm	-47.0 ± 22.2	-45.0 ± 17.4	>0.1
15 Hogenkamp *et al.* (2010), Part A	Texture	33	35[v]	Liquid/Solid	8	mm	-14.3 ± 15.8	-15.0 ± 16.8	>0.05	mm	-15.5 ± 16.7	-19.0 ± 15.9	>0.05
16 Hogenkamp *et al.* (2010), Part B	Texture	34	35[vi]	Liquid/Solid	8	mm	-18.3 ± 19.8	-21.7 ± 20.2	<0.05	mm	-17.5 ± 19.4	-22.2 ± 21.1	>0.05
19 Hogenkamp *et al.* (2012b), Part A	Texture	81	78[vii]	Liquid/Solid	8	10-points	-16.4 ± 20.2	-25.7 ± 22.7	<0.0001	10-points	-9.8 ± 21.8	-15.9 ± 24.0	<0.0001
20 Hogenkamp *et al.* (2012b), Part B	Texture	81	81	Liquid/Solid	8	10-points	-10.9 ± 20.2	-15.9 ± 21.4	<0.0001	10-points	-7.3 ± 17.3	-9.3 ± 19.4	<0.0001
21 Hogenkamp *et al.* (2012b), Part C	Texture	81	81	Liquid/Solid	8	10-points	-12.3 ± 21.1	-15.7 ± 20.6	<0.0001	10-points	-7.6 ± 20.3	-10.8 ± 18.6	<0.0001
22 Hogenkamp *et al.* (2012b), Part D	Texture	81	81	Liquid/Solid	8	10-points	-18.0 ± 22.7	-22.7 ± 22.6	<0.0001	10-points	-6.5 ± 24.3	-13.6 ± 24.3	<0.0001
23 Hogenkamp *et al.* (2012b), Part E	Texture	81	80[viii]	Liquid/Solid	8	10-points	-15.6 ± 22.5	-21.7 ± 21.2	<0.0001	10-points	-6.6 ± 21.5	-13.4 ± 21.4	<0.0001
24 Hogenkamp *et al.* (2012b), Part F	Texture	79	81[ix]	Liquid/Solid	8	10-points	-14.6 ± 22.2	-19.5 ± 20.6	<0.0001	10-points	-9.2 ± 20.1	-11.4 ± 20.1	<0.0001
26 Komai *et al.* (2016)	Number of chews	10		Meal	10	mm	-64.6 ± 30.9	-67.1 ± 30.9	0.959				
27 Labouré *et al.* (2002), Product A	Texture	12		Liquid/Semi-solid	5.5	mm	-62.3 ± 27.2	-71.4 ± 22.2	>0.05				
28 Labouré *et al.* (2002), Product B	Texture	12		Liquid/Solid	5.5	mm	-54.3 ± 11.7	-72.8 ± 22.2	>0.05				
29 Larsen *et al.* (2016), Step 1	Texture complexity	26		Solid	3	mm	-8.7 ± 29.7	-11.9 ± 32.0	>0.05	mm	-8.1 ± 23.8	-10.8 ± 32.1	<0.05

Study ID and reference	Category	*n* 1	*n* 2	Test food type	Fasting time (h)	Hunger units	Mean Hunger[i] ± SD	Mean Hunger[ii] ± SD	Hunger p-value	DE[iii] units	Mean DE[i] ± SD	Mean DE[ii] ± SD	DE p-value
33 Lavin *et al.* (2002)	Texture	20		Liquid/Solid	2.5	mm	-7.0 ± 28.3	-2.0 ± 33.9	0.35				
36 Martens *et al.* (2011)	Texture	10		Liquid/Solid	3	mm	-44.1 ± 28.3	-51.4 ± 20.9	>0.05	mm	-38.6 ± 17.5	-54.0 ± 15.6	>0.05
52 Tang *et al.* (2016), Step 1	Texture complexity	38		Solid	3	mm	-3.7 ± 24.9	-5.5 ± 26.9	>0.05	mm	-5.4 ± 25.3	-7.0 ± 28.5	>0.05
57 Zhang, Leidy and Vardhanabhuti (2015)	Texture	12		Liquid/Solid	2.5	mm	-9.0 ± 10.4	-7.5 ± 8.0	>0.05	mm	-5.0 ± 8.7	-9.2 ± 8.7	>0.05
59 Zhu, Hsu and Hollis (2013)	Number of chews	21		Meal	8	mm	-23.5 ± 20.0	-25.0 ± 23.9	0.009	mm	-22.6 ± 20.0	-25.0 ± 20.0	0.002
67 Zijlstra *et al.* (2009a)	Texture	32		Liquid/Solid	12	10-points	-15.0 ± 23.0	-21.3 ± 22.5	>0.05	10-points	-10.8 ± 25.9	-19.2 ± 25.8	>0.05

[i] Large bite size, fast eating rate, low number of chews, short oral residence time and soft texture conditions
[ii] Small bite size, slow eating rate, high number of chews, long oral residence time and hard texture conditions
[iii] DE: Desire to Eat
[iv] Between subjects design
[v] Between subjects design; decreased sample size in n1 due to missing values
[vi] Between subjects design
[vii] Within subjects design, 27 participants * 3 meals per day = 81 observations, decreased sample size in n2 due to missing values
[viii] Within subjects design, 27 participants * 3 meals per day = 81 observations, decreased sample size in n2 due to missing values
[ix] Within subjects design, 27 participants * 3 meals per day = 81 observations, decreased sample size in n1 due to missing values

Table 2.4. Meta-analysis data on food intake.

Study ID and reference	Category	n 1	n 2	Mean Food intake[i] ± SD	Mean Food intake[ii] ± SD	Intake p-value
1 Andrade, Greene and Melanson (2008)	Eating rate	30		645.7 ± 155.9	579.0 ± 154.7	<0.01
2 Bolhuis *et al.* (2011), Part A	Eating rate	55		66.0 ± 33.6	90.0 ± 39.6	<0.05
3 Bolhuis *et al.* (2011), Part B	Eating rate	55		60.0 ± 30.0	82.8 ± 34.8	<0.05
4 Bolhuis *et al.* (2014), Step 1	Texture	50		737.0 ± 155.0	644.0 ± 173.0	<0.001
5 Bolhuis *et al.* (2014), Step 2	Texture	50		565.5 ± 179.4	540.2 ± 170.1	0.16
7 Ferriday *et al.* (2016), Step 1	Eating rate	24		640.8 ± 321.4	529.8 ± 238.5	0.004
8 Ferriday *et al.* (2016), Step 2	Eating rate	24		338.5 ± 190.6	297.8 ± 167.9	0.004
9 Ferriday *et al.* (2016), Step 3	Eating rate	24		196.3 ± 190.0	223.3 ± 189.6	0.35
10 Ferriday *et al.* (2016), Step 4	Eating rate	24		389.3 ± 223.2	423.2 ± 233.0	0.35
11 Hetherington and Boyland (2007), Part A	Chewing gum	60		461.3 ± 199.1	407.1 ± 217.1	<0.05
12 Hetherington and Boyland (2007), Part B	Chewing gum	60		164.8 ± 198.3	351.0 ± 176.8	>0.05
13 Hetherington and Regan (2011)	Chewing gum	60		247.5 ± 106.9	222.4 ± 108.4	0.029
14 Higgs and Jones (2013)	Chewing duration	14	13[iii]	270.5 ± 121.5	127.6 ± 97.8	0.01
15 Hogenkamp *et al.* (2010), Part A + B	Texture	68	70[iv]	555.9 ± 236.5	431.6 ± 186.0	0.03
17 Hogenkamp *et al.* (2012a), Part A	Texture	53		374.1 ± 198.5	274.4 ± 119.9	<0.0001
18 Hogenkamp *et al.* (2012a), Part B	Texture	53		458.3 ± 171.3	369.3 ± 165.5	<0.0001
19 Hogenkamp *et al.* (2012b), Part A	Texture	81		1767.0 ± 581.0	1720.0 ± 583.0	0.56
20 Hogenkamp *et al.* (2012b), Part B	Texture	81		1886.0 ± 465.0	1850.0 ± 546.0	0.56
21 Hogenkamp *et al.* (2012b), Part C	Texture	81		2016.0 ± 582.0	1941.0 ± 560.0	0.56
22 Hogenkamp *et al.* (2012b), Part D	Texture	81		1549.0 ± 427.0	1496.0 ± 438.0	0.56
23 Hogenkamp *et al.* (2012b), Part E	Texture	81		1537.0 ± 418.0	1554.0 ± 460.0	0.56
24 Hogenkamp *et al.* (2012b), Part F	Texture	81		1589.0 ± 448.0	1588.0 ± 407.0	0.56
25 Julis and Mattes (2007)	Chewing gum	47		1415.0 ± 747.3	1330.0 ± 822.7	>0.05
27 Labouré *et al.* (2002), Product A	Texture	12		776.9 ± 299.6	790.3 ± 204.4	>0.05
28 Labouré *et al.* (2002), Product B	Texture	12		939.6 ± 301.2	703.6 ± 376.6	>0.05
29 Larsen *et al.* (2016), Step 1	Texture complexity	26		982.4 ± 445.6	622.4 ± 302.6	<0.01
30 Larsen *et al.* (2016), Step 2	Texture complexity	26		377.8 ± 197.4	292.0 ± 175.6	0.08
31 Lasschuijt *et al.* (2017), Part A	Texture	58		75.9 ± 21.7	54.1 ± 22.1	<0.001
32 Lasschuijt *et al.* (2017), Part B	Texture	58		70.6 ± 21.7	51.7 ± 22.1	<0.001
33 Lavin *et al.* (2002)	Texture	20		884.4 ± 209.4	766.6 ± 222.2	<0.05
34 Li *et al.* (2011), Group A	Number of chews	16		555.0 ± 111.0	477.8 ± 72.4	0.021
35 Li *et al.* (2011), Group B	Number of chews	14		695.0 ± 127.9	625.0 ± 106.2	0.021
37 Martin *et al.* (2007), Group A	Eating rate	22		1020.0 ± 248.0	918.0 ± 225.0	<0.05
38 Martin *et al.* (2007), Group B	Eating rate	26		588.0 ± 212.0	585.0 ± 216.0	>0.05

Study ID and reference	Category	n 1	n 2	Mean Food intake[i] ± SD	Mean Food intake[ii] ± SD	Intake p-value
39 Mattes and Considine (2013), Group A	Chewing gum	30		2009.2 ± 414.6	1879.2 ± 452.4	0.056
40 Mattes and Considine (2013), Group B	Chewing gum	30		2146.8 ± 452.4	2339.8 ± 452.4	0.059
41 McCrickerd *et al.* (2017), Part A	Texture	58		300.0 ± 84.5	271.6 ± 72.3	<0.001
42 McCrickerd *et al.* (2017), Part B	Texture	58		546.3 ± 216.3	483.9 ± 204.1	<0.001
43 Mourao *et al.* (2007), Product A	Texture	40		1915.0 ± 815.9	1665.0 ± 638.8	0.03
44 Mourao *et al.* (2007), Product B	Texture	40		1970.0 ± 619.8	1752.0 ± 619.8	0.026
45 Mourao *et al.* (2007), Product C	Texture	40		2517.0 ± 1138.4	2116.0 ± 695.7	0.016
46 Park *et al.* (2016), Group A	Chewing gum	25		563.0 ± 270.0	511.0 ± 270.0	>0.05
47 Park *et al.* (2016), Group B	Chewing gum	25		676.0 ± 270.0	613.0 ± 270.0	>0.05
48 Smit *et al.* (2011)	Number of chews	11		702.2 ± 125.0	612.8 ± 111.9	0.006
49 Spiegel *et al.* (1993), Product A	Bite size	18		770.0 ± 237.7	784.0 ± 297.1	>0.05
50 Spiegel *et al.* (1993), Product B	Bite size	18		883.3 ± 283.0	833.3 ± 283.0	>0.05
51 Swoboda and Temple (2013)	Chewing gum	44		254.5 ± 150.6	227.3 ± 195.7	>0.05
52 Tang *et al.* (2016), Step 1	Texture complexity	38		793.0 ± 246.7	696.9 ± 296.1	<0.01
53 Tang *et al.* (2016), Step 2	Texture complexity	38		235.2 ± 73.1	246.8 ± 90.6	0.839
54 Weijzen *et al.* (2008)	Bite size	59		192.0 ± 132.1	169.1 ± 128.6	0.02
55 de Wijk *et al.* (2008), Study 1	Texture	9		402.5 ± 213.5	222.8 ± 27.1	0.003
57 Zhang, Leidy and Vardhanabhuti (2015)	Texture	12		830.0 ± 405.3	809.0 ± 426.1	>0.05
58 Zhu and Hollis (2014)	Number of chews	47		760.0 ± 371.1	647.1 ± 322.6	0.001
59 Zhu, Hsu and Hollis (2013)	Number of chews	21		1098.3 ± 546.0	1117.6 ± 668.9	0.851
60 Zijlstra *et al.* (2011), Group A	Texture	27		572.5 ± 270.0	376.5 ± 198.3	<0.05
61 Zijlstra *et al.* (2011), Group B	Texture	27		600.0 ± 251.3	369.8 ± 166.2	<0.05
62 Zijlstra *et al.* (2010), Product A	Texture	106		406.6 ± 323.8	393.7 ± 321.9	>0.05
63 Zijlstra *et al.* (2010), Product B	Texture	106		174.4 ± 113.2	164.8 ± 112.3	>0.05
64 Zijlstra *et al.* (2010), Product C	Texture	106		592.0 ± 372.6	565.8 ± 340.3	>0.05
65 Zijlstra *et al.* (2008), Study 1	Texture	108		788.5 ± 386.0	567.9 ± 312.1	<0.0001
66 Zijlstra *et al.* (2008), Study 2	Eating rate	49		226.8 ± 122.4	176.6 ± 88.3	0.01
67 Zijlstra *et al.* (2009a)	Texture	32		394.8 ± 212.9	371.5 ± 178.0	>0.05
68 Zijlstra *et al.* (2009b), Condition 1	Chewing duration	22		427.7 ± 185.2	416.4 ± 189.9	0.0008
69 Zijlstra *et al.* (2009b), Condition 2	Bite size	22		406.1 ± 153.2	294.2 ± 159.8	<0.0001
70 Zijlstra *et al.* (2009b), Condition 3	Bite size	22		447.4 ± 165.4	359.1 ± 185.2	<0.0001

[i] Large bite size, fast eating rate, low number of chews, short oral residence time, soft texture and no chewing gum conditions

[ii] Small bite size, slow eating rate, high number of chews, long oral residence time, hard texture and chewing gum conditions

[iii] Between subjects design

[iv] Between subjects design; n1: 34 participants * 2 energy density products = 68 observations, n2: 35 participants * 2 energy density products = 70 observations

Appetite ratings were measured on 100 mm Visual Analog Scales (VAS) or categorical rating scales. The 10-point or 5-point scores were converted to a 100 point scale, so appetite ratings could be better compared against each other. When appetite was assessed at multiple time points after the oral processing manipulation, the ratings directly after the end of manipulation were retrieved. To control for differences in appetite levels before the start of the study due to varying fasting states, for example, the change in mean appetite level was computed (raw mean difference, *e.g.* hunger level after chewing intervention minus the baseline hunger level). Food intake was measured after the chewing manipulation in either weight (g) or energy (kcal or kJ). Where needed, given values were converted to kcal to standardize the measurement units. Mean, standard deviation and sample size for each group were extracted for all papers where they were reported. To account for differences in the measurement scales, the standardized mean difference (SMD) was used to compute the effect size (Borenstein *et al.* 2009). The studies employing a between subjects design were treated as independent studies, whereas the studies employing a within subjects design were considered as dependent studies. For the food intake studies a correlation coefficient of 0.5 was assumed and for the appetite studies a correlation coefficient of 0.2. Both correlation coefficients were based on the few studies where raw data was available to determine the actual correlation coefficients (Cassady *et al.* 2009; Ferriday *et al.* 2016; Hetherington and Boyland 2007; Hogenkamp *et al.* 2012b; Smit *et al.* 2011).

Since the studies from our sample used different methodologies, the meta-analysis was performed using a random effects (RE) model. The heterogeneity was assessed with the I^2 statistic as indicator for the percentage of statistically

meaningful variability between studies. An I^2 value of 0% means there is no heterogeneity that needs to be explained, values of 25% are considered low, 50% moderate and above 75% is considered high (Higgins *et al.* 2003). If heterogeneity between studies was considered high, we tried to explain this further by implementing a mixed effects (ME) model with a number of moderators, such as fasting time, participants' age and BMI status. To investigate risk of publication bias across the studies, funnel plots were produced. A funnel plot is used to visually represent high oral processing effect estimates from individual studies against the standard error of each study. Typically the precision of an estimate increases with the size of the study, with studies with a small sample size distributed towards the bottom of the plot and studies with a larger sample size scattered towards the narrower top of the funnel plot as they are more precise. The different shades of the funnel plot correspond to the 90% confidence interval CI (white), 95% CI (light grey) and 99% CI (dark grey). The free statistical software R® (version 3.3.1) and the metaphor package (version 1.9-9) were used to conduct the meta-analyses (forest plots and funnel plots). The software Comprehensive Meta-Analysis (version 2.2) was used to conduct the sensitivity and group effect analyses, as well as the Egger's tests to assess publication bias (Egger *et al.* 1997).

Table 2.5. Characteristics of studies included in the systematic review[i].

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Andrade, Greene and Melanson (2008)	30	0/30	UW, NW, OW and OB	Randomized, 2-arm, within subjects design	Pasta meal	*Ad libitum* lunch with fast/big bite/no pauses and slow/small bite/chew 20-30 times/pauses condition	VAS	No difference in appetite ratings	Weighing	Yes, under slow eating condition weight and energy intake ↓ compared to fast eating
Bolhuis *et al.* (2011)	55	55/0	NW	Randomized, 6-arm, cross-over design	Tomato soup	Three conditions (2 or 3 s oral exposure each of 5 or 15 s, respectively, or free bite size) for two salt concentrations	VAS	No difference in appetite ratings	Weighing	Yes, intake was ↑ in short oral exposure condition compared to long (34 %)
Bolhuis *et al.* (2014)	50	11/39	NW	Randomized, 2-arm, cross-over study, within subjects	Hamburger/ rice salad	*Ad libitum* lunch of hard or soft foods, followed by *ad libitum* dinner to test if energy intake was compensated	VAS	No difference in appetite ratings	Weighing	Yes, ↓ intake of hard foods, ↓ energy intake and ↓ eating rate compared to soft foods
Cassady *et al.* (2009)	13	8/5	NW	Randomized, 3-arm, cross-over design, within subjects (no control group, *i.e.* 0 g almonds)	Almonds	55 g almonds (11 x 5 g portions) chewed for 10, 25 or 40 times	VAS	Yes, ↓ hunger with 40 chews than with 25 chews (no diff. with 10 chews)		NA
Ferriday *et al.* (2016)[ii], Product A and B	24	12/12	NW	Counterbalanced, randomized, 4-arm, cross-over design, within subjects, sample size power calculation	Beef stew with dumplings/ fish, chips and peas	Two fixed test meals with maximized differences in oral processing, followed by *ad libitum* same meal or dessert, and 1 h later *ad libitum* snack intake	VAS	Yes, ↑ fullness after eating slow meal than after fast meal	Weighing	Yes, ↓ food intake after slow meal than after fast meal
Forde *et al.* (2013)	15	5/10	NW	Full cross-over design, within subjects, randomized within test days, sample size power calculation	35 different food items	50 g portions of 35 different food items, across 5 consecutive days, images of 200 g portions for expected satiety assessment (separate descriptive sensory analysis panel, n = 11)	VAS	Yes, ↓ hunger with increased chewing and longer oral exposure time and smaller bite size		NA

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Hetherington and Boyland (2007)	60	20/40	UW, NW and OB	Repeated measures, counter-balanced (Latin-square), within subjects design	Sweet or salty snack	Fixed lunch, followed by 4 conditions (no gum sweet snack; no gum salty snack; gum sweet snack; gum salty snack), with gum chewed at 3 time points after lunch and ad libitum intake measured 3 h later	VAS	Yes, ↓ hunger and ↑ fullness in chewing gum condition for sweet and savory snacks, with ↓ desire to eat sweet snacks but not savory snacks	Weighing	Yes, ↓ snack intake in chewing gum condition for sweet and savory snacks
Hetherington and Regan (2011)	60	7/53	NW, OW and OB	Repeated measures, counter-balanced, within subjects design	Sweet or salty snack	Restrained eaters: given a fixed lunch, followed by 4 conditions (no gum sweet snack; no gum salty snack; gum sweet snack; gum salty snack), with gum chewed at 4 time points after lunch and ad libitum intake measured 3 h later	VAS	Yes, ↓ hunger, desire to eat and ↑ fullness in chewing gum condition at 2 and 3 h after lunch	Weighing	Yes, ↓ snack intake in chewing gum condition
Higgs and Jones (2013)	41	7/34	NW	Three groups, between subjects design	Sandwich	Fixed lunch with 3 conditions (habitual chewing n = 13; 10 s pauses between each mouthful n = 14; 30 s chewing before swallowing n = 14) and its influence on *ad libitum* snack intake 2 h later	VAS	No difference in appetite ratings	Weighing	Yes, ↓ snack intake in 30s chewing condition
Hogenkamp *et al.* (2010)	105	46/59	NW	Randomized, 3-arm, between subjects design	Yoghurts	*Ad libitum* yoghurt presented in three groups (liquid-yoghurt/straw n = 34, liquid-yoghurt/spoon n = 36 and yoghurt-pudding/spoon n = 35)	VAS	No difference in appetite ratings	Weighing	Yes, intake on first exposure ↑ for liquid/straw compared to semi-solid/spoon
Hogenkamp *et al.* (2012a)	53	12/41	NW	Randomized, 2-arm, cross-over, within subjects design, sample size power calculation	Milk-based custards	*Ad libitum* intake on day 1 and 5, and fixed amount on day 2, 3, and 4 of low vs high expected satiety samples	VAS	No difference between *ad libitum* liquid and solid	Weighing	Yes, liquid product intake ↑ than semi-solid

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Hogenkamp *et al.* (2012b)	27	9/18	NW	Randomized, 4-arm, cross-over, within subjects design	Novel gelatine products	Fixed product conditions (liquid/semi-solid and low/high energy density) eaten with 3 *ad libitum* main meals a day for three days	10-point categorical scale	Yes, ↑ hunger directly after liquid compared to semi-solid food	Weighing	No difference in intake between liquid and semi-solid preload condition
Julis and Mattes (2007)	47	29/18	OW and OB	Randomized, 3-arm, within subjects design	Free	Fixed lunch 3 conditions (no chewing gum, fixed time gum chewing and gum chewing after first hunger occurrence)	VAS	No difference in appetite ratings	Questionnaire	No difference in snack intake between chewing gum conditions
Komai *et al.* (2016)[iii]	10	0/10	NW	Randomized, 2-arm, within subjects design	Hamburger, rice and soup	Fixed solid meal with 30 CPM or pureed meal without chewing (0 CPM)	VAS	No difference in appetite ratings		NA
Labouré *et al.* (2002), Product A and B	12	12/0	NW	Randomized, 5-arm, within subjects design	Soups and rusks	Fixed lunch sessions with five products with different textures, followed by an *ad libitum* dinner	VAS	No difference in appetite ratings	Dinner energy and macro-nutrient content	No difference in energy intake at dinner
Larsen *et al.* (2016)	26	m/f	NW	Randomized, 2-arm, cross-over, within subjects design	Gelatine-agar gels	Fixed preload of high or low complexity model foods, followed by a two-course *ad libitum* meal	VAS	No difference in appetite ratings	Weighing	Yes, ↓ intake after high complex food compared to low complex food
Lasschuijt *et al.* (2017)	58	14/44	NW	Randomized, 4-arm, cross-over, within subjects design, samples size power calculation	*κ*-carrageenan /locust bean gum gels	*Ad libitum* portion of model foods varying in hardness and sweetness	VAS	No difference in appetite ratings	Weighing	Yes, ↓ intake after hard compared to soft model foods
Lavin *et al.* (2002)	20	10/10	NW and OW	Four-arm, within subjects design, randomization unclear	Sucrose containing drink/jelly/ pastilles and water	Four preloads (consumed with varying oral durations) with *ad libitum* meal served immediately after preload	VAS	No difference in appetite ratings	Weighing	Yes, energy intake ↓ after pastilles compared to water and the sweet drink

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Li *et al.* (2011)[iv]	30	30/0	NW + OB	Randomized, 2-arm, within subjects design	Pork pie	*Ad libitum* habitual breakfast with 2 conditions (15 chews or 40 chews, found to be lowest and highest possible chews/bite)	VAS	No difference in appetite ratings	Weighing	Yes, after 40 chews energy intake ↓ than after 15 chews
Martens *et al.* (2011)	10	10/0	NW	Randomized, 2-arm, cross-over, within subjects design, sample size power calculation	Chicken breast	Fixed lunch of whole or blended chicken breast (soup)	VAS	No difference in appetite ratings		NA
Martin *et al.* (2007)	48	22/0	OW and OB	Randomized, 3-arm, between subjects design, sample size power calculation	Chicken	Baseline meal (normal eating rate), reduced-rate meal (by 50 %), combined-rate meal (50 % slower during second half of the meal)	VAS	No difference in appetite ratings	Weighing	No, food intake did not differ between conditions
Mattes and Considine (2013)	60	30/30	NW + OB	Randomized, 3-arm, cross-over, within subjects design	Pasta meal	Three treatments (no gum, soft or hard gum) chewed at 1 chew/s for 15 min while sipping grape juice through a straw, followed by a 6 h blood collection and *ad libitum* lunch and free dinner at home	VAS	No difference in appetite ratings	Weighing + Food record	No difference in energy intake in any of the meals during the test day, however, trend to reduce energy intake in lean participants and increase energy intake in obese participants
McCrickerd *et al.* (2017)[v]	61	30/31	NW	Counterbalanced, randomized, 4-arm, between subjects design, sample size power calculation	Rice based porridge	*Ad libitum* intake at breakfast of thin and thick porridge with low and high energy density	VAS	No difference in appetite ratings	Weighing	Yes, ↓ intake of thick compared to thin porridge
Mourao *et al.* (2007), Product A, B and C	40	20/20?	NW and OB	Randomized, 6-arm, cross-over, between subjects design (in sub-groups within subjects design)	Milk/cheese, Watermelon juice/fruit and Coconut milk/coconut meat	*Ad libitum* lunch and fixed amount of water, liquid or solid test food with either high carbohydrate, high protein or high fat content	VAS	No difference in appetite ratings between products or BMI status	Weighing	Yes, for all three foods daily intake was ↑ in liquid condition compared to solid foods

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Park *et al.* (2016)	25	0/25	NW + OB	Randomized, 2-arm, cross-over, within subjects design	Sweet or salty snack	Fixed lunch, followed by 4 conditions (no gum sweet snack; no gum salty snack; gum sweet snack; gum salty snack), with gum chewed at 3 time points after lunch and ad libitum intake measured 3 h later	VAS	Yes, chewing gum ↓ hunger over time compared to not chewing gum	Weighing	No difference in snack intake between chewing gum conditions
Smit *et al.* (2011)	11	4/7	NW and OB	Counterbalanced, randomized (for last 2 treatments), within subjects design	Pasta meal	Pilot study with 3 treatments (*ad libitum* chewing, 10 or 35 chews per mouthful: CPM)	VAS	No difference in appetite ratings	Weighing	Yes, after 35 CPM food intake ↓ than after 10 CPM
Spiegel *et al.* (1993), Product A and B	18	0/18	NW and OB	Counterbalanced for bite size, randomized, alternating products between sessions, within subjects design	Sandwich rolls and bagels	*Ad libitum* lunch with food varying in bite size (sandwiches 5, 10 and 15 g; bagels 6 or 12 g) tested on separate days	VAS	No difference in appetite ratings due to bite size	Weighing	No difference in meal size due to different bite sizes in either products even though the food texture was very different and was eaten at very different ingestion rates (g/min)
Swoboda and Temple (2013)[vi]	44	21/23	OW	Randomized, within subjects design (with different subjects for part 1 and 2)	Fruit, sweet or savory snack	Two separate studies: one-day acute effect of chewing gum and effect of chewing gum before each meal for a week	VAS	Yes, chewing either mint or fruit gum ↓ hunger compared to no gum	Weighing	Yes, chewing mint-flavored gum ↓ healthy food intake compared to no gum (however no effect on snack food or total energy intake, nor with fruit gum)
Tang *et al.* (2016)	38	22/16	NW	Single-blind, randomized, 3-arm, cross-over, within subjects design	Gelatine-Agar gels	Fixed preload of high, medium or low complexity model foods, followed by 2 *ad libitum* meal courses	VAS	No difference in appetite ratings	Weighing	Yes, ↓ intake after high complex food compared to low and medium complex food
Weijzen *et al.* (2008)	59	5/54	NW and OW	Randomized, 4-arm cross-over, within subjects design	Biscuits with chocolate/ hazelnut cream filling	Either morning or afternoon *ad libitum* snack intake with snacks varying in size and weight, as well as usual or extra attention paid during consumption	5-point categorical scale	Not reported	Weighing	Yes, snack intake of nibbles ↓ than of bars

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
de Wijk *et al.* (2008), Study 1	9	4/5	NW and OW	Counterbalanced, randomized, 2-arm, within subjects design (different subjects between Study A and Study B)	Chocolate dairy products	*Ad libitum* intake by straw with fixed eating rate and fixed meal duration (20 s intervals over 15 min = 45 bites of *ad lib* bite size)	10-point categorical scale	No difference in appetite ratings between liquid and semi-solid foods	Weighing	Yes, semi-solid food intake ↓ than liquid food intake
de Wijk *et al.* (2008), Study 2	10	6/4	NW and OW	Counterbalanced, randomized, 3-arm, within subjects design (different subjects between Study A and Study B)	Chocolate dairy products	*Ad libitum* intake of 45 bites by peristaltic pump with varying oral processing time (5 or 9 s for semi-solid only) and with eliminated bite effort (*ad lib* bite size)	10-point categorical scale	No difference in appetite ratings between liquid and semi-solid foods	Weighing	No difference in energy intake between liquid and semi-solid food, nor due to oral processing time for semi-solid food
Zandian *et al.* (2009)	47	0/47	NW	Two groups (decelerated and linear eating rate), within subjects design	Rice meal	Increased eating rate (40 % more food in same amount of time) and decreased eating rate (30 % less food in same time)	VAS	No difference in appetite ratings	Mandometer	Yes, changing someone's habitual eating rate affected food intake
Zhang, Leidy and Vardhanabhuti (2015)	12	m/f	NW and OW	Randomized, 5-arm, cross-over, within subjects design, sample size power calculation	Protein snacks	Protein beverages at pH 3 or pH 7, or acid or heated treated gels compared to a water control sample, followed by *ad libitum* lunch	VAS	No difference in appetite ratings	Weighing	No difference in food intake between protein snacks
Zhu and Hollis (2014)	47	24/23	NW, OW and OB	Randomized, 3-arm, cross-over, within subjects design, sample size power calculation	Pizza rolls	*Ad libitum* lunch (no beverage) with predetermined average number of chewing cycles used as baseline for the three treatments (100, 150 and 200 %)	VAS	No difference in appetite ratings for treatment or BMI even after a 60 min period	Weighing	Yes, food intake ↓ for 200 % chews compared to 100 % baseline number of chews

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Zhu, Hsu and Hollis (2013)	21	21/0	NW and OW	Randomized, 2-arm, within subjects design, sample size power calculation	Pasta meal	Fixed pizza meal with 2 chewing conditions (15 and 40 chews), followed by *ad libitum* pasta meal 3h later	VAS	Yes, hunger after 40 chews ↓ compared to 15 chews (however fullness not different)	Weighing	No difference in food intake at lunch meal 3h after chewing intervention
Zijlstra *et al.* (2011)	54	12/42	NW + OB	Randomized, cross-over, within subjects design	Rice meal and yoghurt	*Ad libitum* lunch, two sessions of 45 min with a neutrally and highly liked product	VAS	No, satiety ratings for both products were similar, while significantly more calories were consumed with yoghurt	Weighing over time	Yes, ↑ *ad libitum* intake for yoghurt compared to rice
Zijlstra *et al.* (2010), Product A, B and C	106	45/61	NW	Randomized, 6-arm, cross-over, within subjects design (with 7th session to measure eating rate)	Luncheon meat, vegetarian meat replacer and chewy candy	*Ad libitum* snack intake while watching 90 min movie (with two breaks of 15 min in between) receiving 3 x 400 g) of three different product types with different levels of hardness	VAS	No difference in appetite ratings between hard and soft versions of all food products	Weighing	No difference in intake between hard and soft version of all food products
Zijlstra *et al.* (2008), Study 1	108	36/72	NW	Randomized, 3-arm, cross-over, within subjects design (different subjects between study 1 and 2)	Chocolate dairy products	*Ad libitum* intake while watching 90 min movie (with two breaks of 15 min in between) receiving 3 x 1500 g portions	10-point categorical scale	No difference in appetite ratings between liquid, semi-liquid and semi-solid foods	Weighing	Yes, semi-solid food intake ↓ than liquid food intake
Zijlstra *et al.* (2008), Study 2	49	14/35	NW	Randomized, 6-arm, cross-over, within subjects design (different subjects between study 1 and 2)	Chocolate dairy products	*Ad libitum* snack intake under 3 conditions (free eating rate with effort, free eating rate without effort and fixed eating rate without effort at 10 s intervals)	10-point categorical scale	No difference in appetite ratings between liquid and semi-solid foods	Weighing	Yes, controlling eating rate and effort had an effect on food intake (for both products, no difference between products). No effect of effort alone (but semi-solid intake ↓ compared to liquid food intake)

Reference	Participants			Study information			Outcomes			
	n	**Gender (M/F)**	**BMI groups**	**Study design**	**Test food**	**Test procedure**	**Appetite method**	**Effect appetite**	**Food intake method**	**Effect food intake**
Zijlstra *et al.* (2009a)	32	12/20	NW	Randomized, 2-arm, cross-over, within subjects design	Chocolate dairy products	*Ad libitum* snack intake after fixed intake of liquids and semi-solids as breakfast time	10-point categorical scale	No difference in appetite ratings between liquid and semi-solid foods	Weighing	No difference in chocolate cake intake after consumption of a liquid or semi-solid product
Zijlstra *et al.* (2009b), Condition 1, 2 and 3	22	8/14	NW	Randomized, 7-arm, cross-over, within subjects design	Chocolate dairy product	Control vs different bite size (free, 5 or 15 g) and oral processing time (3 or 9 s) for at least 30 min	10-point categorical scale	Yes, significant effect of condition on hunger after intake	Weighing	Yes, ↓ intake for 9 s oral processing time than for 3 s Yes, ↓ intake for 5 g bite size than for 15 g

[i] CPM: Chews Per Mouthful, NW: Normal Weight, OB: Obese: OW: Over Weight, UW: Under Weight, VAS: Visual Analytical Scale
[ii] Two studies were reported, only Study 2 was included in this review
[iii] Two studies were reported, only Study 2 was included in this review
[iv] Two studies were reported, only Study 2 was included in this review
[v] Two studies were reported, only Study 1 was included in this review
[vi] Two studies were reported, only Study 1 was included in this review

2.3. Results

A total of 40 articles, that included 42 studies, were found suitable for qualitative analysis (see **Figure 2.1** and **Table 2.5**).

2.3.1. Effect of food oral processing on appetite

Based on the 42 studies that measured appetite ratings, 10 found significant effects on the appetite ratings, such as hunger, fullness and desire to eat. This disparity in the results may be associated with the study methodology employed, such as having a fixed amount of food to chew. For example, Cassady *et al.* (2009) provided their participants with a fixed amount of almonds to chew for different number of times (10, 25 or 40 chews). They found that a larger number of chews significantly reduced appetite. A fixed amount of food was also given during the manipulation of oral processing in five other studies that found a significant effect on appetite (Ferriday *et al.* 2016; Forde *et al.* 2013; Hogenkamp *et al.* 2012b; Zhu, Hsu and Hollis 2013; Zijlstra *et al.* 2009b). When *ad libitum* meals were provided, participants ate until they reached a certain level of fullness, so the change in appetite ratings was similar regardless of the amount consumed or how much energy was ingested. If an excess amount of food is offered in an *ad libitum* meal, the motivation to eat may be stronger than the oral processing manipulation itself.

2.3.2. Effect of oral processing on food intake

Four studies did not measure *ad libitum* food intake during or after the oral processing intervention (Cassady *et al.* 2009; Forde *et al.* 2013; Komai *et al.* 2016; Martens *et al.* 2011), and therefore were not considered in this section of the review. Thus, the total number of studies that measured food intake was 38. Food intake was measured either at the same time as the oral processing intervention

occurred, *e.g.* number of chews was manipulated during an *ad libitum* meal (Li *et al.* 2011), or after the oral processing manipulation, *e.g.* Zhu, Hsu and Hollis (2013).

The effect of oral processing on objective measures of food intake was significant in 26 studies, but no clear patterns were evident. The provision of a fixed meal to standardize hunger before the oral processing intervention was linked to a significant effect in food intake in seven studies (Bolhuis *et al.* 2011; Hetherington and Boyland 2007; Hetherington and Regan 2011; Lasschuijt *et al.* 2017; Mourao *et al.* 2007; Zijlstra *et al.* 2008, Study 1 and 2), which seems to highlight the importance of a standardized meal to ensure a similar level of hunger between participants before the oral processing manipulations.

2.3.3. Effect of lubrication on appetite and food intake

Six articles were identified that mentioned some links between lubrication and satiety (see **Table 2.6**). McCrickerd, Chambers and Yeomans (2014) tested the satiety effects of fruit drinks varying in thickness and creaminess. The viscosity and lubrication profiles of the test drinks showed that the thickened drinks were more viscous and more lubricating, having a lower traction coefficient than the thin drinks. No effect was found on satiety ratings, but they did observe a difference in food intake where female participants self-selected a smaller portion size when the drink's visual sensory characteristics indicated it would be more satiating (McCrickerd, Chambers and Yeomans 2014). A limitation of this study was that participants were allowed to self-select their own portion size in a glass from a larger amount of the drink in a jug, after assessing the sensory characteristics. The results might have been clearer if the sensory aspects were evaluated by a different

panel, and if the panellists were instructed to drink directly from a larger or fixed amount to ensure satiation. A mindful assessment of the drink attending to the sensory features of the drinks before *ad libitum* intake might have influenced the results. Moreover, as also suggested by the authors, the portion size effect might have had a bigger influence on intake than the texture manipulation. It was suggested that the average portion size for men was bigger than the serving glass could hold, but was smaller for women. Therefore the portion size could explain the lack of effect found in male participants, while there was an effect for female participants.

In a study by Morell *et al.* (2014) the effect of four different hydrocolloids in milkshakes with similar viscosity during pouring and handling conditions on expected satiety was investigated. They found that the starch granules (mainly in modified starch) swell up and disintegrate in presence of artificial saliva. However, the structural properties of guar gum and λ-carrageenan milkshakes remained more or less intact. In addition, the modified starch milkshake had a higher expected satiety. It was hypothesized that expected satiety was more linked to the initially perceived thickness and creaminess of foods and that the loss of structure in presence of saliva is linked to a melting sensation of the modified starch in the mouth (Morell *et al.* 2014). However, this melting sensation could be a function of better lubrication, which in this case seems to be related to higher expected satiety, suggesting later stages of oral processing could be just as important to satiety perceptions as the initial stages. In addition, Stribeck analysis of these milkshakes with or without saliva was not performed to confirm whether the milkshakes had significantly different friction coefficients in the mixed regime.

Table 2.6. Characteristics of studies involving lubrication measures.

Reference	Participants			Study information			Outcomes		
	n	**Age**	**BMI status**	**Study design**	**Test food**	**Test procedure**	**Lubrication measure**	**Effect appetite**	**Effect food intake**
Gavião, Engelen and van der Bilt (2004)	16	35 ± 13	NA	Randomized, 3-arm, within subjects design	Parafilm, Melba toast with and without margarine, breakfast cake and cheese	Parafilm and 3 different types of food products were chewed and expectorated in duplicate, and salivary flow rate was measured	Flow rate significantly increased due to mechanical stimulation by Parafilm and by food. Dry foods had longer oral exposure time than more moist products, while flow rate was similar. Toast with margarine reduced chewing duration and number of chewing cycles	NA	NA
Joyner, Pernell and Daubert (2014)	7	NA	NA	Randomized, 16-arm, within subjects design	Acid milk gels containing thickeners	16 acid milk gel samples, tested for sensory texture attributes in QDA, as well as instrumental rheological and tribological properties	Starch had an impact on friction behaviour of acid milk gels, and addition of artificial saliva resulted in a change of frictional behaviour across the entire range of sliding speeds	NA	NA
Lett, Norton and Yeomans (2016)	34	***Range:*** 18-37	22.7 ± 1.6	Randomized, 2-arm, within subjects design	Emulsions with different droplet size	Fixed preload emulsions with a droplet size of 2 or 50 µm, followed by an *ad libitum* pasta lunch	Rheological and lubrication properties for the two emulsions were comparable (results not published at this time)	Yes, ↓ hunger after 2 µm compared to 50 µm preload (however fullness not different)	Yes, food intake after 2 µm preload ↓ than after the 50 µm preload
McCrickerd, Chambers and Yeomans (2014)	48	20.8 ± 5.3	NW	Randomized, 4-arm, within subjects design	Fruit drinks,, containing thickeners and creamy flavourings	*Ad libitum* intake of 4 iso-energetic fruit drinks varying in texture (thin vs thick) and creamy flavour (low vs high creaminess)	Both instrumental viscosity and lubrication (Stribeck) properties were measured, with the thick drinks being more viscous and more lubricating. The creamy flavour additions did not affect the physical texture of the drinks (both viscosity and lubrication)	No difference in appetite ratings	Yes, for females consumption of the thick drink ↓ than the thin drink. However, no differences found in food intake for males, or due to creamy flavour, regardless of gender

Reference	Participants			Study information			Outcomes		
	n	**Age**	**BMI status**	**Study design**	**Test food**	**Test procedure**	**Lubrication measure**	**Effect appetite**	**Effect food intake**
Morell *et al.* (2014)	106	***Range:*** 18-61	NA	Randomized, 4-arm, within subjects design	Milkshakes, containing thickeners	Sip-test of 4 milkshakes with consumer panel using CATA questionnaires	The swollen starch granules in modified starch disintegrated in presence of artificial saliva	Yes, modified starch had the highest satiety expectation score, and native starch, guar gum and λ-carrageenan the lowest as linked to their sensory creamy sensations when entering the mouth	NA
Morell *et al.* (2015)	121	NA	NA	Randomized, 6-arm, within subjects design	Yoghurts, containing added protein and thickeners	Spoonful test of 6 yoghurts with consumer panel	Physically modified starch granules remain unaltered in presence of α-amylase from artificial saliva leading to a thick, dense and creamy yoghurt that could lead to a longer oro-sensory exposure	Yes, samples which were perceived as thicker and denser were perceived as having a higher satiating capacity	NA

In another study by Morell *et al.* (2015) the influence of different proteins and presence of starch in yoghurts was studied in relation to expected satiety. In line with their previous study, it was found that addition of starch, as well as addition of protein, increased expected satiety with whey protein having more potential to increase expected satiety than skimmed milk powder. The breakdown of starch in presence of saliva and linked melting sensation was not found here, as the starch granules were incorporated in the protein network, aggregating upon exposure to artificial saliva (Morell *et al.* 2015).

In a study by Gavião, Engelen and van der Bilt (2004) several oral processing characteristics of different food products were determined. Dry Melba toast resulted in a longer oral residence time with more chewing cycles, whereas the addition of margarine reduced the time until swallowing as well as the number of chews. This was largely attributed to the lubricating effects of butter facilitating bolus formation (Gavião, Engelen and van der Bilt 2004), however no quantitative tribological measurement of the bolus was performed to confirm such findings. Joyner, Pernell and Daubert (2014) tested the friction behaviour of acid milk gels with and without the addition of saliva. The addition of saliva was found to cause a significant change in the frictional behaviour of the acid milk gels, with a stronger effect seen in samples containing starch (Joyner, Pernell and Daubert 2014). However, in both of these studies no direct link was made with any satiety parameters. Finally, Lett, Norton and Yeomans (2016) have shown the effects of physicochemical characteristics (*e.g.* droplet size) of model (emulsions) affecting hunger and food intake. They highlight that the tribological and rheological properties of these emulsions are the same; however, exact coefficients of friction

at orally relevant speeds are not mentioned (Lett, Norton and Yeomans 2016; Lett *et al.* 2016). These reports suggests that there is growing interest in lubrication measurements but these have yet to be studied in depth for a potential contribution (if any) to satiety and food intake.

2.3.4. Meta-analysis

The 38 articles included in the meta-analysis were divided into 70 individual subgroups. The narrative part of this systematic review indicated that for the two appetite ratings (hunger and desire to eat), the different methodology of a fixed or *ad libitum* meal might have significant effects on the study outcomes. The studies were divided into groups where either a fixed amount was used for the oral processing manipulation (Type 1), or where an *ad libitum* amount of food was presented (Type 2). For the meta-analysis on hunger ratings, 14 Type 1 studies including 22 subgroups and 14 Type 2 studies with 22 subgroups reported data. The studies where chewing gum was used to manipulate oral processing, and thus no food was ingested, were not included in the meta-analysis for appetite.

Figure 2.2 shows the meta-analysis results of the Type 1 studies. The results confirmed that a higher level of oral processing had a significant effect on reducing hunger ratings (-0.20 effect size, 95% confidence interval CI: -0.30, -0.11, I^2 statistic = 0%). The meta-analysis was also performed with both the Type 1 and Type 2 studies included, and the results remained similar (-0.21 effect size, 95% CI: -0.27, -0.15, I^2 = 0%). The ME model using moderators indicated that the included moderators were unable to better explain the total amount of heterogeneity, as the heterogeneity level was already 0%. Subgroup analysis revealed that the oral processing variables eating rate and texture had a significant

effect on hunger ratings, whereas bite size, oral residence time, number of chews and texture complexity on their own did not affect hunger. It is however important to note that few studies were included for the latter variables, where no significant effect was found.

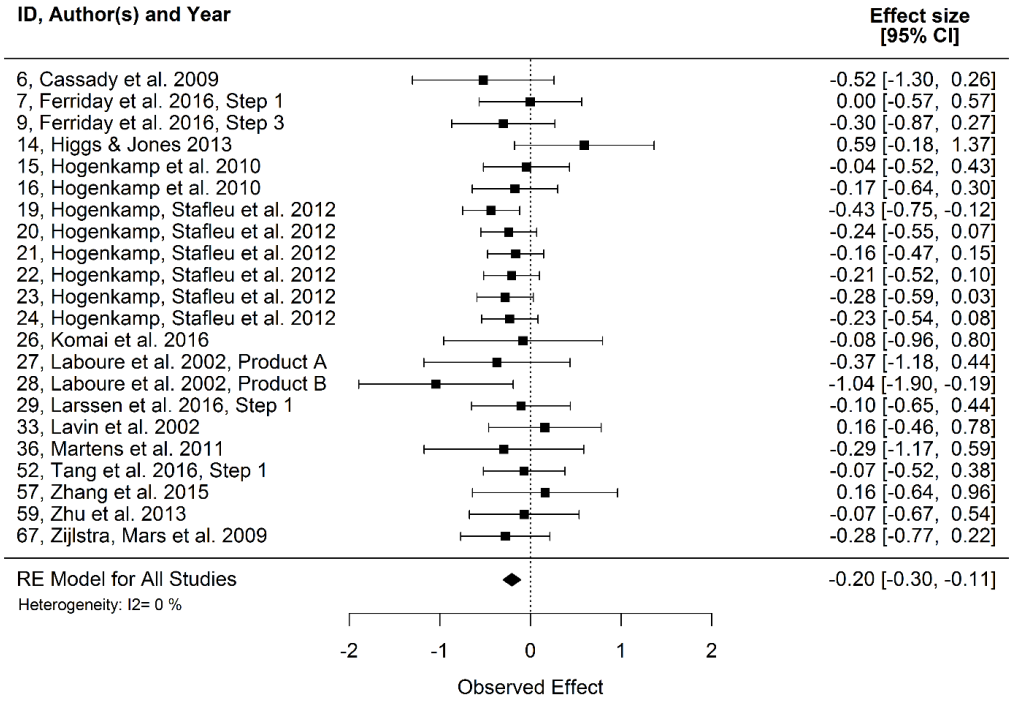

Figure 2.2. Forest plot of oral processing effects on the SMD of hunger ratings with corresponding 95% CI. The pooled estimates were obtained using RE modelling. The I^2 value is a measure of the approximate proportion of total variability in point estimates that can be attributed to heterogeneity.

For the desire to eat ratings, 9 studies including 15 subgroups reported data. The meta-analysis showed similar results to that of the hunger ratings namely that higher oral processing reduced self-reported desire to eat (-0.21 effect size, 95% CI: -0.31, -0.10, I^2 = 0%, see **Figure 2.3**).

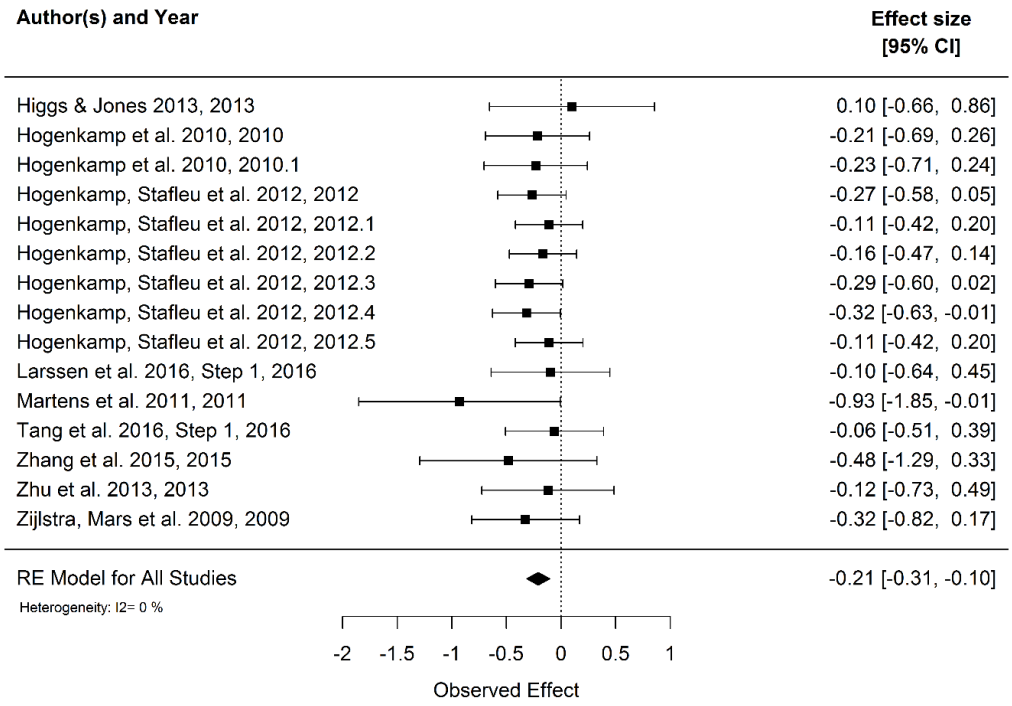

Figure 2.3. Forest plot of oral processing effects on the standardized mean difference (SMD) of desire to eat ratings with corresponding 95% confidence interval (CI). The pooled estimates were obtained using a random effects (RE) modelling.

Meta-analysis of the food intake data included 35 studies with 65 subgroups. Study 2 by de Wijk *et al.* (2008) did not provide the standard deviations for food intake and therefore was not included in the meta-analysis. A significant effect of oral processing reducing food intake was found (-0.28 effect size, 95% CI: -0.36, -0.19, I^2 = 61.52%), as can be observed in **Figure 2.4**. This is in line with what we expected, given the large amount of individual studies that found a significant effect. The I^2 value did indicate a moderate level of heterogeneity, however the ME model using moderators did not result in a consistent improvement. Subgroup analysis revealed that there was no significant effect of oral residence time alone on food intake, however there were only two studies that looked specifically at oral residence time.

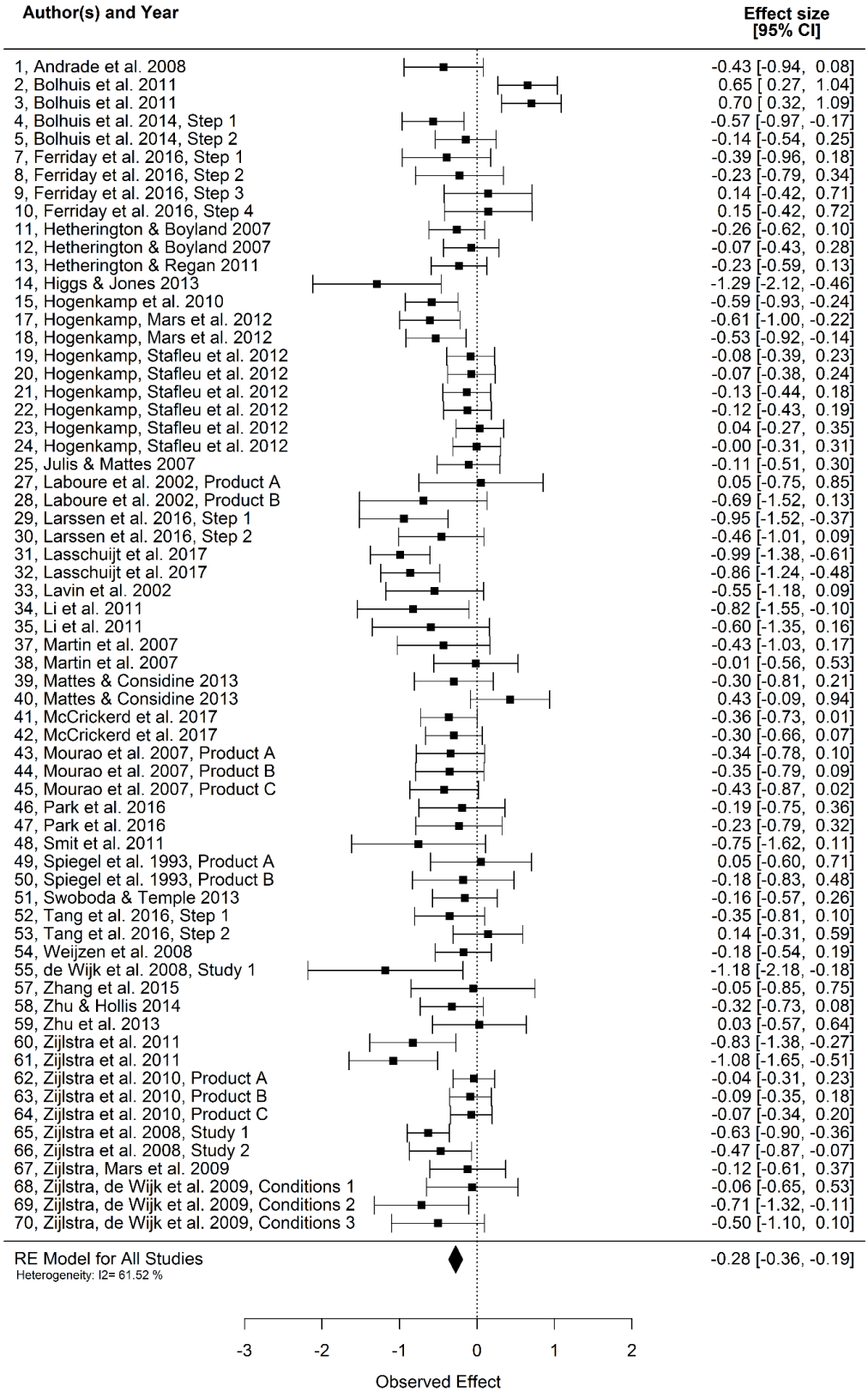

Figure 2.4. Forest plot of oral processing effects on the SMD of food intake with corresponding 95% CI. The pooled estimates were obtained using RE modelling.

The other oral processing factors all included more than two studies, and all showed a significant effect on reducing food intake. Furthermore, as there are different processes that might affect food intake over time, such as cephalic-phase responses in anticipation of food after eating chewing gum or cognitive processes due to the increased expected satiating power of harder, thicker and chewier food, the meta-analysis outcome was tested when Type 1 studies were excluded. However, when only looking at the studies that measured *ad libitum* food intake at the same time as the oral processing intervention, the outcome was not affected (-0.45 effect size, 95% CI: -0.55, -0.35, I^2 = 69.06%).

Publication bias was assessed using funnel plots and the Egger's regression test. The funnel plot for the hunger ratings (**Figure 2.5**) shows a relatively good distribution over the vertical axis, indicating that studies with different sample sizes were included.

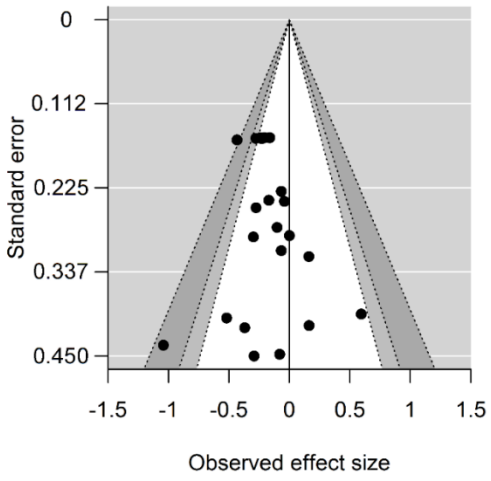

Figure 2.5. Funnel plot of oral processing effects on hunger ratings.

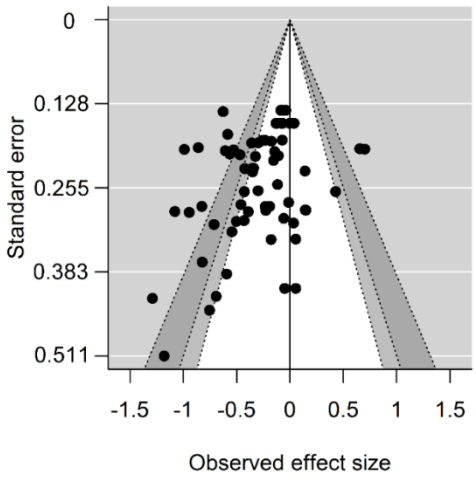

Figure 2.6. Funnel plot of oral processing effects on food intake[i].

[i] The different shades in the plot correspond to the 90% CI (white), 95% CI (light grey) and 99% CI (dark grey).

However, the majority of the studies clustered towards to the left of the mean, indicating there might be evidence of publication bias. Nevertheless, this visual impression was not supported by the Egger's test (*p = 0.17*, CI: -1.01, 0.18). The asymmetry in the funnel plot for food intake in **Figure 2.6** also shows a potential bias in favour of studies that found oral processing had an effect on lowering food intake. This was confirmed by the Eggers's test (*p = 0.000*, CI: -3.59, -1.25).

2.4. Discussion

The main aim of this systematic review and meta-analysis was to understand the impact of oral processing, including chewing and lubrication, on appetite and food intake. It was hypothesized that enhanced oral processing would affect appetite sensations, and reduce food intake. Oral processing is an important factor in the development of satiation and satiety. The results of this review indicate that self-reported appetite and measured food intake are influenced by manipulating components of oral processing such as eating rate, texture and chewing. Thus, where participants are instructed to use a certain oral processing strategy such as the number of times a food is chewed, this will alter how much is eaten. Where participants are provided with foods which increase oral residence time, and/or slow the rate of eating, this reduced subjective appetite. The analyses demonstrate that increased oral processing appears to promote satiation, although it is difficult to isolate which specific component is directly influencing the outcome. Larsen *et al.* (2016) developed a model food where the oral residence time was kept constant while texture complexity was varied. This enabled the study to examine texture complexity controlling for oral exposure time. They found that providing a more complex, orally stimulating first course promoted satiation and reduced food intake

at a subsequent second course. Therefore, enhanced oral processing through greater textural complexity, can lead to enhanced satiety.

Few studies have been performed focusing on the effects of oral lubrication on appetite and satiety, even though this is an aspect that is also manipulated when looking at foods with differently designed textures (*e.g.* soft vs hard). Additionally, it is worth noting that saliva has an important role in the cephalic phase linked to amylase digestion (Giduck, Threatte and Kare 1987), however this was not within the scope of the present review and we have only considered the lubrication (tribological) aspects of saliva.

The results of these meta-analyses suggest that varying different components of oral processing taken together, can have a significant influence on reducing hunger ratings and food intake. Overall, from the literature included in this systematic review, it is clear that all studies involved a relatively small number of participants (varying from 9 to 120) and short-term exposures (only once in most studies). Studies with a larger sample size involving longer well-described replicable interventions (from weeks to months) are needed to understand the impact of oral processing on long-term satiety enhancement and its potential in weight management. In addition, product differences need to be large enough to be detectable by consumers to find a potential influence due to oral processing.

The lack in standardization of study design is a key limitation identified by this systematic review. Blundell *et al.* (2010) have advocated that for all studies of satiation and satiety, a framework should be applied to standardize procedures; as was also suggested by the results in this review, by standardization of prior hunger levels using a fixed meal before the oral processing intervention takes place, the

actual study effects can be studied more carefully (Blundell *et al.* 2010). The recommended study procedure for satiation studies includes a standard, fixed meal based on individuals' estimated daily energy needs before oral processing is manipulated. Furthermore, for satiety studies, the satiety quotient, the time until the next eating occasion, should be reported in addition to subjective hunger ratings and how much is eaten at the next eating occasion (Blundell *et al.* 2010). Thus, conclusions regarding the effects of oral processing on satiety must be made with caution since varying results may be attributable to differences in study design. Moreover, dimensions such as food type, meal occasion, differences between individuals or specific participant groups, such as male/female (Martin *et al.* 2007) or low/high BMI status (Mattes and Considine 2013; Zhu and Hollis 2014), appeared to have an influence on the outcome as well.

A systematic review and meta-analysis by Robinson *et al.* (2014) studied the effects of the specific oral processing characteristic of eating rate on hunger and energy intake. They concluded that a slower eating rate led to a lower energy intake as compared to a faster eating rate, and that different ways in which eating rate could be manipulated (directly or indirectly) did not alter the outcome. No effect of eating rate on hunger was found directly after the meal or up to 3.5h after the meal, both in the analysis with *ad libitum* studies as well as the fixed studies. The difference with our results on the hunger ratings could be explained by including more oral processing variables, and also many more studies were included (five compared to 22 subgroups in the current review with fixed amounts of foods). Another systematic review by Miquel-Kergoat *et al.* (2015) compared the outcome measure of hunger ratings and energy intake under different oral processing conditions, with the addition of gut hormones and metabolites. Besides hunger

ratings, meta-analyses in the current review focused on food intake and desire to eat data, thereby broadening the scope of the review. Also, the oral processing definition was expanded to include aspects of lubrication and saliva incorporation. Finally, oral processing parameters were grouped together according to the recommended oral processing strategies commonly suggested for better weight management such as slow eating rates, high number of chews and longer oral resident time (Christen and Christen 1997; Ford *et al.* 2010; Smit *et al.* 2011). Moreover, additional data not included in the original publication was requested from authors. Instead of comparing 13 subgroups as was reported by Miquel-Kergoat *et al.* (2015), the current review included hunger ratings from 22 subgroups. Therefore, the present review allows a more comprehensive and advanced analysis by broadening the scope of the used measures, expanding the search to include lubrication, and performing detailed analysis using raw data from authors.

2.5. Conclusions

In this study we conducted a comprehensive systematic review to assess different oral processing characteristics on appetite ratings and food intake. In order to address this quantitatively, a meta-analysis was undertaken to test the effect size of self-reported appetite ratings and objectively measured food intake in studies that manipulated oral processing parameters, such as oral residence time, texture, eating rate, chewing and lubrication. The meta-analysis demonstrated that manipulating oral processing through slow eating rates and textural complexity reduced subjective appetite and greater oral processing through strategies such as greater chewing reduced food intake.

Although evidence was found for the effects of oral processing on appetite ratings and food intake, this systematic review identified a clear gap in knowledge on the influence of saliva incorporation and oral lubrication on appetite ratings and food intake. The influence of the lubrication parameters of food (pre and post mixing with saliva) on appetite and food intake remains largely unquantified. Furthermore, the studies involving lubrication did not perform tribological measurements of the food and the bolus to quantify differences in lubrication profiles. Future research should be conducted following the framework outlined by Blundell *et al.* (2010) and standardize prior hunger before oral processing manipulations, which should be apparent and not subtle. With carefully planned and standardized procedures, the knowledge base on the importance of all aspects of oral processing, including both chewing and lubrication, for satiation and satiety development will be expanded and potential application to weight management can be explored. Such knowledge, together with longer interventions, are needed to underpin the creation of the next generation of foods for weight management and allow the development of coordinated public health strategies to tackle obesity.

The next chapters of this thesis will focus on studying the influences of oral processing including both chewing and lubrication on satiation and satiety. The instrumental fracture properties of different hydrocolloid gels were measured (**Chapter 3**) and a number of hydrogels with varying properties were selected for further study. Then, the selected hydrogels were characterised by instrumental and sensory techniques as related to chewing (fracture properties) and oral lubrication (friction properties) (**Chapter 4**). After that, it was attempted to explain the oral processing behaviour of the different hydrogels for a group of participants by the food structural properties and by the eating capabilities of the tested participants

(**Chapter 5**). And then finally, the effects of model hydrogels with different chewing and oral lubrication characteristics on the hunger levels and food intake of a snack were examined (**Chapter 6**).

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Chapter 3

Selection of hydrogels

Abstract

In order to study the effects of oral processing on satiety, focussing both on the chewing and oral lubrication properties, model preload foods were designed based on their fracture properties. The fracture properties of different κ-carrageenan (κC) hydrocolloid gels were determined using a wide range of concentrations, mixing ratios with locust bean gum (LBG) or sodium alginate (NaA), and levels of complexity using calcium alginate beads (CaA) of 300 or 1000 μm, at 1-4 wt% total hydrocolloid concentrations. First bite oral processing fracture properties were determined using a Texture Analyser with a Volodkevitch bite jaw probe. Seven hydrogel samples were selected that were considered to be edible for a human trial, and that differed in their fracture stress. In addition, hydrogels were selected based on their potential differences in lubricity and complexity, so that differences may be achieved in their oral lubrication properties.

3.1. Background on hydrocolloids

Results of the systematic review and meta-analysis identified gaps in the literature in relation to the specific effects of both chewing and oral lubrication on satiation and satiety. Ideally, to investigate the potential effects of chewing and oral

lubrication on short-term regulation of satiety and food intake, a preload-test meal paradigm is applied (Blundell *et al.* 2009). A preload is defined as an eating episode smaller than a meal (usually about 1 MJ) and is given at a particular time interval (often 30 to 90 min) before a test meal (Blundell *et al.* 2009). Hydrocolloid based model systems have been widely used to study the influence of rheological properties on food oral processing (Hayakawa *et al.* 2014; Hori *et al.* 2015; Kohyama *et al.* 2015), and can easily be used as a small-scale preload food. Although the influence of hydrogel hardness on oral residence time is relatively well researched, there has been limited literature on the influence of structural properties of gels on other oral processing characteristics.

Hydrocolloids are a heterogeneous group of hydrophilic long chain polymers that can form viscous dispersions and/or gels upon dispersion in water due to the presence of large number of hydroxyl (-OH) groups (Saha and Bhattacharya 2010). By using hydrocolloid model systems, the structural and rheological properties can be specifically modified within a relatively simple matrix. Texture design of real food products, by comparison, is more challenging, where it is much more difficult to change specific aspects of texture due to their rather complex structures and interactions between different components (Szczesniak 1990). In order to study the oral processing effects of chewing and oral lubrication, a selection of different hydrocolloid model gels were investigated. Hydrocolloid gelation can be either thermo-reversible or thermo-irreversible (Ahmed 2015). The textural properties of hydrogels vary widely due to the type of used hydrocolloid, their concentration as well as any mixture of hydrocolloids, and thus may have very different sensory perceptions (Hayakawa *et al.* 2014). Hydrocolloids that have been frequently used in research on gel systems are

alginate, agar, carrageenan, locust bean gum, gelatin, gellan, pectin and xanthan (Banerjee and Bhattacharya 2012; Nishinari, Zhang and Ikeda 2000; Saha and Bhattacharya 2010). Using a combination of multiple hydrocolloids can be useful to improve the rheological and sensory characteristics of gels. The type of synergy may depend on the association or non-association of two different hydrocolloid molecules (Williams and Phillips 2000). Oppositely charged molecules are likely to associate and may form a precipitate or result in gel formation. Hydrocolloids that do not associate may appear to form a single homogeneous phase in the case of low concentrations, whereas at higher concentrations they might phase separate (Williams and Phillips 2000).

For the purposes of this PhD project, no fats or sugars were introduced to the model systems and hydrocolloids were selected for their ability to form gels that do not melt in the mouth during oral processing. Thus, carrageenans, locust bean gum and alginates were selected based on their thermo-irreversibility, *i.e.* no change in gel structure is expected when exposed to oral temperatures of 37 °C. Also, the selected gels were targeted for their resistance to oral enzymes, and therefore starch gels were not considered.

3.1.1. Carrageenans

Carrageenans are large, highly flexible molecules, consisting of linear sulphated polysaccharide chains, and are extracted from red seaweeds (*Rhodophyceae*). They consist of high molecular weight linear polysaccharides comprising repeating galactose units and 3,6-anhydrogalactose, both sulphated and non-sulphated, joined by alternating α-(1,3) and β-(1,4) glycosidic links (Imeson 2000). The three major carrageenan types are kappa (κC), iota (ιC) and lambda (λC), as shown in

Figure 3.1, with each differing in degree of sulphation (Kariduraganavar, Kittur and Kamble 2014). The thickening and gelling properties of the different carrageenan types vary widely. Upon cooling hot aqueous dispersions of *κ*C and *ι*C to between 40 and 60 °C, gelation occurs. Carrageenan gels are thermos-reversible at 5 to 20 °C above the gelling temperatures, but properties do not change at 37 °C, and the gels can exhibit hysteresis (Imeson 2000). Whereas *ι*C can form soft elastic gels in the presence of calcium ions (Ca^{2+}), *λ*C does not form a gel and is mainly used as a thickener in dairy products (Kariduraganavar, Kittur and Kamble 2014).

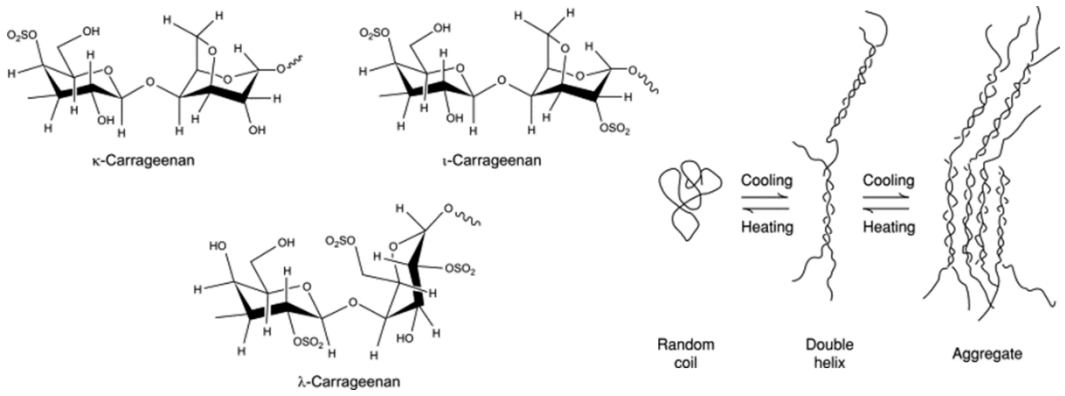

Figure 3.1. Schematic representation of the molecular structures of *κ*-, *ι*- and *λ*-carrageenan (left), and gelation mechanism (right), adapted from Kariduraganavar, Kittur and Kamble (2014) and Williams and Phillips (2003).

The gelation process of *κ*C involves coil-to-helix transition of the molecules followed by aggregation that occurs upon cooling (see **Figure 3.1**), particularly in presence of potassium ions (K^+), and forms strong, rigid gels (Morris, Rees and Robinson 1980; Kariduraganavar, Kittur and Kamble 2014). In the presence of K^+, the *κ*C network forms rigid rod-like aggregates (Meunier *et al.* 1999), increasing the hardness and fracture properties of the gels (Zhu, Bhandari and Prakash 2018). The electrostatic repulsions along the carrageenan polymer backbone are screened

by like charges from the K^+ ions and weaken the electrostatic attraction between semi-ester sulphates, neutralising them. As a consequence, helix-helix formations are promoted and *κ*C aggregates are more easily formed, increasing the gel strength (Rey and Labuza 1981; Zhu, Bhandari and Prakash 2018). Similarly, Ca^{2+} ions might act between adjacent *κ*C helices, neutralising the repulsion between carrageenan strands (Zhu, Bhandari and Prakash 2018). However, the excess presence of the divalent Ca^{2+} ions might cause precipitation, resulting in syneresis (*i.e.* the release of water from the *κ*C network) and a decrease in gel strength (Lai, Wong and Lii 2000; MacArtain, Jacquier and Dawson 2003). Thus, *κ*C in presence of K^+ ions was used in this PhD for its ability to form strong gels.

3.1.2. Locust bean gum (LBG)

Locust bean gum (LBG), also known as carob gum, is a galactomannan vegetable gum that is extracted from the seeds of the carob tree (*Ceratonia siliqua*). As can be seen in **Figure 3.2**, the structure consists of linear *β*-(1,4)-D-mannan chains with single D-galactose substituents linked to the main backbone by *α*-(1,6)-glycosidic bonds, in a ratio of 4:1 (Wielinga 2000). LBG is only slightly soluble in cold water, and must be heated to at least 80 °C for full hydration (Pegg 2012). Solutions tend to be a little cloudy due to the presence of small quantities of protein and fibre.

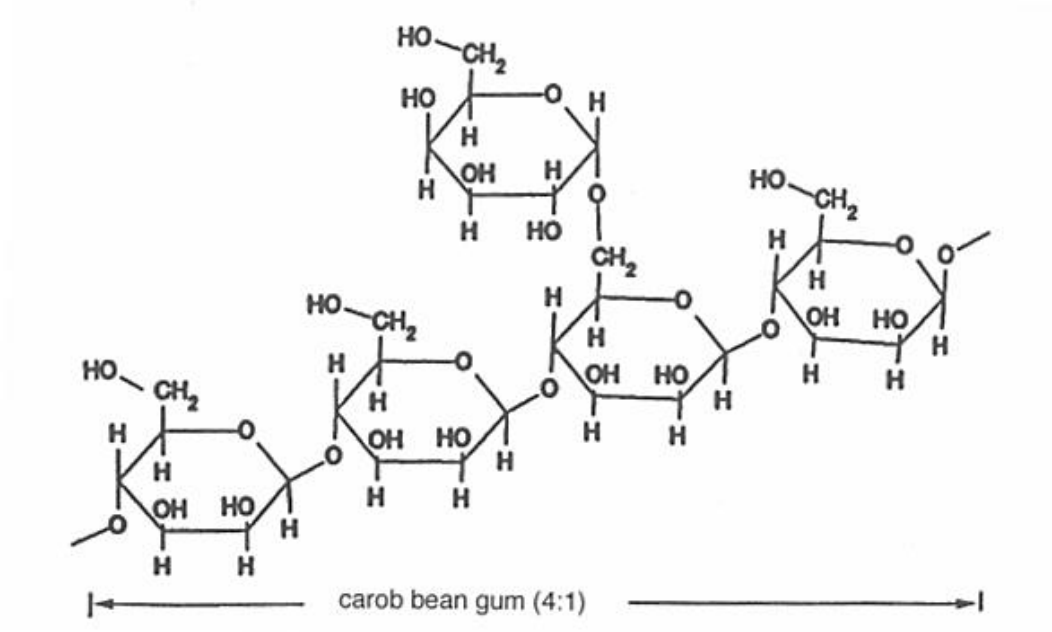

Figure 3.2. Schematic representation of the molecular structure of LBG (Wielinga 2000).

Although able to form a gel on its own (Richardson and Norton 1998), it is more commonly used to form a gel in combination with other hydrocolloids such as *κ*C, agar or xanthan (Pegg 2012). Incorporation of LBG strengthens *κ*C's continuous network, promotes elastic properties and reduces syneresis (Arnaud, Choplin and Lacrox 1989; Dunstan *et al.* 2001). This synergistic interaction is attributed to the ability of LBG to form stable cross-links with the rigid *κ*C structure, increasing the elasticity of the gels (Stading and Hermansson 1993; Imeson 2000).

3.1.4. Alginates

Alginate is extracted from natural brown seaweeds or algae (*Phaeophycea*), and forms heat-stable gels in the presence of acids and calcium. Sodium alginate (NaA) is an unbranched linear polysaccharide consisting of β-(1,4)-D-mannuronic acid (M-block) and α-(1,4)-L-glucuronic acid (G-block) residues, see **Figure 3.3**, and varies widely in composition and sequence (Draget 2000; Yoo *et al.* 2006).

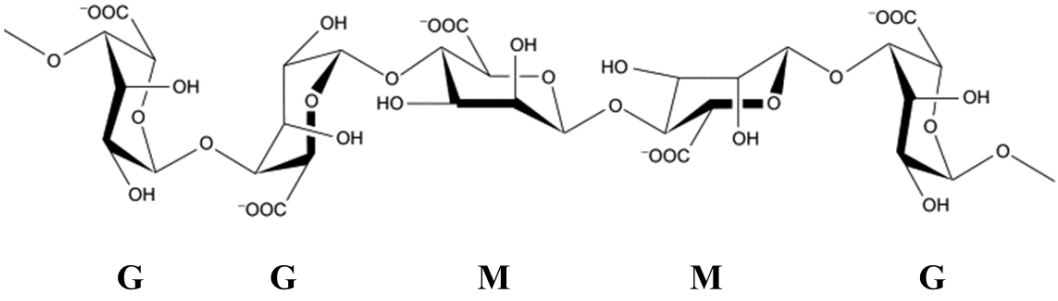

Figure 3.3. Schematic representation of the molecular structure of alginate, as adapted from Draget (2000) and Kariduraganavar, Kittur and Kamble (2014).

The gelling characteristics of NaA are based on their ion binding properties, where the alginate undergoes ionic crosslinking in presence of Ca^{2+} ions to form a so-called "egg-box model" gel structure, as shown in **Figure 3.4** (Yoo *et al.* 2006; Draget 2000). The divalent calcium displaces the sodium ions (Na^{+}), and due to the physical crosslinking or chelation between the carboxylate anions of guluronate-units in NaA and the Ca^{2+} ions, calcium alginate gels (CaA) are formed.

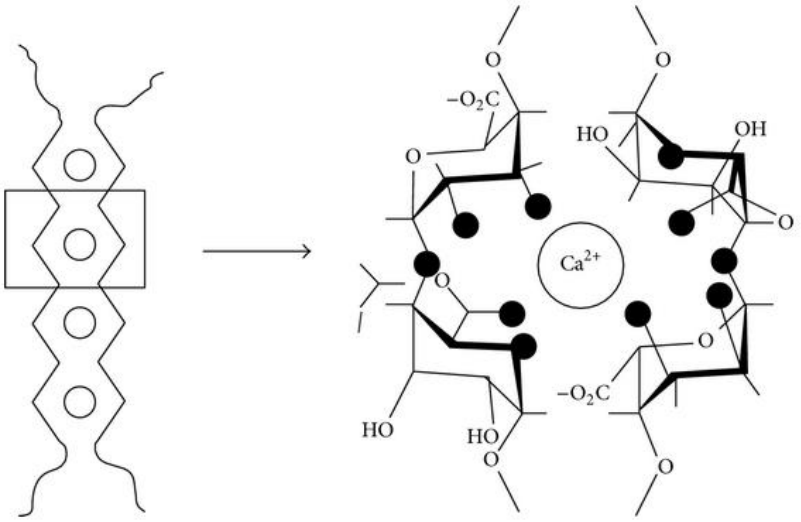

Figure 3.4. Schematic illustration of the "egg-box model" gel structure for alginates in presence of calcium ions (Braccini and Pérez 2001).

To achieve structural complexity, *κ*C gels mixed with NaA or with the inclusion of CaA beads differing in particle size can be created. CaA beads can be created via extrusion techniques. By dropping a liquid NaA solution into a ionic water bath, a gel capsule is formed around the liquid NaA droplets (Torres, Murray and Sarkar 2017). The Buchi Encapsulator® was developed based on this technique (see **Figure 3.5**), in which the NaA solution is pumped through the encapsulator system, passing it through a vibrating nozzle before dropping it into a turbulently stirred Ca^{2+} solution.

Figure 3.5. Buchi Encapsulator® set-up (left) and dropping mechanism of the NaA solution into a water bath with Ca^{2+} ions (right).

3.2. Materials and methods

3.2.1. Materials

Food grade quality kappa-carrageenan, locust bean gum and sodium alginate were purchased from Special Ingredients Ltd (Chesterfield, UK). Potassium chloride was purchased from Minerals Water Ltd (Purfleet, UK) and calcium chloride from VWR International (Leuven, Belgium). Gels were prepared with demineralised water and all materials were used without further purification.

3.2.2. Preparation of the hydrogels

A wide range of different single and mixed hydrogels were prepared with κC, LBG and NaA/CaA. Typically 400 g of sample was prepared per batch and poured into petri-dishes to a height of 1 cm. These petri-dishes were kept overnight at 4 °C to allow the gels to set, and cylindrical pieces of the hydrogels were cut out from the petri-dishes using a circular cookie cutter (diameter 24 mm, height 10 mm). Visual images of the hydrogels are shown in **Supplementary Figures A.1 to A.3**.

The single matrix κC hydrogels were prepared by dispersing the desired amounts of κC in a 0.2 M KCl solution and letting them stir for 30 min to ensure maximum hydration. Then, the solutions were heated up in a shaking water bath at 98 °C for 1 h under constant mixing until completely dissolved. In the case of mixed hydrogels, LBG or NaA were mixed with the κC first, before continuing the gel preparation. CaA beads of various sizes (nozzle size 300, 450, 750 and 100 μm) were prepared separately, and then added as a separate layer in the petri-dish before adding the hot κC mixture. A 1 wt% NaA solution was passed through the Buchi Encapsulator B-390® (Buchi UK Ltd, Chadderton, UK) at 250 mbar with a vibrating nozzle at 500 Hz, and then dropped into stirring 0.05 M $CaCl_2$ water bath. The aqueous Ca^{2+} solution was stirred for 20 min, after which the CaA beads were filtered and washed three times using demi water to remove residual Ca^{2+} ions.

3.2.3. Texture analysis

The textural properties of the different hydrogels consisting of a κC matrix were measured with a puncture test in a Texture Analyzer (TA-TX2, Stable Micro Systems Ltd., Surrey, UK), attached with a 30 kg load cell. Samples were compressed using a 10 mm Volodkevitch bite jaw probe, at a constant speed of 2 mm/sec with a deformation of 80 % strain. All tests were carried out at 22 °C, and at least three replicates were recorded for each hydrogel. The software Exponent (TEE32, v6.1.9.0, Stable Micro Systems Ltd., Surrey, UK) was used to obtain the force-distance curves, and the fracture mechanics were calculated from these curves. Analysis of variance (one-way ANOVA) with Bonferroni *post-hoc* testing was performed using SPSS (IBM® SPSS® Statistics, v24, SPSS Inc, Chicago, USA) to study the statistically relevant differences between hydrogel samples and significance level was set at $p < 0.05$.

3.3. Results

3.3.1. Fracture properties of *κ*C hydrogels

First of all, a wide concentration range of single matrix *κ*C hydrogels was prepared, increasing the concentrations stepwise by 0.5 wt%. The lowest *κ*C concentration that formed a gel firm enough to measure with the texture analyser was 0.5 wt%, but it was not very stable. The highest concentration that we were able to dissolve was 5.0 wt%. The samples formed translucent gels, and visibly increased in opaqueness with increasing concentration (see **Supplementary Figure A.1**).

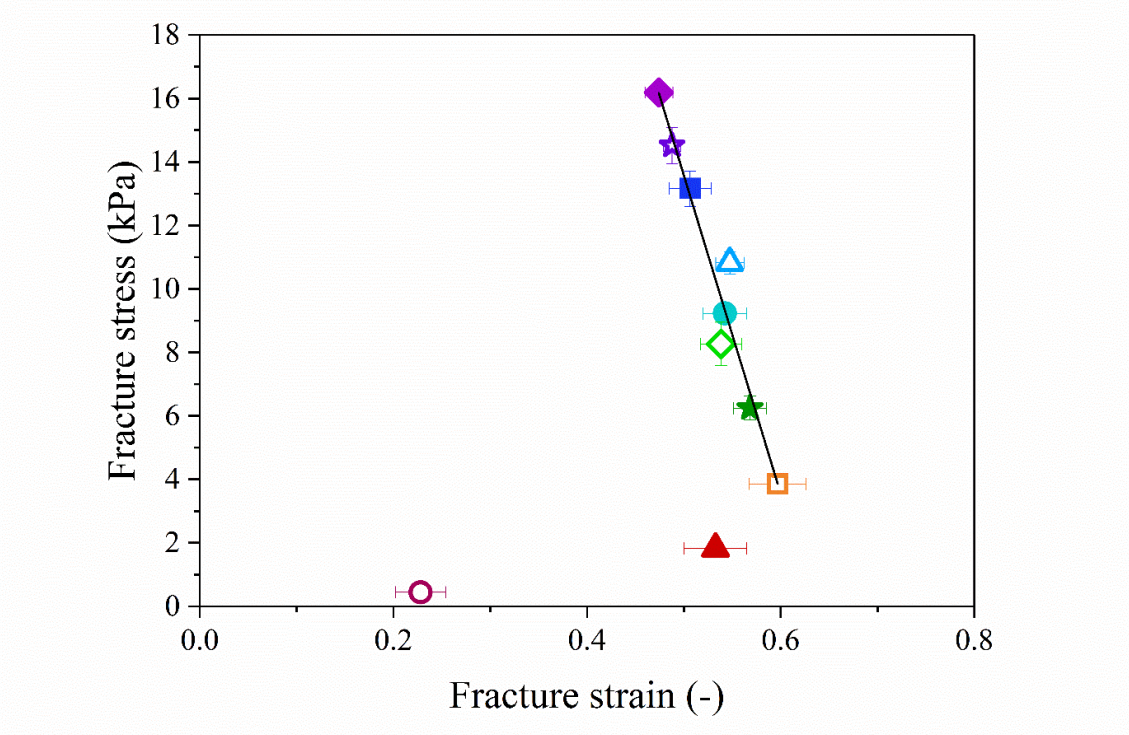

Figure 3.6. Mean (± SD) fracture stress and fracture strain of puncture tests with Volodkevitch probe of *κ*C hydrogels, with concentrations 0.5*κ*C (○), 1*κ*C (▲), 1.5*κ*C (□), 2*κ*C (★), 2.5*κ*C (◊), 3*κ*C (●), 3.5*κ*C (△), 4*κ*C (■), 4.5*κ*C (☆) and 5*κ*C (♦). The trendline is shown in black for the *κ*C concentrations 1.5 – 5.0 wt%: y = -99.5x + 63.3 and R^2 = 0.95. The fracture stress differed significantly between hydrogels ($p < 0.05$).

Figure 3.6 shows the fracture stress and fracture strain of these gels, and a clear trendline could be observed from the increasing concentrations, starting with 1.5*κ*C. The higher the concentration, the higher the fracture stress and the lower the fracture strain. With each 0.5 wt% increase in concentration, the fracture stress increased by about 1-2 kPa. This strongly indicates the gel strength increases with concentration, but decreases in elasticity.

3.3.2. Fracture properties of *κ*C/LBG hydrogels

Subsequently, different mixed hydrogels with a *κ*C matrix were evaluated. The fracture stress and fracture strain of mixed *κ*C hydrogels with LBG are shown in **Figure 3.7**. Hydrogels with 1, 2 and 3 wt% total hydrocolloid concentrations were measured in various *κ*C and LBG ratios, indicated in red, green and purple, respectively. As can be seen, the fracture stress increases with increasing total hydrocolloid concentration. No clear trend could be seen within the hydrogels with the same total hydrocolloid concentrations with different ratios. For the ratios with total biopolymer concentrations 2 and 3 wt%, the fracture stress and fracture strain were not significantly different from each other ($p < 0.05$), except for the fracture stress of 1.67*κ*C0.33LBG being a little higher than that of 1.5*κ*C0.5LBG and 1.8*κ*C0.2LBG. For the ratios with a total biopolymer concentration of 1 wt%, the fracture stress and fracture strain increased with decreasing *κ*C content and increasing LBG content.

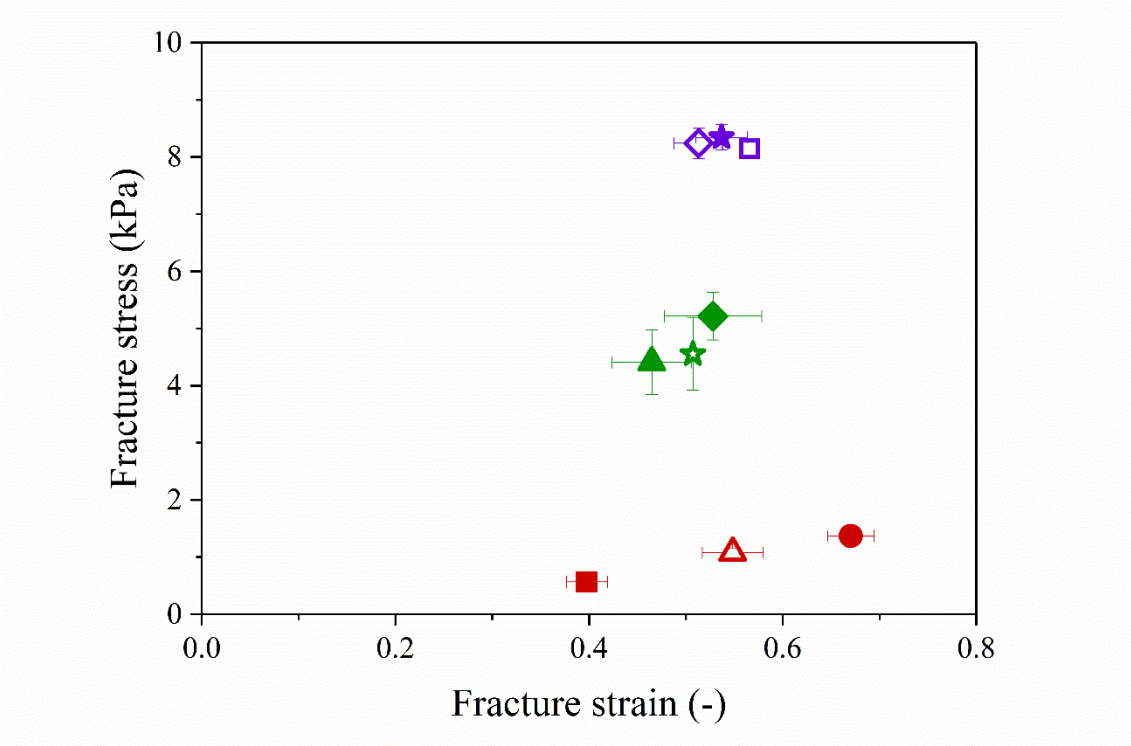

Figure 3.7. Mean (± SD) fracture stress and fracture strain of puncture tests with Volodkevitch probe of mixed *κ*C and LBG hydrogels, with concentrations 0.7*κ*C0.3LBG (●), 0.8*κ*C0.2LBG (Δ), 0.9*κ*C0.1LBG (■), 1.5*κ*C0.5LBG (☆), 1.67*κ*C0.33LBG (♦), 1.8*κ*C0.2LBG (▲), 2.25*κ*C0.75LBG (□), 2.5*κ*C0.5LBG (★) and 2.7*κ*C0.3LBG (◊).

This suggests that the *κ*C network can be strengthened with LBG at lower biopolymer concentrations, but a certain limit is reached at higher concentrations where the network is no longer strengthened by the increase in LBG content. In addition, the fracture stress of the single 3*κ*C hydrogel was slightly higher than that of the mixed hydrogel 2.25*κ*C0.75LBG, with values of 9.2 kPa compared to 7.9 kPa, respectively ($p < 0.05$), but the other 3 wt% *κ*C/LBG ratios were not significantly different ($p > 0.05$). The fracture stress of 2*κ*C and 1*κ*C was consistently higher than that of all 2 wt% *κ*C/LBG or 1 wt% *κ*C/LBG ratios, respectively ($p > 0.05$). This would indicate that the addition of LBG at the tested concentrations weakened the *κ*C network slightly.

3.3.3. Fracture properties of alginate hydrogels (NaA and CaA)

The fracture stress and fracture strain of mixed *κ*C hydrogels with alginates are shown in **Figures 3.8**. Mixed *κ*C hydrogels with NaA at 2 and 3 wt% total hydrocolloid concentrations were measured in various ratios, indicated in yellow and blue, respectively. With increasing *κ*C, and thus reducing NaA concentrations, the fracture stress and fracture strain increased. Compared to *κ*C alone, *κ*C/NaA mixed hydrogels at 2 and 3 wt% total hydrocolloid concentrations were much weaker suggesting that the *κ*C network in interrupted by the NaA molecules disrupting the strong *κ*C bonds. The 3 wt% *κ*C/NaA hydrogel with the highest fracture stress (4.9 kPa) was found to be half as much as the single 3*κ*C (9.2 kPa).

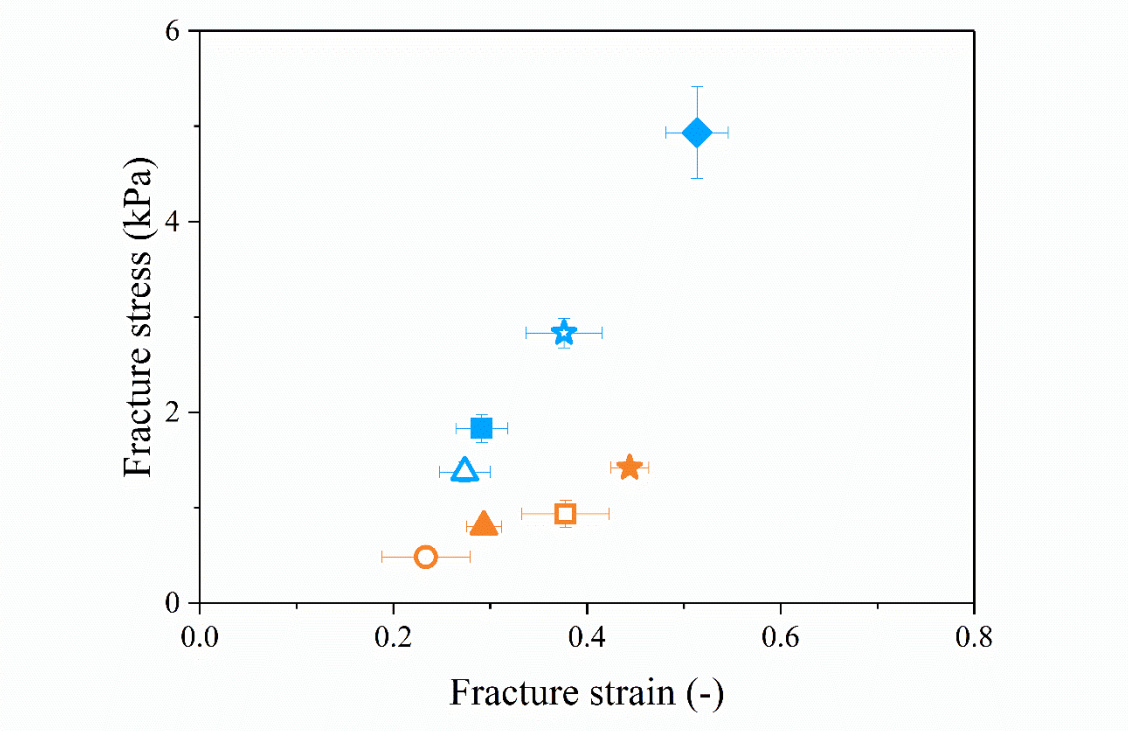

Figure 3.8. Mean (± SD) fracture stress and fracture strain curve of puncture tests with Volodkevitch probe of mixed *κ*C and NaA hydrogels, with concentrations 1*κ*C1NaA (○), 1.4*κ*C0.6NaA (▲), 1.5*κ*C0.5NaA (□), 1.8*κ*C0.2NaA (★), 1.5*κ*C1.5NaA (△), 1.8*κ*C1.2NaA (■), 2.25*κ*C0.75NaA (☆) and 2.7*κ*C0.3NaA (♦).

Measurements with CaA beads revealed the complexity of hydrogel measurements with a level of inhomogeneity. Due to the layers, two main fracture peaks were identified: one for the beads layer and another as a result of the *κ*C matrix. The fracture point here, however, was considered at the highest peak. It was found that the main determining factor for fracture stress was the *κ*C matrix, where the higher concentration of *κ*C increased the fracture stress (**Figure 3.9**). The presence of CaA beads lowered the gel strength compared to the single hydrogels with the same *κ*C concentration.

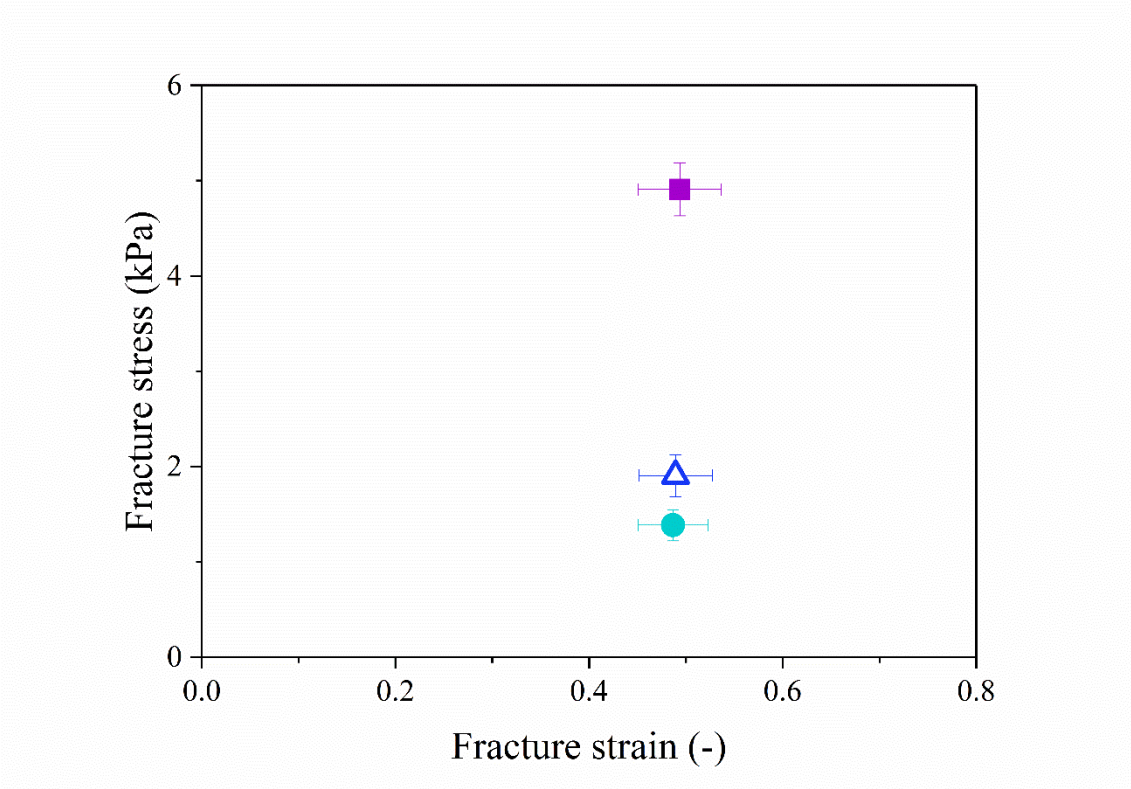

Figure 3.9. Mean (± SD) fracture stress and fracture strain curve of puncture tests with Volodkevitch probe of mixed *κ*C hydrogels of various concentrations and an added layer of CaA beads, with 1.6*κ*C0.2CaA$_{300}$ (●), 1.6*κ*C0.2CaA$_{1000}$ (Δ), 2.4*κ*C0.2CaA$_{300}$ (■).

3.4. Selection of hydrogels

To study any further differences in instrumental and oral processing properties, two single κC hydrogel concentrations were nominated, reflecting differences in fracture strain, but that were also considered to be edible. If the concentration, and thus the fracture stress, becomes too high, the hydrogel sample might require so much chewing that it will no longer considered to be palatable. Therefore, the concentrations 2κC and 3κC were selected for further study. Furthermore, a mixed gel with LBG and another with NaA were chosen. The sample 2.25κC0.75LBG was selected to study any influences of the addition of LBG, where the fracture properties were similar to the single 3κC hydrogel. The 1.5κC0.5NaA hydrogel was selected to study the influence of NaA with a much lower fracture stress, yet still have a measurable amount of oral processing. In addition, the concept of texture complexity was explored further by examining κC hydrogels with added CaA beads of various sizes. Two basic κC concentrations were chosen for the hydrogel matrix, and bead sizes of 300 and 1000 μm were selected as they might potentially have the most different lubrication experiences during oral processing. The instrumental (rheology, tribology) and sensory properties of the selected hydrogels are further explored and described in the next chapter, **Chapter 4**.

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Chapter 4

On relating rheology and oral tribology to sensory properties in hydrogels[b]

Abstract

The aim of this study was to understand the relationship between rheological, tribological and sensory properties of hydrogels differing in hydrocolloid type, concentration and degree of inhomogeneity. Fracture properties of hydrogels containing different ratios of κ-carrageenan (κC) and/or locust bean gum (LBG), sodium alginate (NaA), 300/1000 μm calcium alginate beads (CaA) at 1-4 wt% concentration were determined, and the texture perception was analysed by descriptive sensory analysis (n=11). Viscosity and friction coefficients (μ) of the hydrogel-boli after simulated oral processing were characterized. Tribology measurements were conducted in a polydimethylsiloxane ball/disc set-up with pre-adsorbed artificial salivary film at 37 °C. 'Scaling' with boli viscosity showed good agreement of observed data with the Stribeck master curve, however only in the mixed regime *i.e.* at intermediate values of the product of velocity and lubricant viscosity (Uη). Low μ values of gel boli in the boundary regime were

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largely driven by the formation of a viscous layer of bolus fragments between opposing surfaces. Fracture properties of hydrogels and boli viscosity were correlated with all chewing-related texture attributes *i.e.* 'firm', 'elastic', 'chewy' and 'cohesive' and inversely correlated with lubrication-related attributes 'melting' and 'pasty' ($p<0.05$). On the other hand, μ of the bolus filtrate at orally relevant speeds (50 mm/s) was inversely correlated with lubrication-related attributes 'pasty' and positively with 'slippery' ($p<0.05$). The lack of correlations with 'smooth' could be explained due to sample inhomogeneity and the absence of 'ball-bearing'-ability of the gel beads. A combination of initial fracture properties, boli viscosity and tribology of bolus filtrates (mixed regime) impacted the lubrication-related attribute 'salivating' ($p<0.05$).

4.1. Introduction

Oral processing strategies, such as a high number of chews and a long oral residence time have recently been linked to lower self-reported hunger and food intake in controlled experiments (Krop *et al.* 2018; Miquel-Kergoat *et al.* 2015). Hence, there has been a gradual increase in research efforts to understand and alter oral processing *i.e.* in-mouth chewing and lubrication by means of microstructural engineering (Laguna *et al.* 2016; Laguna and Sarkar 2016). Understanding the characteristics of oral processing (chewing, lubrication) has drawn significant research attention with the focal point recently shifting from rheology to tribology. This is largely due to the current consensus on the transformation during oral processing from rheology (bulk property) to tribology (surface property of food-saliva bolus based lubricants) (Pradal and Stokes 2016; Stokes, Boehm and Baier 2013; Chen and Stokes 2012; Prakash, Tan and Chen

2013; Garrec and Norton 2013; van Stee, de Hoog and van de Velde 2017; Sarkar *et al.* 2017; Laguna and Sarkar 2017).

An important aspect of oral processing of solid and semi-solid foods is the incorporation of saliva to form a swallowable food bolus. Saliva is a complex biological fluid that consists of mainly water (~99.5%), various enzymes (*α*-amylase, lysozyme) and proteins, (~0.3%), small organic compounds and inorganic salts (Sarkar, Goh and Singh 2009; Sarkar and Singh 2012; Sarkar, Ye and Singh 2017). The key protein component in human saliva is highly glycosylated mucin, which mainly contributes to the lubrication and shear-thinning properties of saliva (Schipper, Silletti and Vingerhoeds 2007; Vijay *et al.* 2015). The incorporation of saliva over time within a single bite episode has a major effect on the texture perception (Funami *et al.* 2012; Hutchings and Lillford 1988). The in-mouth friction properties might change significantly due to the interactions between food and salivary components, such as mucins and salts. However, few studies have used real human saliva, or artificial saliva formulation, within *in vitro* oral processing experiments to understand its impact on the mechanical properties, such as viscosity or friction coefficient, and correlated such data to sensory perception (Morell, Chen and Fiszman 2017; Laguna *et al.* 2017a; Laguna *et al.* 2017b).

This study creates a unique body of evidence on the initial fracture properties of hydrogels, viscosity and tribology of hydrogel boli created using simulated oral processing (using artificial saliva formulation) and sensory profiling (descriptive analysis) to understand the relationship between mechanical and sensory properties. To investigate food oral processing, biopolymeric

‘hydrogels’ have been selected as model solids and semi-solid foods in the literature (Hayakawa *et al.* 2014; Kohyama *et al.* 2015; Hori *et al.* 2015; Santagiuliana *et al.* 2018; Laguna *et al.* 2016; Laguna and Sarkar 2016). This is because hydrogels have a relatively low level of complexity as compared to most composite foods systems. They can be structurally manipulated in a systematic manner, and exclude prior learning, emotional associations and expected postprandial satisfaction (if any) during sensory testing.

Recently, there has been an increase in research efforts directed towards designing hydrogels with structural complexity for various applications (Tang *et al.* 2016; Santagiuliana *et al.* 2018; Laguna and Sarkar 2016). For instance, Laguna & Sarkar (2016) demonstrated that incorporation of calcium alginate gel beads of 185-2380 μm size in *κ*-carrageenan hydrogel matrix enabled to increase the oral residence time. On the other hand, Tang *et al.* (2016) showed the impact of using textural heterogeneity with seeds as well as layering arrangements within gelatine-agar hydrogels on increasing satiation. Temporal perception of texture contrast was recently investigated by Santagiuliana *et al.* (2018), where authors employed layering approaches to generate mechanical contrast in agar, *κ*-carrageenan, and gelatine hydrogels and suggested that a combined effect of mechanical and physicochemical properties influenced the dynamic perception of inhomogeneity over time. Nevertheless, to our knowledge, creation of hydrogels with systematic manipulation of structural complexity and understanding the impact of those manipulations on ‘chewing’ and ‘lubrication’ related texture attributes that are perceived during early and later stages of oral processing, respectively, have not been investigated to date.

The aim of this study was to understand the relationship between rheological, tribological and sensory properties of hydrogels differing in hydrocolloid type, concentration and degree of inhomogeneity. Our hypothesis was that initial fracture properties of the hydrogels and apparent viscosities of the gel boli would be correlated with chewing-related texture attributes, whereas tribological properties (*i.e.* friction coefficients in boundary and mixed lubrication) of the gel boli would be correlated with lubrication-related texture attributes. A range of hydrogels using *κ*-carrageenan, locust bean gum (LBG), sodium alginate and calcium alginate with different degrees of structural complexity and inhomogeneity were employed to test this hypothesis.

Kappa-carrageenan forms a tight-knit molecular network that results in the formation of a strong homogenous gel matrix. Since real food is not homogeneous, a degree of structural complexity was achieved in the samples by manipulating *κ*-carrageenan gels using LBG or sodium alginate to form mixed gels. Incorporation of LBG can strengthen *κ*-carrageenan's continuous network, promoting elastic properties and reducing syneresis. This synergistic interaction is attributed to the ability of LBG to form stable cross-links with *κ*-carrageenan (Stading and Hermansson 1993). On the other hand, sodium alginate is known to interfere with the incipient coil-to-helix transition during the formation of the *κ*-carrageenan gel, and thus the sodium alginate + *κ*-carrageenan mixture is expected to create a weaker mixed gel (Laguna and Sarkar 2016). To add another dimension to the structural complexity, a level of inhomogeneity was introduced in the *κ*-carrageenan gels by inclusion of calcium alginate beads of different particle sizes, where the latter behaved as "inactive filler particles" (Laguna and Sarkar 2016). Presence of calcium alginate beads would likely lead to a decrease

of mechanical strength due to interruption of the continuous *κ*-carrageenan network by these beads acting as structural defects. To our knowledge, this is the first study that attempts to examine the relationship between rheology, tribology and sensory perception in hydrogels and findings from this study should provide useful information for the design of novel foods with specifically tailored oral texture and sensory properties.

4.2. Materials and methods

4.2.1. Materials

Food grade quality kappa-carrageenan, locust bean gum and sodium alginate were purchased from Special Ingredients Ltd (Chesterfield, UK). Green food colouring was obtained from AmeriColor (Placentia, USA) and American peppermint extract was purchased at a local supermarket (Leeds, UK). Potassium chloride was purchased from Minerals Water Ltd (Purfleet, UK) and calcium chloride from VWR International (Leuven, Belgium). Additionally, sodium chloride, potassium phosphate, potassium citrate, uric acid sodium salt, urea, lactic acid sodium salt, and porcine gastric Mucin Type II were obtained from Sigma-Aldrich (St. Louis, USA). All materials were used without further purification. Demineralised water was used in preparation for all the gels and the artificial saliva formulation.

4.2.2. Preparation of the hydrogels

The composition of the hydrogels is shown in **Table 4.1**. Visual images of the seven hydrogels are shown in **Supplementary Figure B.1**. Typically 400 g of sample was prepared and poured into petri-dishes (150 g gel per petri-dish), and

then kept overnight at 4 °C. Cylindrical pieces of the hydrogels were cut out from the petri-dish using a circular cookie cutter (diameter 25 mm, height 10 mm), and used as such for all measurements.

Table 4.1. Final composition of the hydrogels.

Hydrogel samples[i]	*κ*-carrageenan (wt%)	Locust bean gum (wt%)	Na-alginate (wt%)	Ca-alginate beads (wt%)	Water (wt%)
2*κ*C	2				97
3*κ*C	3				96
2.25*κ*C0.75LBG	2.25	0.75			96
1.5*κ*C0.5NaA	1.5		0.5		97
1.6*κ*C0.2CaA$_{1000}$	1.6			0.2	97
1.6*κ*C0.2CaA$_{300}$	1.6			0.2	97
2.4*κ*C0.2CaA$_{300}$	2.4			0.2	96

[i] All hydrogels contained 0.5 wt% green food colouring and 0.5 wt% peppermint flavouring. The two *κ*-carrageenan hydrogels contained 0.145 wt% KCl. The composition of the mixed hydrogels containing Ca-alginate beads was determined based on the ratio between *κ*-carrageenan gel matrix (2 or 3 wt%) and Ca-alginate beads (1 wt%), irrespective of bead size.

4.2.2.1. Kappa-carrageenan hydrogels

For preparation of kappa-carrageenan hydrogels (*κ*C), appropriate quantities of *κ*C were dispersed in a 0.2 M potassium chloride (KCl) solution and stirred for 30 min to ensure maximum hydration. Then, the solution was heated up in a shaking water bath at 98 °C for 1 h. The gelling solutions were allowed to cool down for 5 min, and finally the green colouring and peppermint flavouring were added before being allowed to set in petri-dishes.

4.2.2.2. Kappa-carrageenan/LBG or kappa-carrageenan/sodium alginate hydrogels

The mixed hydrogels were prepared by mixing the appropriate quantities of powdered κC and LBG or sodium alginate (NaA) together before adding the respective powder mixtures to distilled water and mixing for 30 min. Then, the solutions were heated up in a shaking water bath at 98 °C for 1 h. The solutions were allowed to cool down for 5 min, and finally the green colouring and peppermint flavouring were added before being allowed to set in petri-dishes.

4.2.2.3. Kappa-carrageenan/calcium alginate hydrogels

The calcium alginate (CaA) beads were prepared first and then added as a layer in the κC hydrogels (before the gels were allowed to set) to create a level of inhomogeneity within the gels, based on a previous study (Laguna and Sarkar 2016). The beads were prepared by making a 1 wt% NaA solution in water, and stirring for 1 h to ensure complete hydration. Calcium chloride ($CaCl_2$) solutions of 0.01 M and 0.05 M were prepared to make the 300 μm and 1000 μm sized beads, respectively. The 1 wt% NaA solution was passed through a Buchi Encapsulator B-390® (Buchi UK Ltd, Chadderton, UK) with a vibrating nozzle and then dropped into the appropriate $CaCl_2$ solutions while being stirred to create the CaA beads. A vibrating nozzle of 300 μm (frequency 500 Hz, air pressure 250 mbar) or 1000 μm (frequency 700 Hz, air pressure 300 mbar) was used depending on the required bead size. The beads were allowed to set in the $CaCl_2$ solution at room temperature for 30 min under constant stirring. The beads were subsequently washed thrice with distilled water and then air-dried. Meanwhile, the κC solution was prepared by dissolving the appropriate amount in

distilled water and mixing for 30 min. Then, the solution was heated up in a shaking water bath at 98 °C for 1 h and allowed to cool down for 5 min followed by adding the green colouring and peppermint flavouring. The appropriate amount of CaA beads was weighed and added to the petri-dish and the *κ*C gels solution was poured, before allowing the gel to set similar to the preparation method of the aforementioned hydrogels.

4.2.3. Texture analysis

Uniaxial single compression tests were carried out using a TA-TX2 Texture Analyser (Stable Micro Systems Ltd., Surrey, UK), attached with a 50 kg load cell. In the compression test, the samples were compressed using a cylindrical probe (diameter 59 mm). The tests were carried out at 22 °C, at a constant speed of 2 mm/s and the deformation level was set at 80 % strain. At least three repeats were recorded for each gel on at least four different gel preparation days. The software Exponent (TEE32, v6.1.9.0, Stable Micro Systems Ltd., Surrey, UK) was used to obtain the force-distance curves, and the fracture mechanics were calculated from these curves. The fracture properties were determined at the maximum point of the stress-strain curves. The fracture energy was determined as the area under the curve up to the fracture point (Peleg 1984). The initial slope of all samples was determined up to a stress of 500 Pa, as this was considered within the viscoelastic limit.

4.2.4. Preparation of artificial saliva

Artificial saliva was prepared according to the method previously described by Sarkar, Goh and Singh (2009). Briefly, artificial saliva was composed of 1.59 g/L NaCl, 0.64 g/L K_2HPO_4, 0.2 g/L KCl, 0.31 g/L $K_3C_6H_5O_7.H_2O$, 0.02 g/L

$C_5H_3N_4O_3Na$, 0.2 g/L H_2NCONH_2, 0.15 g/L $C_3H_5O_3Na$ and 3 g/L mucin. The pH of the saliva solution was adjusted to pH 6.8 using 1 M NaOH. Noteworthy, porcine mucin was used in the artificial saliva to simulate the human salivary viscosity at comparable concentrations present in human saliva. However, bovine submaxillary mucin could be a promising alternative considering its ability to form more elastic films and its higher lubricating properties particularly in elastomeric contact surfaces (Madsen *et al.* 2016).

4.2.5. Simulated oral processing

The hydrogels were broken down mechanically in the presence of artificial saliva to mimic oral processing. The samples were put into a mechanical blender (Andrew James UK Ltd, Bowburn, UK) with artificial saliva in a ratio 2:1 w/w and homogenized for 15 seconds at low speed (speed 1). Depending on the hydrogel tested, the obtained particle size was < 2-5 mm. After grinding, the gel was mixed with artificial saliva (final sample to saliva ratio 4:3 w/w) and left to rest for 30 min. It is worth highlighting that the amount of saliva incorporated in the food bolus has varied across studies from as low as 8 wt% saliva *in vivo* in emulsion gels (Devezeaux de Lavergne *et al.* 2015b) to 18 wt% artificial saliva incorporation *in vitro* to create model hydrogel boli (Ishihara *et al.* 2011) to 50 wt% simulated saliva addition for food matrices in case of harmonized INFOGEST static model (Minekus *et al.* 2014). For our study, we used a ratio of 4:3 (w/w) sample:saliva to have the same level of saliva incorporation across all samples to enable comparison, though we highlight the limitation that during oral processing (*in vivo*), the amount of saliva added to the samples would not be the same across the different hydrogels with varying degrees of complexity.

The broken down hydrogel:saliva mixture samples, from here on defined as ‘gel bolus fragments’, were used for the rheological and tribological measurements. To understand the thin-film properties, the tribological properties were also measured for the samples where any large gel particles (> 500 μm) were filtered out, from here on defined as ‘gel bolus filtrate’.

4.2.6. Apparent viscosity

The apparent viscosities of the gel fragments in presence of artificial saliva were measured using a rheometer (Kinexus Ultra+, Malvern Instruments Ltd, Worcestershire, UK) equipped with a plate-plate geometry (diameter 60 mm). The gap size (ranging from 0.01-0.15 mm) was individually adjusted for each gel, depending on their particle size once broken down. To prevent evaporation, the samples were sealed off with a thin layer of silicone oil. Flow curves were obtained for all gel samples after simulated oral processing at shear rates ranging from 0.0001 to 100 s^{-1} at 37 °C. A minimum of three measurements were performed for each sample. Associated Ostwald de Waele power law (equation 4.1) was fitted to the viscosity of each sample:

$$\eta = K\gamma^{n-1} \tag{4.1}$$

where η is the apparent viscosity, K is the consistency index (Pa s) and n is the behaviour index. These parameters were utilised in the determination and validation of the corresponding viscosities calculated by entrainment speeds and permitted friction coefficients to be plotted against the entrainment speed and viscosity products as described in the tribology section.

It is noteworthy that detailed rheological characterization of the viscoelasticity of the hydrogels and the corresponding bolus fragments was not carried out in this study.

4.2.7. Oral tribology

The oral tribological properties of the gel bolus fragments and gel bolus filtrates were determined using a ball-on-disc set up in a Mini Traction Machine (MTM2, PCS Instruments, London, UK). The gel bolus samples were prepared according to the method described above. Commercially available polydimethylsiloxane (PDMS) ball (diameter of 19 mm, MTM ball Slygard 184, 50 Duro, PCS Instruments, London, UK) and disc (diameter of 46 mm, thickness of 4 mm, MTM disc Slygard 184, 50 Duro, PCS Instruments, London, UK) were used for the measurements (surface roughness of PDMS tribopairs, R_a < 50 nm). The PDMS surface contacts were kept a minimum of 2 h submerged in artificial saliva to create a mucin film with the intent to simulate the oral conditions. The sample was loaded into the pot equipped with the PDMS disc; the ball was lowered onto the disc and then the pot was covered with a lid. The PDMS ball and disc were rotated at different speeds to create a relative motion between the surface of the ball and the disc, resulting in a slide-to-roll ratio (SRR) of 50%, to impart both rolling and sliding motions (Sarkar *et al.* 2017) and the temperature was maintained at 37 °C, simulating oral conditions.

Two parameters have been used for both the ball speed and the disc speed: one with $V_{ball} > V_{disc}$ and one with $V_{ball} < V_{disc}$, while keeping the SRR constant. The entrainment speed was calculated as the average of the two measures to remove any offset errors in the lateral force measurement, as well to remove any

friction that did not reverse sign when the speeds were reversed, such as rolling friction (Bongaerts, Fourtouni and Stokes 2007). Thus, the entrainment speed was defined as:

$$\bar{U} = \frac{1}{2}(U_1 + U_2) \tag{4.2}$$

where U is the entrainment speed and U_1 and U_2 are the velocities of the two contacting surfaces (i.e. ball and disc). The rolling speed was reduced from 1000 to 1 mm/s and friction forces were measured to obtain a Stribeck curve. All tests were performed at a load of 2 N, as this is a good representative value of loads occurring in the mouth while maintaining sensitivity in the tribometer. Average and standard deviation were calculated from three measurements on replicate samples. Following the studies by de Vicente, Stokes and Spikes (2005) and Bongaerts, Fourtouni and Stokes (2007), we utilized the Stribeck 'master curve' (equation 4.3) to enable comparison of sample friction coefficient μ against the product of entrainment velocity U and sample viscosity η:

$$\mu_{total} = \mu_{EHL} + \left(\frac{\mu_b - \mu_{EHL}}{1 + \left({}^{U\eta}/_{B}\right)^{m}}\right) \tag{4.3}$$

where

$$\mu_{EHL} = k(U\eta)^n \tag{4.4}$$

and

$$\mu_b = h(U\eta)^l \quad (4.5)$$

where, (*k*, *n*) and (*h*, *l*) are the elastohydrodynamic lubrication (EHL) and boundary layer power law coefficient and index respectively. Here, *B* relates to the threshold value of $U\eta$ for boundary friction and *m* represents the mixed regime exponent. It is worth pointing out that the flow curves (in the above section) were only determined for gel fragments to relate to the early stages of oral processing where bulk properties tend to dominate. However, friction coefficients were determined for both bolus fragments and filtrates, latter resemble the thin layer formed between the contact surfaces (*e.g.* tongue and palate) in later stages of oral processing, where surface properties dominate (Chen and Stokes 2012; Stokes, Boehm and Baier 2013; Laguna and Sarkar 2017).

4.2.8. Descriptive sensory analysis

A panel was recruited from the University of Leeds to participate in a descriptive sensory analysis. The panel was selected and familiarized with the hydrogel samples followed by generation of attributes and introduction to the rating scale. The study was reviewed and approved by the Faculty Research Ethics Committee at the University of Leeds (ethics reference MEEC 16-006). A group of 11 participants (4 male, mean (± SD) age = 28.8 (± 5.5) years, range 21-40 years) was trained to familiarize them with the different hydrogel samples and to create a list of relevant attributes related to the chewing as well as the lubrication aspects of the gels.

Three training sessions of 1 h each were conducted with the seven hydrogel samples. During the first training session, the hydrogels were tasted to familiarise the participants with the type of samples, and participants were encouraged to come up with terms to describe the different texture aspects of the gels. Subsequently, an extensive list of potential attributes related to both the chewing and lubrication aspects was introduced to the participants and their applicability and definitions were discussed in the group. During the second session, the list of attributes generated during the first training session was further specified to describe the difference between the textural aspects of the gels as best as possible and to reach a consensus within the panel. Finally, in the last training session the rating scales were introduced and a group discussion arrived at consensus on how to use the scales for the different attributes in the included samples and the best order in which to rate these attributes.

After completion of the training, the samples were evaluated in individual sensory test booths under normal lighting conditions, with the samples presented in randomised order in triplicate in a balanced block design divided over two test days (with 11 samples rated on day one and 10 samples on day two). All samples were prepared 24 h prior to the sensory assessment, presented in individual cups labelled with a three-digit code, and moved to room temperature 20 min before the start of the test. A practice sample was provided on each test day to get a sense of the samples before the start of the test due to the novelty of these hydrogels. As determined by the training sessions, nine different texture attributes were rated for each sample in a fixed order (see **Table 4.2**).

Table 4.2. List of attributes and descriptions as included in the sensory analysis.

Textural attributes from sensory profiling	Definition
Smooth	Degree of abrasiveness of the products surface as perceived by the tongue
Firm	The force needed to compress the sample between tongue and palate (hardness)
Elastic	The ease in which the sample bounces back after chewing (springiness)/force with which the sample returns to its original shape after partial compression (without fracture) between the tongue and palate
Chewy	The amount of chews needed to break down the sample to be ready for swallowing
Cohesiveness	Degree to which the samples deforms/holds together rather than crumbles/breaks/ruptures (it conforms to the palate rather than shears)
Pasty	The sensation of the presence of wet/soft (immiscible) solids in the mouth (muddy)
Slippery	The ease in which the sample slides through the mouth during chewing (slimy)
Salivating	The amount of saliva released during chewing
Melting	The amount of sample that dissolves/disappears over time (loss of structure in the mouth) rather than cracking or breaking apart

The intensities of the different attributes were rated on an unstructured line scale of 100 mm, as presented with the software CompuSense (v5.0, Ontario, Canada), anchored from ‘not at all’ (0) to ‘very’ (100). All panellists followed the same tasting procedure, putting the whole sample in the mouth. It was optional for the panellists to choose whether to swallow the sample at the end or spit it out in provided cups. Between each sample, panellists were instructed to rinse their mouth with water and eat a cracker to cleanse their palate. Data was extracted

from the software and exported to SPSS (IBM® SPSS® Statistics, v24, SPSS Inc, Chicago, USA) for analysis.

4.2.9. Statistical analysis

Mean values and standard deviations (SD) were calculated using Excel (Microsoft Office 2010). For each sample, sensory attribute and assessor in the sensory analysis, the panel performance was checked to make sure there were no clear outliers or obvious errors using the software PanelCheck (v1.4.2). The panel agreement, discrimination and repeatability among assessors were assessed using Tucker-1 plots, F-plots and MSE plots, respectively. Panel agreement was considered high when all assessors were clustered around the same point on the outer ellipse of the Tucker-1 plot. The F-plots showed the discriminative ability for each panellist, with the higher values signifying better sample discrimination, and the MSE plot shows the mean square error values for each panellist as a measure of repeatability. The lower the repeatability of an assessor, the higher the MSE values (Tomic *et al.* 2010). The overall panel performance was considered to be acceptable, and no data was removed.

In addition, Principal Component Analysis (PCA) was conducted on the nine sensory attributes with orthogonal rotation (Direct Oblimin). The Kaiser-Meyer-Olkin measure verified the sampling adequacy for the analysis: KMO = 0.788, which is well above the acceptable limit of 0.5 (Field 2017). Bartlett's test of sphericity χ^2 (36) = 1197.985, $p < .001$, indicated that correlations between items were sufficiently large for PCA. An initial analysis was run to obtain eigenvalues for each component in the data. Two components had Eigenvalues

over Kaiser's criterion of 1 and in combination explained 64.1 % of the variance, and thus were retained in the final analysis.

In order to study the differences in samples for all selected attributes, analysis of variance (one-way ANOVA) was applied to the ratings data from the sensory panel with the samples as fixed factor; least significant differences were calculated by Bonferroni's *post-hoc* test. Similarly, differences between samples for the mechanical analyses (uniaxial compression test of hydrogels, flow curves of bolus fragments, friction coefficients of gel bolus fragments and filtrates) were determined with one-way ANOVA and Bonferroni *post-hoc* testing. Pearson's product moment correlations were calculated to assess the simple relationships between the different instrumental and sensory characteristics of the hydrogels. All statistical analyses were performed in SPSS (IBM® SPSS® Statistics, v24, SPSS Inc, Chicago, USA), and statistical significance level was set at $p < 0.05$.

4.3. Results and discussion

4.3.1. Mechanical characterisation of hydrogels and simulated boli

4.3.1.1. Texture analysis of the hydrogels

The fracture stress and strain of the seven hydrogels are shown in **Figure 4.1**. The samples can be categorized into three groups: 1) high fracture stress/high fracture strain, 2) intermediate fracture stress/fracture strain, and 3) low fracture stress/low fracture strain. Group 1 included the two *κ*C samples (2 wt% and 3 wt%) and the *κ*C/LBG sample, averaging a fracture stress of 190 kPa and a fracture strain of 1.17. Group 2 included the samples containing the CaA beads, with an average fracture stress and fracture strain of 71 kPa and 0.93, respectively, where the

particle size of CaA beads (300 or 1000 µm) did not show any significant contribution to the fracture mechanics at equivalent biopolymer concentration ($p > 0.05$). Group 3 consisted of the *κ*C/NaA hydrogels with an average fracture stress of 27 kPa and a fracture strain of 0.70. The high and low fracture stress samples varied by a factor 7 and the samples in the low and high fracture strain groups varied by a factor 1.8. The fracture energy of the hydrogels, shown in **Table 4.3**, also indicate that the samples were categorized in similar groups as in **Figure 4.1**. Based on these groupings, **Figure 4.2** shows a schematic representation of the structures of these hydrogels.

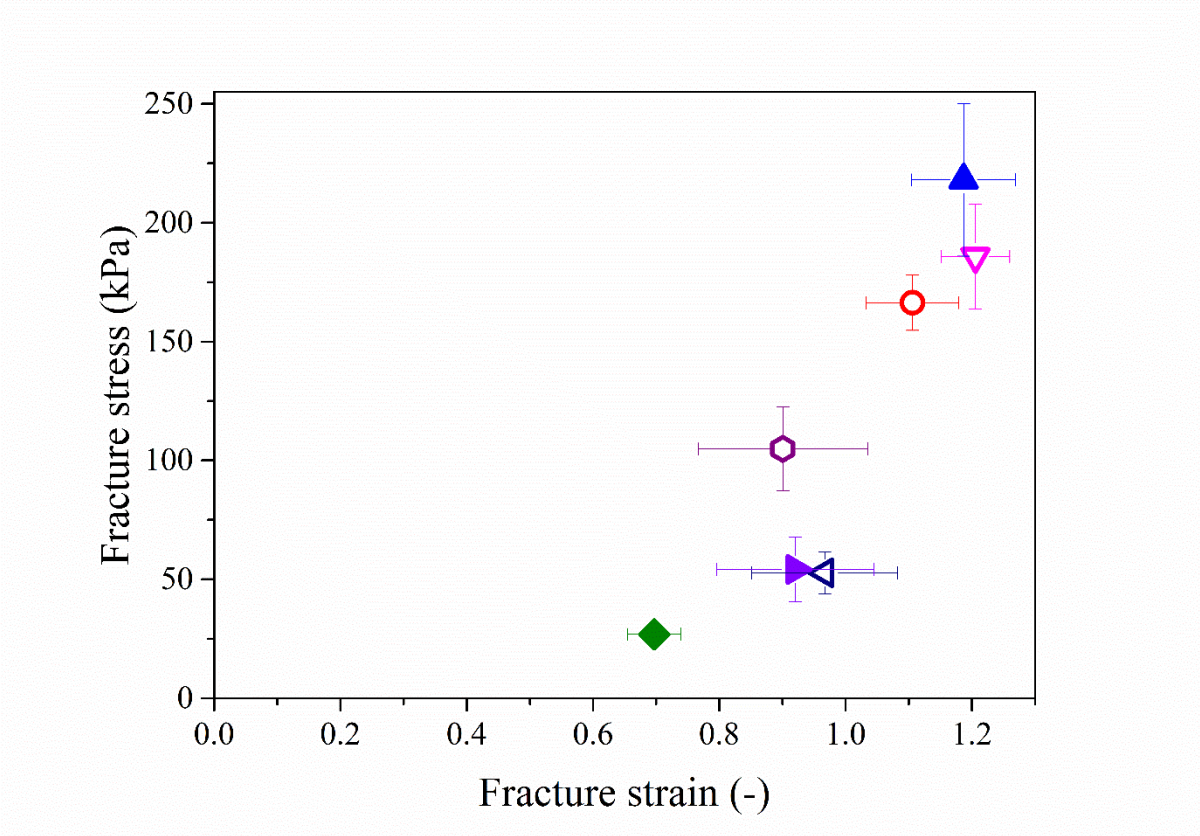

Figure 4.1. Fracture stress and strain of 2*κ*C (○), 3*κ*C (▲), 2.25*κ*C0.75LBG (▽), 1.5*κ*C0.5NaA (◆), 1.6*κ*C0.2CaA$_{1000}$ (◁), 1.6*κ*C0.2CaA$_{300}$ (▶) and 2.4*κ*C0.2CaA$_{300}$ (⬡) hydrogels in uniaxial compression test. Data points represent the average of at least three measurements on four different preparation days. Error bars indicate the standard deviation.

Table 4.3. Textural properties of the hydrogels obtained from uniaxial compression test. A different lower case letter denotes a statistically significant difference ($p < 0.05$).

Samples	Fracture energy (kPa)	
	Mean	SD
2κC	91.52^{b}	8.31
3κC	147.87^{a}	17.75
2.25κC0.75LBG	71.13^{c}	43.74
1.5κC0.5NaA	6.54^{f}	0.98
1.6κC0.2CaA$_{1000}$	26.60^{e}	5.11
1.6κC0.2CaA$_{300}$	22.39^{ef}	7.03
2.4κC0.2CaA$_{300}$	47.27^{d}	12.85

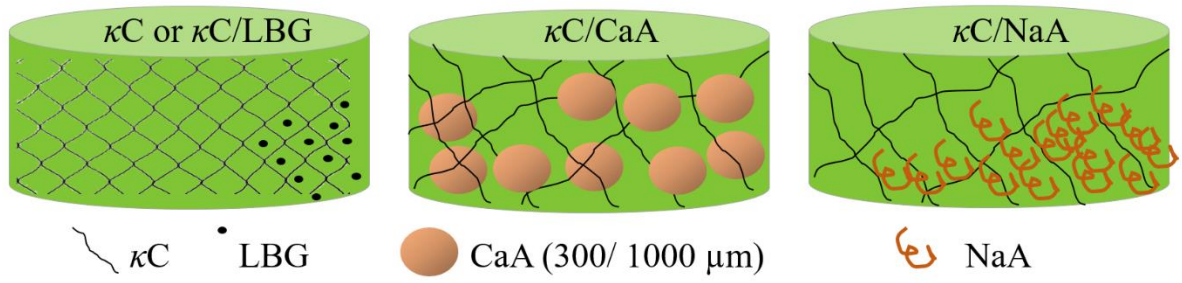

Figure 4.2. Schematic representation of the hydrogels.

As expected, the fracture stress followed a power law increase with increased concentration of κC in native κC hydrogels (**Figure 4.1**), allowing the formation of a three-dimensional network structure (as shown schematically in **Figure 4.2**) induced by the supramolecular aggregation of the double helices (Laguna and Sarkar 2016). Interestingly, the fracture stress of the 2.25κC0.75LBG hydrogel was significantly lower than that of 3κC hydrogel at equivalent total biopolymer concentration ($p < 0.05$). This is not in line with previous findings, where it has been reported that LBG has the ability to

strengthen the κC network by forming multiple junction zones between LBG un-substituted mannan backbones and κC helices (Devezeaux de Lavergne *et al.* 2016; Dea and Morrison 1975; Dunstan *et al.* 2001). A possible explanation for this could be the difference in total biopolymer concentrations and the ratio between κC and LBG used in this study versus previous reports. Interestingly, Czaczyk, Olejnik and Trojanowska (1999) also observed similar weakening effect of LBG on κC hydrogels at 2-3 wt% total biopolymer concentration in a ratio of κC: LBG of 2:1 w/w *i.e.* similar to the range used in this study.

Unsurprisingly, the presence of NaA (1.5κC0.5NaA hydrogel) resulted in significant weakening of the κC gel (**Figure 4.1**), which might be attributed to the segregative interaction between NaA and κC, disrupting the coil-to-helix transition during κC hydrogel formation (**Figure 4.2**), finally leading to a phase separated κC/NaA hydrogel (Goh, Sarkar and Singh 2008; Goh, Sarkar and Singh 2014; Laguna and Sarkar 2016). On the other hand, the presence of CaA beads (1.6κC0.2CaA_{300}, 1.6κC0.2CaA_{1000}) contributed to considerable reinforcement of the κC hydrogel as compared to that of the presence of NaA (1.5κC0.5NaA). Introducing defects due to the presence of these CaA beads as "inactive filler particles", resulted in a less defined network (**Figure 4.2**) with less fracture stress as compared to that of a native κC hydrogel (**Figure 4.1**). Based on the texture analysis results, it can be concluded that the chosen hydrogel types covered a wide range of deformation behaviour, which can be hypothesized to have different sensory properties, particularly in terms of chewing-related attributes.

4.3.1.2. Apparent viscosity of the hydrogel boli

Figure 4.3 shows the apparent viscosity (η) of the bolus particles derived from simulated oral processing of the hydrogels in the presence of artificial saliva at 37 °C. All bolus fragments in presence of artificial saliva showed extreme shear thinning behaviour, with slight indications of plateau values being reached only at low shear rate limits (10^{-3} s^{-1}). Such pseudoplastic behaviour is in agreement with that of protein-based microgels, where latter showed similar ranges of η values as a function of volume fraction and shear rate (Sarkar *et al.* 2017).

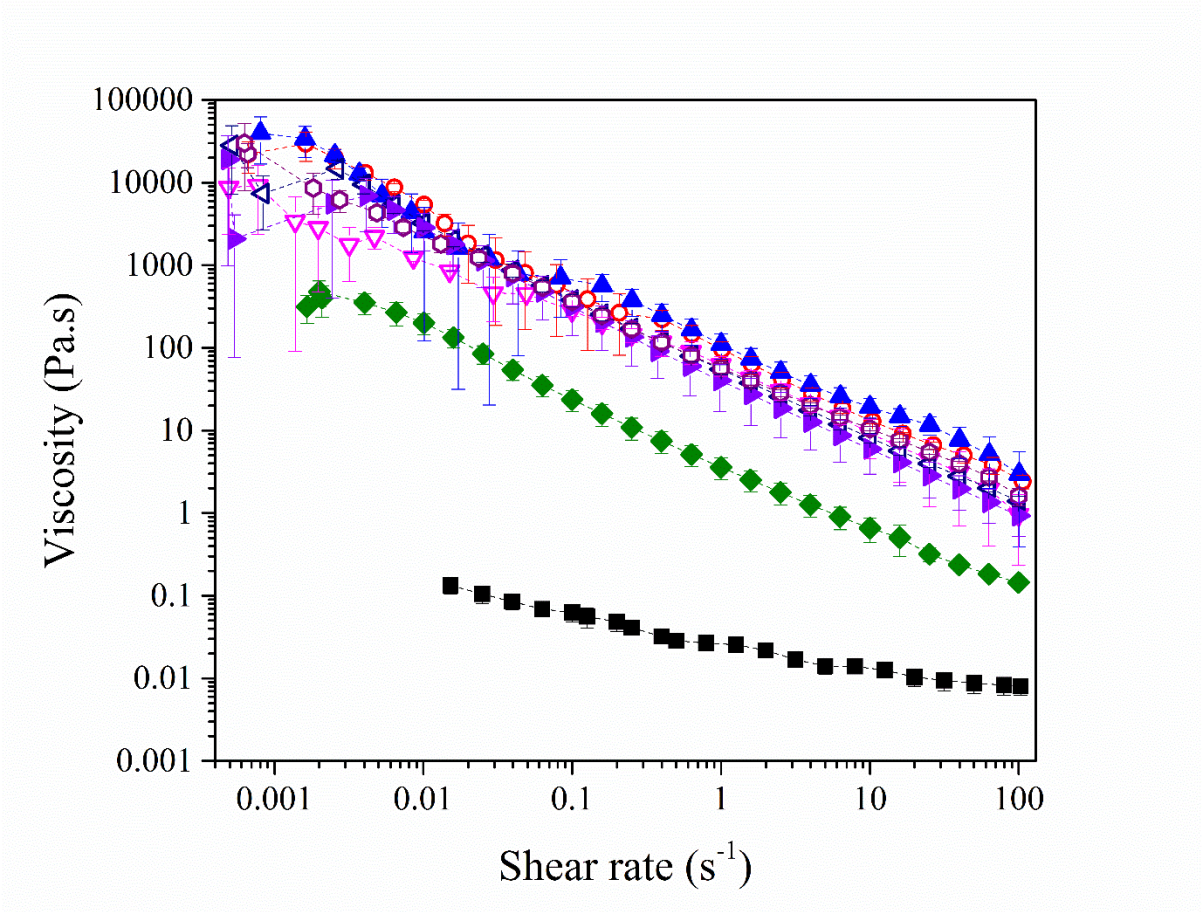

Figure 4.3. Flow curves of artificial saliva (■), 2*κ*C (○), 3*κ*C (▲), 2.25*κ*C0.75LBG (▽), 1.5*κ*C0.5NaA (◆), 1.6*κ*C0.2CaA$_{1000}$ (◁), 1.6*κ*C0.2CaA$_{300}$ (▶) and 2.4*κ*C0.2CaA$_{300}$ (⬡) gel bolus fragments as a function of shear rate at 37 °C. Data points represent the average of at least three measurements. Error bars indicate the standard deviation.

In addition, high values of η persisted in boli of both κC hydrogels and mixed hydrogels even after subjection to fairly high *i.e.* orally relevant shear of 50 s^{-1}. As expected, due to the aforementioned segregative interaction between κC and NaA, the bolus of 1.5κC0.5NaA hydrogels were one to two orders of magnitude lower in η as compared to that of the rest of the hydrogels (κC, κC/LBG and κC/CaA) even though all the systems were highly shear thinning. It is worth noting that at oral shear (50 s^{-1}), η of 1.5κC0.5NaA hydrogel bolus fragments and the rest of the (κC, κC/LBG and κC/CaA) hydrogel bolus fragments was three or four orders of magnitude higher than artificial saliva (**Figure 4.3**) or real human saliva, respectively (Bongaerts, Rossetti and Stokes 2007). This suggest that the rheology might play an important role in driving the load bearing capacity of these gel bolus fragments during oral tribology experiments and consequently sensory perception. However, the viscosity results were insufficient to identify the underlying differences in the friction coefficients (if any) between κC, κC/LBG and κC/CaA hydrogel bolus, as the viscosities were not significantly different between these gel bolus fragments at orally relevant shear rates ($p > 0.05$). Furthermore, one might investigate how the viscoelastic parameters of the bolus fragments may impact the load bearing aspects and oral processing attributes, which is beyond the scope of this study and needs to be studied in future.

4.3.1.3. Oral tribology of the hydrogel bolus fragments and filtrates

It is well recognized that the rheological properties (bulk phase) dominate the textural sensation only in the early stages of oral processing. It is now postulated that oral tribology (surface properties) dictates the thin-film properties and thus

the oral sensation in the later stages of oral processing where the food and/or food-saliva mixture interact with the oral surfaces (Stokes, Boehm and Baier 2013; Chen and Stokes 2012; Pradal and Stokes 2016; Laguna and Sarkar 2017). To understand this surface phenomenon, the coefficient of friction (μ) of both gel bolus fragments and gel bolus filtrate (*i.e.* the thin-film) when sheared between smooth hydrophobic PDMS-PDMS ball and disc tribopairs was plotted as a function of entrainment speed as shown in **Figures 4.4a and 4.4b**, respectively. Although attempts were made to pre-adsorb artificial salivary films to hydrophobic PDMS substrates, there was no change in the water contact angle (θ) of the substrates (data not shown) and the PDMS surface remained hydrophobic (θ = 108 °) as studied previously (Sarkar *et al.* 2017; Yakubov *et al.* 2009).

The plateau boundary ($\bar{U} \leq 10$ mm/s) and mixed regimes ($10 < \bar{U} \leq 300$ mm/s) of lubrication could be clearly identified in the Stribeck curves of the measured samples (**Figures 4.4a and 4.4b)**. Considering the relevance of biologically relevant speeds, such as the speed of the human tongue being ~ 20 mm/s (Steele and van Lieshout 2009), we have focussed only on boundary and mixed lubrication regimes. The artificial saliva, which served as a control, showed a classical Stribeck profile with μ varying from 0.3–0.5 in the boundary regime, falling off by one-order of magnitude in the mixed regime. This is consistent with ranges of values found in a previous study using the same artificial saliva formulation (Laguna *et al.* 2017a).

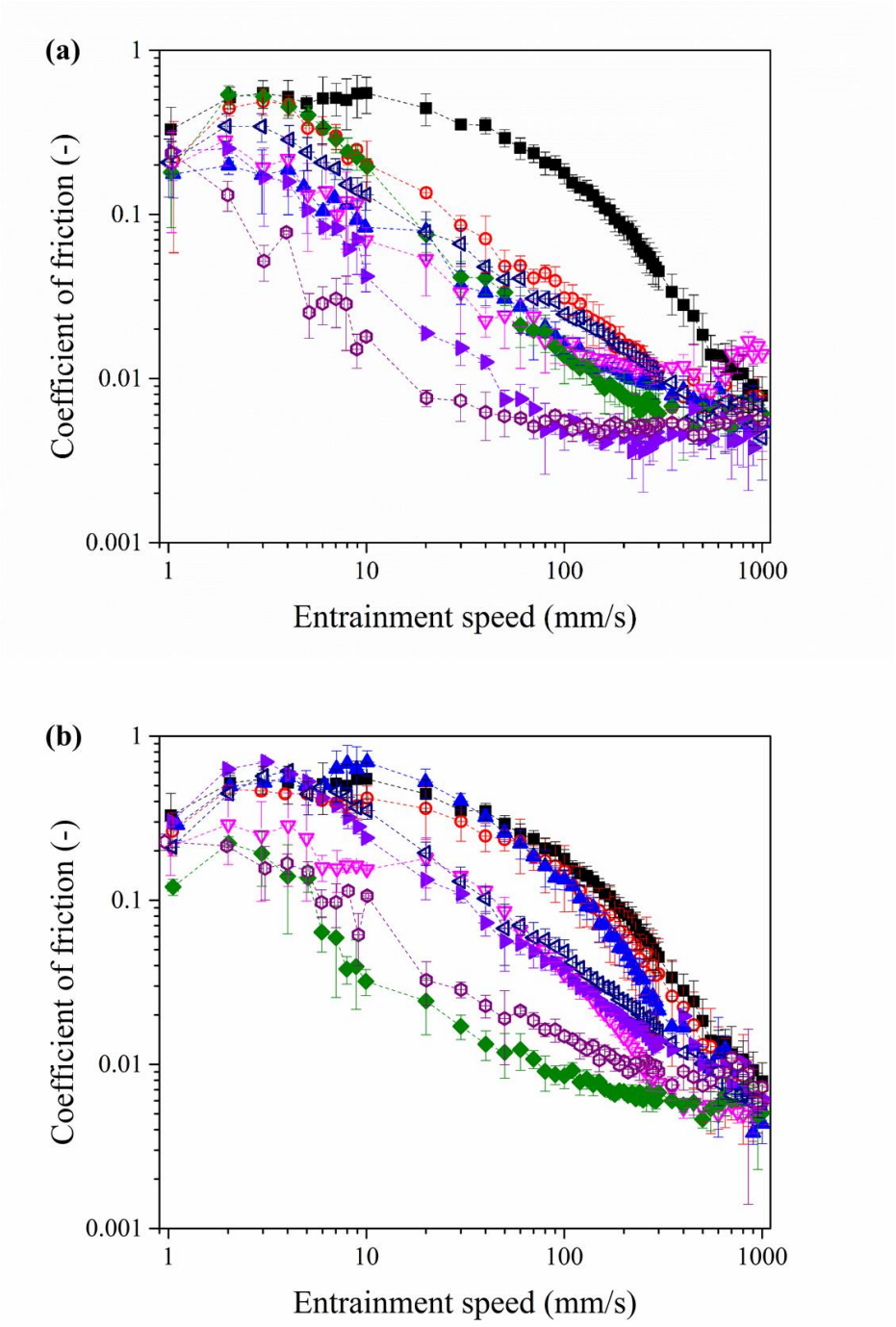

Figure 4.4. Friction coefficients of 2*κ*C (○), 3*κ*C (▲), 2.25*κ*C0.75LBG (▽), 1.5*κ*C0.5NaA (◆), 1.6*κ*C0.2CaA$_{1000}$ (◁), 1.6*κ*C0.2CaA$_{300}$ (▶) and 2.4*κ*C0.2CaA$_{300}$ (⬡) gel bolus fragments (a) and gel bolus filtrates (after filtering out the larger fragments) (b), respectively, after simulated oral processing in presence artificial saliva (■), at 37 °C as a function of entrainment speed. Data points represent the average of at least three measurements. Error bars indicate the standard deviation.

In the boundary conditions, the PDMS ball and disc appeared to be in near-adhesive PDMS-PDMS (intimate) contact, where the entrainment of the hydrogel bolus fragments or filtrates was rather poor (**Figures 4.4a and 4.4b)**. Interestingly, gel fragments containing higher concentration of *κ*C (3*κ*C), LBG (2.25*κ*C0.75LBG) and alginates as beads (1.6*κ*C0.2CaA$_{1000}$, 1.6*κ*C0.2CaA$_{300}$, 2.4*κ*C0.2CaA$_{300}$) showed some sort of entrainment even in the boundary regime reducing the friction force significantly (< 0.4 N) as compared to that of artificial saliva ($p < 0.05$) (**Table 4.4**). Gong & Osada (1998) described a "repulsion–adsorption model" to explain friction in hydrogels, which suggests that the friction force is the sum of elastic force and viscous force, which can be applied to these gel bolus fragments. The elastic force arises from anchorage of the biopolymer to the substrate (adhesive), whereas the viscous force results from the hydration of the polymer (repulsive) (Stokes *et al.* 2011; Gong and Osada 1998; Gong 2006). At a first glance, it seems that the reduction in μ in the boundary regime of these gel fragments (3*κ*C, 2.25*κ*C0.75LBG, 1.6*κ*C0.2CaA$_{1000}$, 1.6*κ*C0.2CaA$_{300}$, 2.4*κ*C0.2CaA$_{300}$) might be associated with interactions between *κ*C, LBG, NaA or CaA hydrogels and the PDMS substrates allowing biopolymer adsorption to some degree. However, this is somewhat unlikely considering the high hydrophobicity of PDMS (Sarkar *et al.* 2017) and hydrophillicity of these gels.

Hence, the relevance of 'opposing substrate' in friction in this case is worth recognizing (Gong *et al.* 2001). Note, both *κ*-carrageenan and alginates are highly negatively-charged biopolymers at pH 6.8. Thus, repulsions from both opposing PDMS substrates (artificial saliva coated *i.e.* negatively charged) (Sarkar *et al.* 2017; Sarkar, Goh and Singh 2009) as well as the opposing gel

surfaces (*i.e.* inter-gel repulsion between negatively-charged gel bolus fragments) (Gong and Osada 2002; Bongaerts, Cooper-White and Stokes 2009; Gong, Kagata and Osada 1999) are highly likely. Such repulsive interactions against the opposing artificial saliva coated PDMS substrate and/or the gel fragment surfaces, might have enabled these hydrogel fragments to remain hydrated forming a thicker solvent layer of 'lubricant', thus providing an effective barrier to the asperity contacts under the low load. This is further justified by the high viscosity values of these specific gel fragments (**Figure 4.3)** suggesting viscous force as the driving factor and separating the PDMS contacts effectively (**Figure 4.4a**).

Table 4.4. Friction force (N) for the gel bolus fluid (thin liquid) after filtering out the larger fragments at 3 mm/s (boundary lubrication regime) and 50 mm/s (mixed lubrication regime) entrainment speed and 37 °C. The samples were prepared using simulated oral processing in the presence of artificial saliva, and compared to artificial saliva as a control measure. A different lower case letter denotes a statistically significant difference ($p < 0.05$).

Samples	Friction force of the gel bolus fragments (N)				Friction force of the gel bolus filtrate (N)			
	Boundary lubrication regime (3 mm/s)		Mixed lubrication regime (50 mm/s)		Boundary lubrication regime (3 mm/s)		Mixed lubrication regime (50 mm/s)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Artificial saliva	1.097^{a}	0.211	0.585^{a}	0.071	1.097^{a}	0.211	0.585^{a}	0.071
2κC	0.978^{ab}	0.179	0.096^{b}	0.024	0.928^{ab}	0.067	0.469^{a}	0.069
3κC	0.348^{cd}	0.146	0.061^{b}	0.010	1.042^{a}	0.047	0.515^{a}	0.059
2.25κC0.75LBG	0.387^{cd}	0.076	0.049^{b}	0.018	0.498^{bc}	0.301	0.172^{b}	0.036
1.5κC0.5NaA	1.046^{ab}	0.125	0.067^{b}	0.011	0.386^{c}	0.143	0.024^{b}	0.007
1.6κC0.2CaA$_{1000}$	0.687^{bc}	0.135	0.080^{b}	0.017	1.141^{a}	0.103	0.135^{b}	0.055
1.6κC0.2CaA$_{300}$	0.338^{cd}	0.168	0.015^{b}	0.002	1.393^{a}	0.003	0.113^{b}	0.039
2.4κC0.2CaA$_{300}$	0.104^{d}	0.026	0.012^{b}	0.003	0.313^{c}	0.112	0.038^{b}	0.018

As the sliding speed of the disc started to increase, μ decreased in all samples (**Figures 4.4a and 4.4b**) and started to fill the gap between the surface asperities of the tribopairs in the mixed lubrication regime. The inclusion or exclusion of gel fragments or gel filtrate largely depends on the gap between the contacting surfaces, the size of the gel fragments compared to the size of the gap and asperities, as well as the interactions of these gel fragments with the PDMS surfaces. In the case of gel bolus fragments containing beads (1.6κC0.2CaA$_{1000}$, 1.6κC0.2CaA$_{300}$, 2.4κC0.2CaA$_{300}$) (**Figure 4.4a**), the μ was one-order of magnitude lower than artificial saliva ($p < 0.05$). The beads were larger in size as compared to the asperities of the PDMS substrates (R_a = 50 nm) and thus the beads released from the gel fragments during simulated oral processing might have rolled into the contact zone between the PDMS tribopairs, thus reducing μ values. It is tempting to propose a "ball-bearing mechanism" for the reduction in μ using CaA, such mechanism has been previously postulated for whey protein particles and protein microgel particles (Sarkar *et al.* 2017; Liu *et al.* 2016b).

To test the possibility of this ball-bearing mechanism occurrence, the Hertz pressure effects for the CaA beads of 1000 and 300 μm were calculated with the assumption that 10% of the beads were entrained between the PDMS ball and disc surfaces (Johnson 2009; Johnston *et al.* 2014; de Vicente, Stokes and Spikes 2005; Puttock and Thwaite 1969). The Young's modulus for CaA beads was assumed to be 20 kPa, based on previous studies (Larsen *et al.* 2015). From Hertz theory, the spherical contact area of the PDMS ball and disc was calculated:

$$\pi\alpha^2 = 1.31\left(\frac{R'F}{E'}\right)^{0.67} \quad (4.6)$$

where, α is the surface contact, *R'* is the reduced radius of the PDMS ball, *F* is the force for each particle entrained between the two contacts and *E'* is the reduced elastic modulus. The E' was defined as:

$$\frac{2}{E'} = \frac{1-\nu_1{}^2}{E_1} + \frac{1-\nu_2{}^2}{E_2} \quad (4.7)$$

where, *v* is the Poisson's ratio, assumed to be 0.5, and *E* is the Young's modulus (de Vicente, Stokes and Spikes 2005).

The number of particles in the contact area was calculated as:

$$N_{particles} = \frac{\varphi_{particles} \times A}{\pi R^2} \quad (4.8)$$

with φ being the concentration of particles in the contact zone, *A* the area of contact ($\pi\alpha^2$) and *R* the radius of the particles. Hence, the force per particle was calculated as:

$$F_{particle} = \frac{F_{total}}{N_{particles}} \quad (4.9)$$

with *F* the total force applied and *N* the number of particles. Finally, the spherical contact area was determined using the approach of distant points (Johnson 2009):

$$\alpha = \left(\frac{9F^2}{16R'E'^2}\right)^{1/3} \tag{4.10}$$

with *E'* defined as:

$$E' = 2\left(\frac{1-{\nu_1}^2}{E_1} + \frac{1-{\nu_2}^2}{E_2}\right)^{-1} \tag{4.11}$$

The results shown in **Table 4.5** clearly indicate that the CaA particles were not capable of rolling as α >> size of the beads, even with 10% particles being entrained between the PDMS surfaces. Interestingly, the 1000 μm CaA beads were too large to actually be entrained in the contact zone. These calculations indicate that there was no "ball-bearing effects" using CaA irrespective of the particle size studied, and the reduction in μ could be explained by the rheological behaviour (**Figure 4.3**) of the gel boli containing beads forming the viscous layer as the 'lubricant', as discussed previously. Also, not to underestimate, that the amount of water within these gel beads might also play an important role in exhibiting low friction (Gong and Osada 2002). The water might be squeezed out the gel beads forming a thin-film and may serve as a 'boundary lubricant'.

In case of the gel filtrates (**Figure 4.4b**), the Stribeck curve of the κC hydrogels almost overlapped irrespective of the biopolymer concentration in both

boundary and mixed lubrication regimes ($p > 0.05$). Similarity in friction forces for both 2κC and 3κC hydrogel bolus filtrates and artificial saliva irrespective of entrainment speeds (**Table 4.4**) suggests that the hydrogel bolus filtrates lacked the ability to migrate into and replenish the confined region in the event that the two PDMS shearing surfaces were almost in direct contact. This is unlike the behaviour in **Figure 4.4a**, where κC gel bolus fragments showed entrainment driven by the viscosity of the hydration layer created by the gel bolus fragments (**Figure 4.3**). As expected, such influence of rheology on tribology was not evident in the hydrogel bolus filtrates owing to the loss of the gel fragments particles during filtration (**Figure 4.4b**). In the mixed lubrication regime, the filtrates from hydrogel boli containing LBG (2.25κC0.75LBG), NaA (1.5κC0.5NaA) or CaA (1.6κC0.2CaA$_{1000}$, 1.6κC0.2CaA$_{300}$, 2.4κC0.2CaA$_{300}$) contributed to significantly lower friction forces as compared to the artificial saliva ($p < 0.05$) (**Table 4.4)**. This complies with the behaviour observed for the corresponding hydrogel bolus fragments (**Figure 4.4a**). Even after filtration, the spherical CaA beads might have been retained in the filtrate enabling some degree of entrainment (**Table 4.5**), or both gel fragments containing NaA and CaA were increasing the lubrication effect, possibly by 'weeping out' the water layer as a thin-film 'boundary lubricant' (Gong and Osada 2002).

Table 4.5. Elastic compression of the CaA beads based on 10% particle entrainment.

Samples	**$N_{particle}$**	**$F_{particle}$ (N)**	**α (mm)**
1.6κC0.2CaA1000	0.5	3.90	18.3
1.6κC0.2CaA300	5.7	0.35	5.5
2.4κC0.2CaA300	5.7	0.35	5.5

In our study, the fitted values *i.e.* $k = 0.0065$, $n = 0.55$, $h = 11$, $l = 0.075$, $B = 0.33 \cdot 10^{-4}$, $m = 1.0$ (see equation 4.3), gave a good fit with the Stribeck master curve as can be seen in **Figure 4.5** (de Vicente, Stokes and Spikes 2005; Bongaerts, Fourtouni and Stokes 2007). Here it should be noted that since the samples were shear thinning (**Figure 4.3**), the viscosity multipliers were calculated at each entrainment velocity. This was achieved by fitting the entrainment speeds to the shear rate by use of a power law relation of the same format as equation 4.1, and which enabled calculation of the associated viscosity. In this study, entrainment speeds of 1 to 1000 mm/s translated to shear rates of 0.1 - 100 s^{-1}. This was validated to ensure the entrainment speeds did indeed coincide with shear rates and that the predicted viscosities agreed with the shear rate/viscosity Ostwald de Waele power law regressions relevant to each sample.

As can be seen in the master curve (**Figure 4.5**), good agreement was achieved in the mixed regime and from the transitionary region into the EHL. However, in contrast to Newtonian lubricants (de Vicente, Stokes and Spikes 2005; Bongaerts, Fourtouni and Stokes 2007), using the particulate hydrogel bolus fragments, the model failed in the boundary regime. This suggests that the hydrogel bolus particles had a different degree of entrainment in the boundary regime and the key mechanism of friction reduction in the boundary regime was due to opposing surface-mediated formation of a viscous layer of ‘gel fragments’ (**Figure 4.4b**). As one might expect, such layer formation varied as a function of sample inhomogeneity under shear conditions in confinement and samples with inhomogeneity indicated a limitation in the Stribeck representation in this regime.

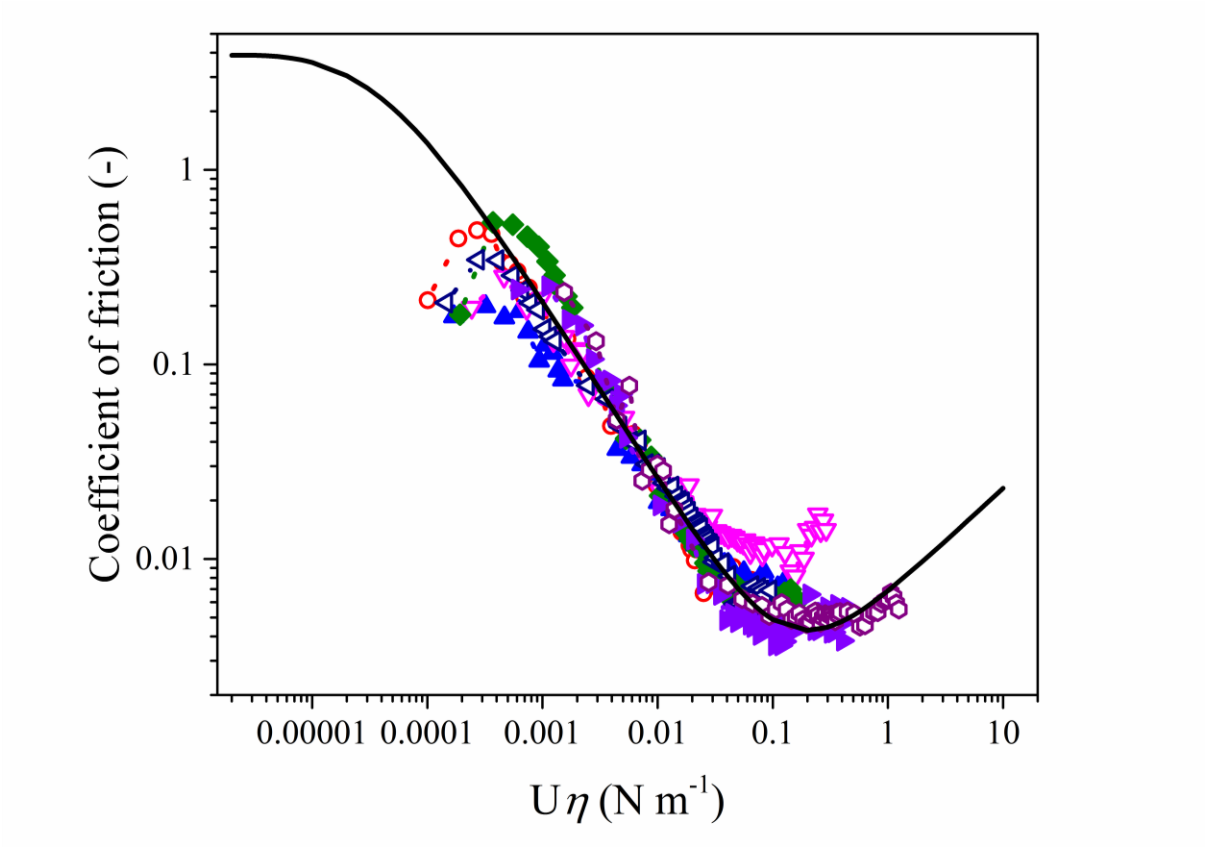

Figure 4.5. Stribeck master curve for 2*κ*C (○), 3*κ*C (▲), 2.25*κ*C0.75LBG (▽), 1.5*κ*C0.5NaA (◆), 1.6*κ*C0.2CaA$_{1000}$ (◁), 1.6*κ*C0.2CaA$_{300}$ (▶) and 2.4*κ*C0.2CaA$_{300}$ (⬡) gel bolus fragments as a function of the product of viscosity and entrainment speed component (*Uη*). The black solid line is the best fit to the data using equation (4.3).

4.3.2. Descriptive sensory analysis of the hydrogels

Table 4.2 summarizes the sensory attributes generated by the sensory panel together with their definitions. Nine different texture attributes were selected that were perceived during oral processing of the hydrogels. With the first two principal components (PC), 64% of the variance in the data was explained and the PCA plot showed that attributes were clustered in three groups (**Figure 4.6**). The pattern matrix, **Table 4.6**, shows that PC1 included the attributes related to the chewing aspects: ‘firm’, ‘elastic’, ‘chewy’ and ‘cohesive’, as well as the inverse

of the attributes more related to lubrication: ‘pasty’ and ‘melting’. The PC2 was represented by attributes that could be considered in the oral lubrication domain: ‘smooth’, ‘slippery’ and ‘salivating’ (**Figure 4.6**). At first bite, perceived firmness of a solid or semi-solid food is known to be related often to the fracture stress (Foegeding *et al.* 2011).

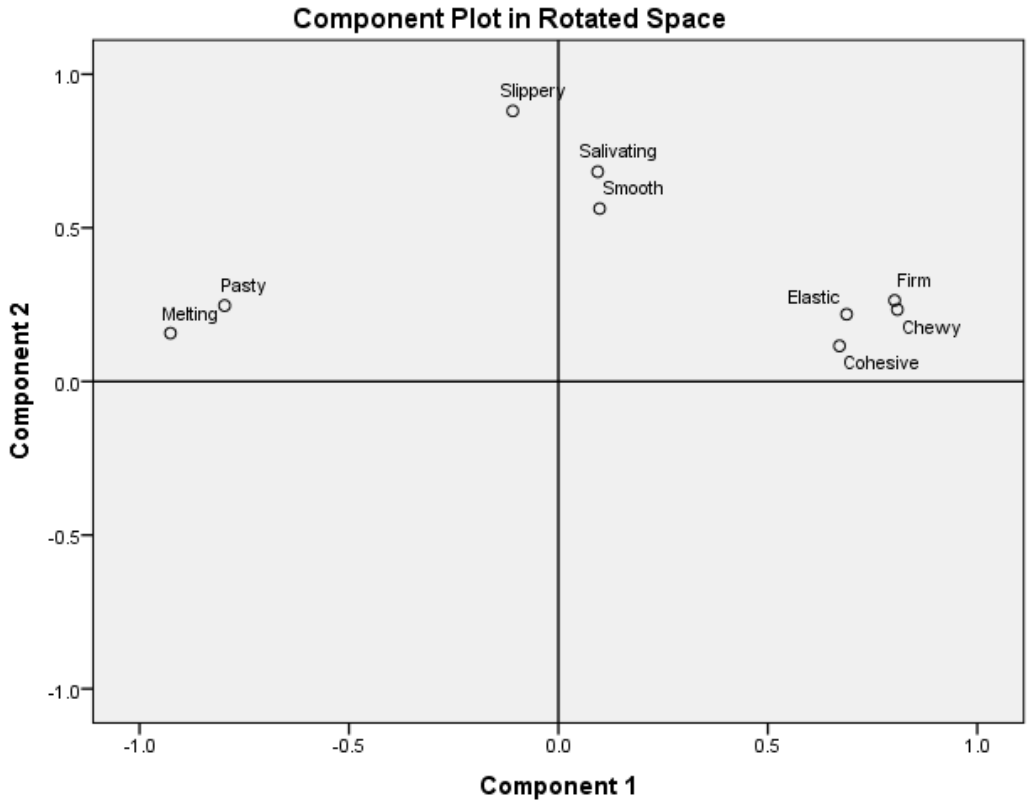

Figure 4.6. Principal component analysis (PCA) of all texture attributes obtained in the descriptive sensory analysis. Principal component 1 (PC1) represents 48.5 % and PC2 15.6 % of the variance in the data.

Table 4.6. Pattern matrix from the Principal Component Analysis (PCA). The Oblimin with Kaiser Normalisation rotation method was applied and the rotation converged in 5 iterations. Highlighted in red shows the sensory attributes best represented in PC1 and PC2 (> 0.500).

	PC 1	PC 2
Smooth	.098	.562
Firm	.803	.264
Elastic	.688	.219
Chewy	.809	.234
Cohesive	.671	.116
Pasty	-.797	.247
Slippery	-.109	.880
Salivating	.094	.682
Melting	-.926	.157

In fact, in this study, for all the chewing-related attributes (see **Figure 4.7**), the hydrogels could be categorised into two key groups. Group 1) included the hydrogels with high fracture stress/high fracture strain (*κ*C and *κ*C/LBG gels) (**Figure 4.1**) that generally scored high on the chewing-related texture attributes, such as 'firm', 'elastic' and 'cohesive', and Group 2) included the hydrogels with low fracture stress/low fracture strain (*κ*C/NaA and *κ*C/CaA gels) that scored low on these attributes. As one might expect, the chewing-related texture attributes were strongly dominated by the concentration of *κ*C *i.e.* higher concentration of *κ*C (3wt%) resulted in more 'firm' and 'chewy' perception as compared to that created using lower concentrations (2 wt%) ($p < 0.05$). Similarly for samples containing beads of the same size (300 µm), a higher concentration of *κ*C (2.4 wt%) resulted in creating samples (2.4*κ*C0.2CaA$_{300}$) that scored on the higher end of the 100 mm scale and were more 'firm', chewy', 'elastic' and 'cohesive', as

compared to that created using lower κC concentrations (1.6 wt%) ($p < 0.05$). Although the presence of beads and their particle size (300 versus 1000 µm) significantly influenced the fracture mechanics during the uniaxial compression test (**Figure 4.1**), this was not apparent in the sensory analysis of the four chewing-related texture attributes ($p > 0.05$) (**Figure 4.7**).

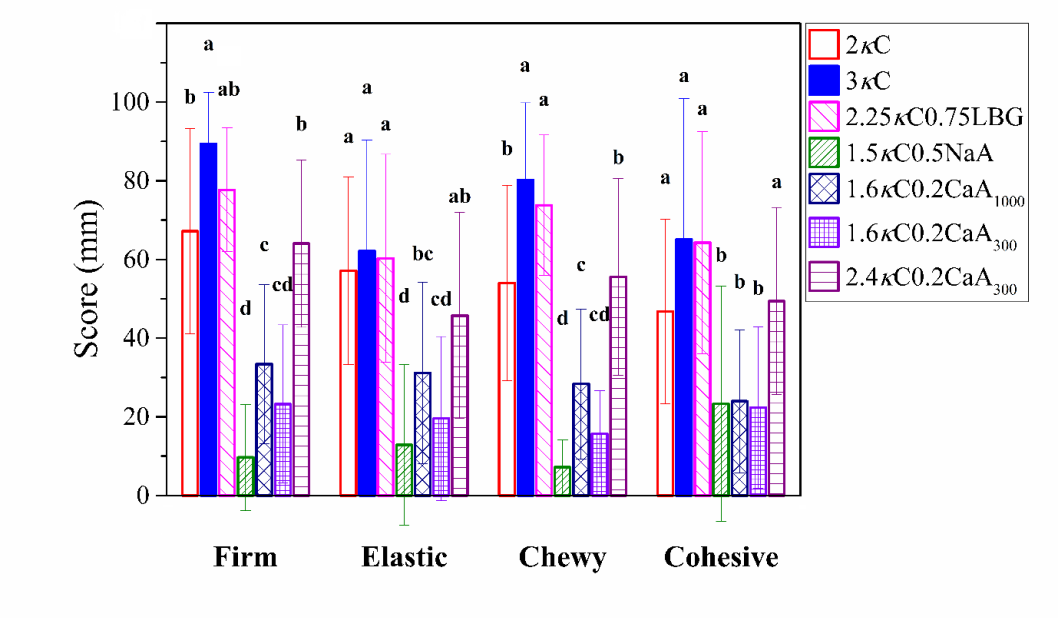

Figure 4.7. The ratings of the chewing-related attributes obtained from sensory profiling of the gels. Data points represent the average of the gels evaluated in triplicate by 11 panellists. Error bars indicate the standard deviation and bars within one attribute with different lower case letters denote a statistically significant difference ($p < 0.05$).

The lubrication-related texture attributes (see **Figure 4.8**) appeared to show a somewhat opposite effect, with the low fracture stress/low fracture strain samples scoring high on 'melting' and 'pasty', whereas the high fracture stress/high fracture strain samples scored relatively low on the 100 mm scale. The κC/NaA hydrogel (1.5κC0.5NaA) scored high on most of the lubrication-related texture attributes, such as 'smooth', 'pasty', 'melting'. It is worth remembering that the 1.5κC0.5NaA hydrogel bolus had the lowest η (though three-orders of

magnitude higher than human saliva) as compared to the other samples (**Figure 4.3**). Nevertheless, the higher scores on the lubrication-related texture attributes of the *κ*C/NaA hydrogel is in close agreement with the lower *μ* values (**Figure 4.4b**), and correspondingly lower friction force in both boundary and mixed lubrication regimes for the hydrogel bolus filtrate (**Table 4.4**). This suggests that the viscosity-parameter could not explain the lubrication-related texture attributes in case of *κ*C/NaA hydrogel and it was the 'weeping' water film that might have acted as a 'boundary lubricant'. Interestingly, the *κ*C and *κ*C/LBG hydrogels scored significantly low on 'pasty' and 'melting' ($p < 0.05$), congruent with the high *μ* of the hydrogel bolus filtrate (**Figure 4.4b**) and their correspondingly high friction forces in both the boundary and mixed regimes (**Table 4.4**).

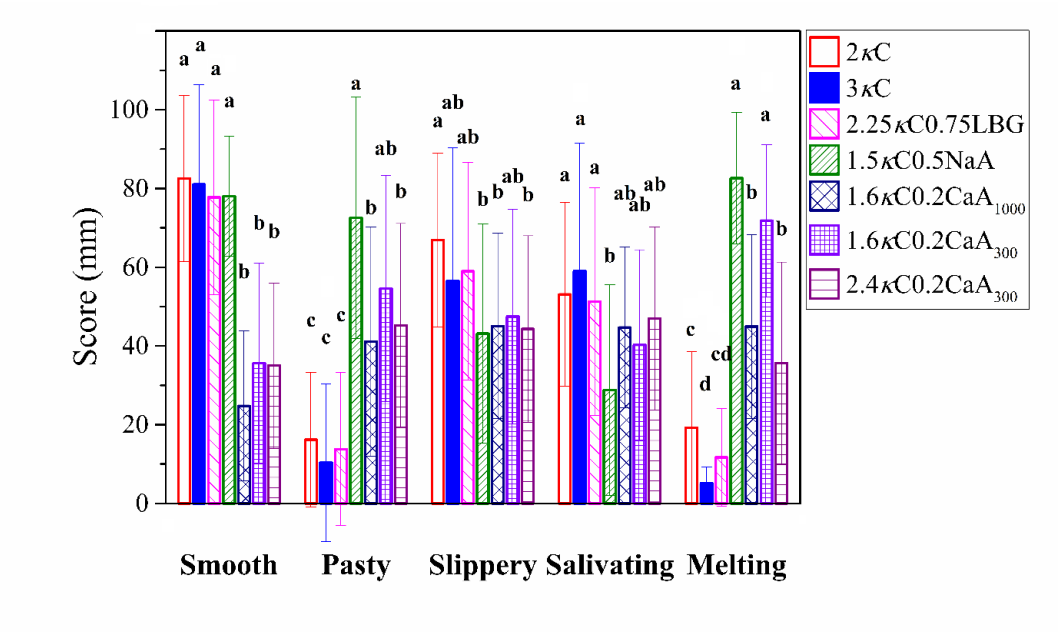

Figure 4.8. The ratings of the lubrication-related attributes obtained from sensory profiling of the gels. Data points represent the average of the gels evaluated in triplicate by 11 panellists. Error bars indicate the standard deviation and bars within one attribute with different lower case letters denote a statistically significant difference ($p < 0.05$).

The *κ*C/CaA hydrogels with beads (1.6*κ*C0.2CaA$_{300}$, 1.6*κ*C0.2CaA$_{1000}$, 2.4*κ*C0.2CaA$_{300}$) scored rather intermediate (30-60 mm) on all lubrication attributes. They were perceived to be more 'melting' and 'pasty' as compared to the *κ*C and *κ*C/LBG hydrogels ($p < 0.05$) (**Figure 4.8**), which corresponds with the reduced friction coefficients (**Figure 4.4b**) and equivalent friction force in the mixed regime for these samples (**Table 4.4**). However, considering that these beads were not rolling, as discussed before, the beads appeared to provide some degree of inhomogeneity perception, which might explain the relatively low scores on the attribute 'smooth' ($p < 0.05$) as compared to that of their absence in the other hydrogels, irrespective of particle size (**Figure 4.8**). It is worth noting that the sensory perception of particles is not only dictated by the particle size, but also by its concentration, shape, roughness and hardness of the particles as well as the properties of the matrix in which it is dispersed. For example, the sensory threshold for particle size in chocolate is ~ 30 μm (Afoakwa, Paterson and Fowler 2007), whereas for sharp-faceted silica particles it is as low as 2 μm (Engelen *et al.* 2005) to be perceived as rough and/or gritty. Also, a thicker matrix can mask the sensory detection of particles (Sala and Scholten 2015; Imai, Hatae and Shimada 1995). Thus, the observed low ratings in sensory smoothness might have resulted from these soft big CaA beads (≥ 300 μm) with sizes much above the sensory detection threshold, the inability of the *κ*C matrix to mask such perceptions as well as the absence of any ball-bearing effects.

It is worth highlighting that although the *κ*C hydrogels scored high on the sensory attribute 'smooth' (**Figure 4.8**), the friction coefficients of *κ*C hydrogel bolus filtrates were highest in the boundary regime ($\mu \sim 0.5$) (**Figure 4.4b**). Noteworthy is that the PDMS substrates used in this study for tribology were

highly hydrophobic (even after pre-adsorption of artificial saliva), which might not have allowed efficient polymer-adsorption of the hydrophilic κC hydrogel bolus particles remaining in the filtrate to the substrates and thus did not reduce friction significantly ($p > 0.05$). This might not be the case during real oral processing as the oral mucosa is highly hydrophilic because of the salivary coating (Laguna and Sarkar 2017). Hence, one might consider introducing some degree of hydrophilicity in these soft PDMS substrates for doing oral tribology measurements in order to accurately understand this sensory smoothness scores for κC hydrogels (Sarkar *et al.* 2017). Interestingly, the friction coefficients of the κC hydrogel bolus fragments (particularly 3κC) was considerably low ($\mu \sim 0.15$) in the boundary regime (**Figure 4.4a**, **Table 4.4**). This suggests that the 3κC gel bolus fragments were responsible for acting as a solvated layer of lubricant to reduce viscous friction, as discussed previously, and consequently were rated high on the sensory attribute, 'smooth' (**Figure 4.8**).

For the attributes 'slippery' and 'salivating', the trend was not very clear for samples containing NaA or CaA (**Figure 4.8**), which might be associated with the rather difficult definitions and the unfamiliarity of the panel with these lubrication-related texture attributes. This can also be seen in the Tucker-1 plots, see **Figure 4.9**, where the attributes 'slippery' and salivating' showed rather random clustering patterns indicative of poor panel agreement. It appears that insufficient training was provided to the participants on these constructs for them to grasp the complexity of these attributes in novel soft solid systems *i.e.* the hydrogels with different textural complexity. Only samples containing NaA scored significantly lower on 'salivating' as compared to that of κC or κC/LBG hydrogels ($p < 0.05$). 'Salivating' was defined as 'amount of saliva released

during chewing' (**Table 4.2**). Therefore, it is likely that panellists rated κC or κC/LBG hydrogels as more 'salivating' compared to that of NaA samples ($p < 0.05$) as possibly a larger quantity of saliva was generated for cleansing the residues of the stiffer hydrogel fragments (**Figure 4.1**). Similarly, samples containing NaA and CaA (except 1.6κC0.2CaA$_{300}$) scored significantly lower on the attribute 'slippery' compared to that of κC hydrogels ($p < 0.05$). This suggests that samples containing alginate as biopolymer or beads did not slip easily and provided some sort of oral coating properties due to the alginate itself or created a 'weeping' layer of water as a lubricant during tribological shearing (Gong and Osada 2002), as discussed in the previous section. The oral coating property of alginates is in agreement with literature suggesting that alginate can create hydrogen bonds with human salivary mucins through carboxyl–hydroxyl interactions (Shtenberg *et al.* 2018; Cook *et al.* 2017).

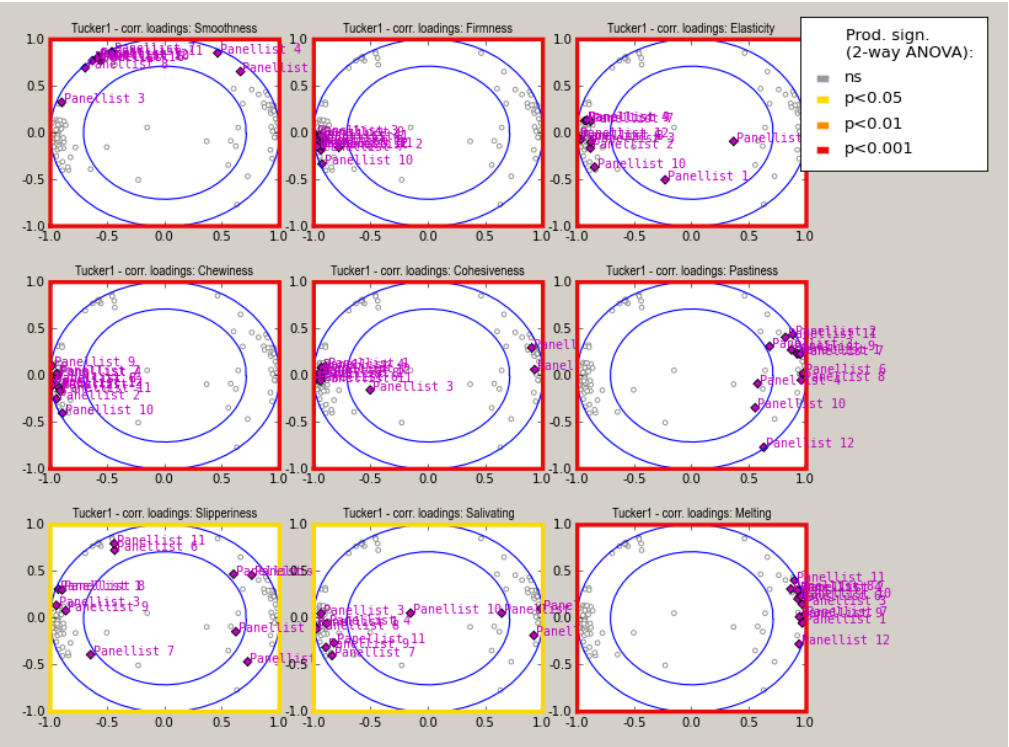

Figure 4.9. Tucker-1 plots for the all attributes from the descriptive sensory analysis, showing the panel agreement.

In general, it can be concluded that all chewing-related attributes were largely controlled by the fracture properties of the hydrogels, whereas the lubrication-related attributes showed significant variations between the hydrogel samples and some of the lubrication-related attributes corroborated the oral tribology results of the gel bolus filtrates in the mixed lubrication regime. Noteworthy is that the relationship between fracture properties, tribology and sensory analysis has been investigated in literature, particularly in emulsion gels (Devezeaux de Lavergne *et al.* 2015a; Liu *et al.* 2016a). Nevertheless, to our knowledge, this is the first study that has been carried out using descriptive sensory analysis focussing on textural attributes related to both chewing and lubrication in aqueous systems *i.e.* hydrogels with varying degree of textural complexity.

4.3.3. Relationship between instrumental and sensory properties of the hydrogels

To understand the complex sensory perceptions in these hydrogels with or without inhomogeneity, an integrative approach of identifying interrelationships between sensory textural attributes and instrumental parameters rather than dependence on a single instrumental test is necessary. **Table 4.7** highlights the statistically significant correlation coefficients between the broad spectrum of mechanical parameters, *i.e.* fracture properties, apparent viscosity, μ in boundary and mixed lubrication regime, and the texture attributes.

Table 4.7. Pearson's correlations of sensory attributes (descriptive analysis) and physical properties (large deformation rheology, apparent viscosity and coefficient of friction) of the hydrogels, where green is positive and red shows a negative correlation with $p < 0.05$ in light colours and $p < 0.01$ in the darker shade.

		Smooth	Firm	Elastic	Chewy	Cohesive	Pasty	Slippery	Salivating	Melting	Fracture stress	Fracture strain	Fracture Energy	Viscosity at 50 s^{-1} shear rate	μ at 50 mm/s	μ at 3 mm/s	μ at 50 mm/s	μ at 3 mm/s
Sensory	**Smooth**																	
	Firm	0.40																
	Elastic	0.44	0.98															
	Chewy	0.41	0.99	0.98														
	Cohesive	0.53	0.96	0.94	0.98													
	Pasty	-0.43	-0.92	-0.95	-0.91	-0.84												
	Slippery	0.66	0.68	0.77	0.64	0.63	-0.84											
	Salivating	0.25	0.94	0.94	0.91	0.82	-0.95	0.71										
	Melting	-0.38	-0.97	-0.99	-0.97	-0.91	0.97	-0.73	-0.96									
Texture analysis	**Fracture stress**	0.59	0.96	0.96	0.95	0.95	-0.94	0.80	0.91	-0.94								
	Fracture strain	0.36	0.87	0.90	0.87	0.80	-0.98	0.80	0.92	-0.92	0.91							
	Fracture Energy	0.55	0.89	0.87	0.87	0.84	-0.88	0.72	0.90	-0.87	0.94	0.82						
Rheology	**Viscosity at 50 s^{-1} shear rate**	0.30	0.91	0.89	0.88	0.80	-0.90	0.67	0.98	-0.91	0.80	0.86	0.95					
Tribology, gel bolus filtrate	**μ at 50 mm/s**	0.56	0.68	0.71	0.63	0.57	-0.80	0.82	0.79	-0.72	0.79	0.74	0.90	0.85				
	μ at 3 mm/s	-0.30	-0.11	-0.11	-0.16	-0.30	-0.15	0.14	0.22	0.04	-0.03	0.25	0.14	0.24	0.39			
Tribology, gel bolus fragments	**μ at 50 mm/s**	0.47	0.06	0.19	0.05	0.00	-0.31	0.48	0.16	-0.24	0.18	0.22	0.25	0.16	0.56	0.15		
	μ at 3 mm/s	0.42	-0.44	-0.31	-0.45	-0.42	0.22	0.17	-0.40	0.31	-0.28	-0.30	-0.23	-0.37	0.18	0.34	0.81	

Positive correlations were obtained between the chewing-related sensory attributes, *i.e.*, 'firm', 'elastic', 'chewy' and 'cohesive' and the instrumental measures of fracture stress, fracture strain, fracture energy and viscosity at 50 s^{-1} (**Table 4.7**). The correlations of the fracture parameters with the chewing-related attributes are in agreement with previous literature dealing with emulsion gels (Devezeaux de Lavergne *et al.* 2015b) and agarose gels (Barrangou *et al.* 2006). This suggests that firm samples, such as, *κ*C and *κ*C/LBG will require more stress to deform, particularly in the early stages of oral processing.

Interestingly, the lubrication-related sensory attribute 'salivating' also showed strong positive correlations with instrumental measures of fracture stress, fracture strain, fracture energy and viscosity at 50 s^{-1}, respectively. As discussed previously, the firm samples might have created residues/particles, which required increased salivary flow for oral cleansing (**Table 4.2**). Hence, it appears that the trained panel might have associated sensory 'salivating' with the quantity rather than the quality of saliva production. In addition, strong inverse relationships were obtained between the chewing-related sensory attributes, *i.e.*, 'pasty', and 'melting' and the instrumental measures of fracture stress, fracture strain, fracture energy of the hydrogels and viscosity of boli at 50 s^{-1} ($p < 0.01$). In other words, 'firm' samples, such as *κ*C or *κ*C/LBG hydrogels might have created bolus fragments during the oral processing that were relatively stiff, retained their integrity and were not melting easily over the duration of the oral residence time. And, due to such fragment creation, firm samples were not perceived to be 'pasty' ($p < 0.01$) *i.e.* did not form a continuous layer of soft solids. Although the sensory attribute 'smooth' showed no correlations with

either the fracture properties of the hydrogels or the viscosity of the boli, the sensory attribute ‘slippery’ showed a positive correlation with initial fracture stress and fracture strain, which suggests that the term ‘slippery’ had some association with early stages of oral processing, which was not expected ($p > 0.05$). Overall, clear relationships existed between all fracture properties of the hydrogels and rheology of the bolus fragments with lubrication-related attributes, such as ‘pasty’, ‘melting’ and ‘salivating’ that were perceived by the panellists during oral processing.

We now shift our focus to investigate whether μ of the hydrogel fragments and/or filtrates could predict the sensory dimensions of both chewing- and lubrication-related texture attributes (**Table 4.7**). As one might expect, no correlations existed between the chewing-related attributes and μ of bolus fragments/filtrates irrespective of the lubrication regimes. However, looking at the lubrication-related sensory attributes (**Table 4.7**), ‘pasty’ was inversely correlated with the μ of hydrogel bolus filtrates in the mixed lubrication regimes ($p < 0.05$). This further suggests that ‘pasty’ was most likely associated with the mouth-coating aspects during oral processing, as discussed previously. For example, samples with lower μ values in the boundary regime (*e.g.* 1.5κC0.5NaA) will be more lubricating and will thus be perceived as more ‘pasty’, forming an oral coating and/or ‘weeping’ layer of water and separating the oral surfaces from the asperity contacts. Although the signs of correlations in case of the tribology experiments performed with hydrogel bolus fragments were similar to that of the hydrogel bolus filtrate, no statistical significance was observed in the former irrespective of the lubrication-related attribute. This confirms that the bolus filtrates being thin-film had more relevance in the oral

tribology domain in this study when relating to the in-mouth sensory perception as compared to that of the bolus gel fragments.

Interestingly, there was a tendency towards an inverse relationship of sensory 'melting' to the μ of the hydrogel bolus filtrates in the mixed lubrication regime, however there was no statistical significance (**Table 4.7**). Moreover, 'salivating' was positively correlated with the μ of the bolus fragments ($p < 0.05$). This supports the explanation for the initial fracture properties, which suggests that hydrogels scoring higher on 'salivating' were the samples that generated more volume of saliva (**Table 4.2**). As can be expected, the generated saliva was perhaps depleting the gel fragments or residues from the oral surfaces. Such 'depletion' of bolus fragments or residues from the oral surfaces might have resulted in apparent surface asperity contacts, which may justify the positive correlation of 'salivating' with friction coefficients as observed in **Table 4.7**. No relationships could be observed between 'smooth' and μ of bolus filtrates, which might be attributed to the inhomogeneity of samples, such as the ones containing CaA beads. For the sensory attribute 'slippery', there was a positive correlation with μ of hydrogel bolus filtrates in the mixed lubrication regime ($p < 0.05$) (**Table 4.7**).

It is worth highlighting this observed anomaly when relating the sensory attribute of 'slippery' to the tribology results. Previously, an inverse relationship of friction coefficient and slipperiness in foods, *i.e.* $slipperiness\ \alpha\ \frac{1}{Viscous\ force + W\mu}$ (where W is the applied load in tribology), has been postulated by Kokini (1987). However, this previous study by Kokini (1987) was done with fat-rich low viscosity fluids, where 'slipperiness' could be

perceived easily due to ‘fatty’ or ‘creamy mouthfeel’. In comparison, the current study has employed semi-solid aqueous hydrogels and their corresponding bolus fragments and filtrates. Furthermore, ‘slippery’ perception was defined by the ease of sliding of the sample (**Table 4.2**). This suggests that highly slippery samples, such as 2κC gels (**Figure 4.8**) were sliding past the oral mucosa easily, which apparently resulted in having no fragments/ filtrate in the confinement, corroborating with the high μ values (**Figure 4.4b**, **Table 4.4**). Nevertheless, it is worth emphasising that both ‘slippery’ and ‘salivating’ were difficult sensory terms for the panellists (**Figure 4.8**), as discussed before, and so these correlations should be taken with caution.

4.4. Conclusions

This study presents hydrogels as model soft solid foods, where systematic manipulation of the structural properties was used to investigate the relationship between mechanical (instrumentally measured) and sensory aspects of oral processing. A range of hydrogels with varying degrees of structural complexity was evaluated using uniaxial compression test of the hydrogels, flow curves and tribology of gel boli (after simulated oral processing) as well the sensory properties, which was investigated using descriptive sensory analysis. Tribology of the bolus fragments and filtrates were explained using theoretical “repulsion-adsorption” model highlighting the role of opposing surfaces (PDMS, gels). A clear correlation was obtained between the initial fracture properties of the hydrogels, viscosity of the bolus fragments and all chewing-related texture attributes *i.e.* ‘firm’, ‘elastic’, ‘chewy’ and ‘cohesive’. Interestingly, all fracture attributes and boli viscosity showed positive correlations with the relatively novel

lubrication-related texture attributes, such as ‘salivating’ and inverse correlations with both ‘pasty’ and ‘melting’. The coefficient of friction of the bolus filtrates in the mixed lubrication regime showed inverse correlations with the lubrication-related attributes, such as sensory ‘pasty’ and positive correlations with ‘slippery’ and ‘salivating’. However, our experimental design could not establish a significant inverse correlation between sensory ‘smooth’ and the friction coefficients, which is largely attributed to the inhomogeneity of the samples employed in the study. Novel findings from this study suggests that lubrication-related attributes were perceived during both early and later stages of oral processing and thus relationships existed with initial fracture properties of gels, boli viscosity and boli tribology and not only to boli tribology, as hypothesized initially. Future studies could conduct more independent systematic studies on hydrogels with varying degrees of structural complexity at micro- to macro-scale to establish the tribology-sensory relationships particularly at the later stages of oral processing. In addition, further training of panel members particularly with respect to lubrication-related texture attributes could be done to achieve more uniform responses and less variability in descriptive analyses. However, it is recognised that there are large individual differences which account for variance in sensory perception, and as an adjunct approach to training panels it is important to discern the contribution of these differences to sensory perception of the physical manipulations of the hydrogels.

In order to explore the impact of both the instrumental and sensory properties of the hydrogels, as well as the individual differences in consumer perception and eating capability, a further study was conducted. The next chapter reveals the results of this study, examining the effects on the oral processing

behaviour of the same hydrogels as characterised here and the eating capabilities of a group of young, healthy participants (**Chapter 5**).

4.5. References

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Chapter 5

Oral processing of hydrogels: influence of food material properties versus individuals' eating capability[c]

Abstract

Food material properties play an important role in the sensory perception and consumer acceptance of foods. However, the actual oral processing behaviour may depend on both the material properties of the food that is being consumed as well as individuals' oral capabilities. This study aimed to examine the relationships between *intrinsic* (oral capabilities of healthy participants), as well as *extrinsic* (food material properties of a set of hydrogels) variables to the real oral processing behaviour. Three *κ*-carrageenan hydrogels (*κ*C), differing in fracture mechanics and oral tribology properties, were prepared: native *κ*C, *κ*C with added Na-alginate and a *κ*C matrix with added Ca-alginate beads of 300 μm. A composite score of eating capability (EC) was measured with non-invasive techniques (maximum bite force and tongue pressure) using a panel of 28 untrained consumers. The oral processing behaviour (number of chews, oral

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residence time and chewing rate) was analysed with the same participants using frame-by-frame video analysis. It was found that the EC scores did not correlate with any of the oral processing behaviours. The number of chews and oral residence time showed a strong correlation to the fracture force and friction force at orally relevant speeds (10-100 mm/s), whereas chewing rate did not vary with these properties. The results from this study indicate that oral processing in healthy adults seems mainly motivated by food material properties, and the chewing rate seems to relate more to individual differences and EC than to food properties.

5.1. Introduction

As global obesity rates increase, there have been intensified efforts to design satiety-enhancing foods that can decrease appetite and thus reduce food intake in the longer term. Previous studies have demonstrated the role of oral processing on satiety (Miquel-Kergoat *et al.* 2015), and in a recent systematic review and meta-analysis it was found that "oral processing" leads to a significant reduction in food intake (-0.28 effect size, 95 % CI: -0.36, -0.19) (Krop *et al.* 2018). Here, the term "oral processing" incorporated a variety of strategies, such as increased number of chews, eating rate or bite size, extended oro-sensory exposure time and/or introducing harder textures as compared to a softer/liquid variant. However, the effects of salivation and food/saliva interactions were not considered in the retrieved studies (Krop *et al.* 2018).

Previous evidence revealed that the actual oral processing strategy is adapted to the *extrinsic* material properties of the food that is being consumed (Koç *et al.* 2014; Le Révérend *et al.* 2016), but also varies between individuals

according to their *intrinsic* oral capabilities (Peyron *et al.* 2011; Wilkinson, Dijksterhuis and Minekus 2000). During oral processing, the food's physical properties are continuously manipulated with the food structure being broken down and mixed with saliva and fluid released from the food matrix to form a cohesive bolus (Chen 2009). Therefore, both texture properties and the degree of moisture of the initial food structure contribute significantly to oral processing (Hutchings and Lillford 1988). The central nervous system uses sensory feedback from the changing physical properties during oral processing to update the oral processing strategy, from visual cues before ingestion, to first bite until swallowing (Koç *et al.* 2014; de Wijk, Engelen and Prinz 2003; van der Bilt *et al.* 1995).

Research on chewing has primarily focussed on solid foods, using various techniques to quantify chewing behaviour. In a study by Hiiemae *et al.* (1996), it was found that the number of chews and oral residence time increased for foods with a more complex initial structure, with banana requiring less chewing than biscuits. In another study by Forde *et al.* (2013), the number of chews related to the number of bites for 50 g food sample, and the eating rate was inversely related to the number of chews. Previous studies have also made the link between commercial food products like cheese, peanuts and carrots, where harder and drier foods required more chewing (Engelen, Fontijn-Tekamp and van der Bilt 2005; Fontijn-Tekamp *et al.* 2004). In addition, Engelen, Fontijn-Tekamp and van der Bilt (2005) found that quantity of saliva and maximum bite force were only weakly correlated with chewing characteristics, accounting for less than 10 % of the variance in the number of chews. However, these products are highly familiar with learned expectancies for processing and satiety.

Hydrocolloids have been used in research to make model foods to study texture and oral processing behaviour without invoking an emotional response and expectancies built up from prior experience with real life products (Funami 2011; Funami *et al.* 2012; Funami *et al.* 2016; Hayakawa *et al.* 2014; Nishinari 2004; Larsen *et al.* 2016; Laguna and Sarkar 2016). Previous studies observed a relationship between food hardness and chewing behaviour, where fracture stress from instrumental texture analyses was correlated to higher number of chews and increased oral residence time (Funami *et al.* 2016; Devezeaux de Lavergne *et al.* 2016; Laguna and Sarkar 2016). Moreover, from bolus particle analysis it was found that harder and more complex model gels break down into significantly higher number of particles that are smaller in size (Larsen *et al.* 2016; Devezeaux de Lavergne *et al.* 2016).

Aside from fracture properties, the effects of food structure on oral lubrication (both internal and external) have gained increased research attention. Human saliva binds particulated food into a cohesive bolus that can be easily swallowed (Pedersen *et al.* 2002; Carpenter 2012). In addition, the moisture content in foods (providing external oral lubrication) has been linked to the used oral processing strategy (Hutchings and Lillford 1988). A dry solid food (*e.g.* biscuits) will generally require a large quantity of saliva in order to form a swallowable bolus, whereas more moist solid foods, such as fruits and vegetables, already contain a large quantity of moisture that is released during oral processing (Chen and Rosenthal 2015). However, due to the continuously changing nature of the food structure during oral processing, the effects of external lubrication by food on oral processing behaviour remains a challenging research topic (Chen 2009).

Besides the *extrinsic* food properties, oral processing also depends on *intrinsic* individual differences in oral physiology, from the size of the oral cavity to the strength of the oro-facial muscles (Alsanei and Chen 2014; Engelen, Fontijn-Tekamp and van der Bilt 2005). Several studies have mentioned that chewing patterns vary not only between individuals but also within the same person. In a study by Lassauzay *et al.* (2000) the number of chews for gelatine based model foods in male individuals varied from 19 to 57, with a similar variability found for the different test foods. Another study by Brown *et al.* (1994) using healthy subjects also reported large variations between subjects for the tested foods, such as apple, salami and toast, with raw carrot showing particularly big differences in number of chews and oral residence time, ranging from 20 to 190 chews and 15 to 125 s, respectively. Furthermore, the effects of gender and age on masticatory ability have been reported in literature. Males have a bigger bite size, faster eating rate and a higher EMG muscle activity than females (Peyron *et al.* 2004; Park and Shin 2015), whereas females chewed more and for a longer time than males ($p < 0.05$) (Park and Shin 2015). Also, due to the decrease of masticatory muscle mass and maximum bite force with age (Bakke *et al.* 1990), the number of chews and EMG activity increased in older participants who still had complete healthy dentition compared to younger adults (Kohyama, Mioche and Martin 2002; Peyron *et al.* 2004). At the same time, salivary secretion, saliva viscosity, and its protein content varies widely between individuals, as well as within the same individual at different times of the day (Carpenter 2012), and would therefore be expected to influence oral processing.

Thus, understanding the relationship between oral processing behaviour, food material properties (determined both instrumentally and sensorially) and

individuals' eating capabilities is important in determining what drives the consumer experience of eating a food and what leads to consumer acceptance and preference. To date, no studies have looked at the oral processing behaviour of hydrogels in young individuals, examining both the food material properties as well as the participants' individual eating capability. Therefore, the aim of this study was to investigate the oral processing response and their potential relationships with 1) the *extrinsic* food material properties of different hydrogels (*i.e.* fracture behaviour and oral lubrication) and 2) the *intrinsic* eating capability of a group of young healthy consumers.

5.2. Materials and methods

5.2.1. Materials

Food grade-quality *κ*-carrageenan (*κ*C) and sodium alginate (NaA) were purchased from Special Ingredients Ltd (Chesterfield, UK). Green food colouring was obtained from AmeriColor (Placentia, USA) and American peppermint extract was purchased at a local supermarket (Leeds, UK). Potassium chloride (KCl) was purchased from Minerals Water Ltd (Purfleet, UK) and calcium chloride (CaCl) from VWR International (Leuven, Belgium). All materials were used without further purification. Demineralised water was used in preparation for all hydrogels.

5.2.2. Hydrogels

Based on a previous study by (Krop *et al.* 2019), three model hydrogels (that did not contain any fat) were selected that had different chewing and oral lubrication properties as determined by instrumental and sensory texture analysis. The

hydrogels consisted of varying concentrations of κC alone or with the addition of NaA or calcium alginate (CaA) beads of 300 μm diameter. The selected model gels were 3κC (3 wt% κC), 1.5κC0.5NaA (1.5 wt% κC and 0.5 wt% NaA) and 2.4κC0.2CaA$_{300}$ (2.4 wt% κC and 0.2 wt% CaA beads of 300 μm). The hydrogels were unsweetened, but flavoured with peppermint aroma and coloured with green food colouring to increase acceptability. Further details on the preparation method, as well as instrumental and sensory characterisation of the hydrogels have been published elsewhere (Krop *et al.* 2019). The samples were presented in bite-size round pieces (diameter 25 mm, height 10 mm) in small, shot-glass type plastic cups.

5.2.3. Puncture test (texture analysis)

The mechanical properties of the hydrogels were determined using uniaxial puncture test with a Texture Analyzer (TA-TX2, Stable Micro Systems Ltd., Surrey, UK) , with a 30 kg load cell. The fracture mechanics were measured using a 10 mm Volodkevitch bite jaw probe to simulate a first bite with human incisor teeth. Tests were carried out at 22 °C, at a constant speed of 2 mm/s and the deformation level was set at 80 % strain. Six replicates were measured for each hydrogel on at least four different preparation days. The software Exponent (TEE32, v6.1.9.0, Stable Micro Systems Ltd., Surrey, UK) was used to plot the force-distance curve.

5.2.4. Tribological measurements

The oral lubrication properties of the hydrogels after simulated oral processing were determined with a Mini Traction Machine (MTM2, PCS Instruments, London, UK), based on a method developed by Krop *et al.* (2019). Briefly, the

hydrogels were broken down in presence of artificial saliva containing mucin at pH 6.8 (Sarkar, Goh and Singh 2009) with a mechanical blender (final sample to saliva ration 4:3 w/w). The larger gel particles (> 500 μm) were filtered out, and the friction behaviour of the bolus filtrate was determined. Commercially available polydimethylsiloxane (PDMS) ball (diameter of 19 mm) and disc (diameter of 46 mm, thickness of 4 mm) were obtained from PCS Instruments (MTM ball and disc, Sylgard 184, 50 Duro, London, UK), with the surface roughness of the PDMS tribopairs, R_a < 50 nm. The friction force in the mixed lubrication regime was determined as a function of the applied entrainment speed, ranging from 10 to 100 mm/s, with an applied load of 2 N, slide-to-roll ratio (SRR) of 50 % at 37 °C. Measurements were performed in triplicate and then averaged to obtain the Stribeck curves.

5.2.5. Participants

Twenty-eight healthy participants with natural, intact dentition were recruited to participate in this study and gave written informed consent before the start of the study. The study was reviewed and approved by the Faculty Research Ethics Committee at the University of Leeds (reference number MEEC 16-006). Participants were aged between 22 and 52 years old (mean 28.5 ± SD 6.2, 9 male and 19 female). Participants with any allergies/intolerances to the gel ingredients were excluded, as well as those who suffered from any condition hampering normal chewing or swallowing. All participants received a financial compensation.

5.2.6. Study procedure

Test sessions were conducted in sensory booths at the School of Food Science and Nutrition, University of Leeds. Prior to the start of the study, participants were instructed that they would be video recorded while eating the three model foods and that afterwards eating capability measures (bite force and tongue pressure) would be taken. Participants were given the opportunity to ask questions in case anything was unclear, after which they provided written, informed consent form. Samples were provided to the panellists in randomised order, and a practice sample was provided to familiarise the panellists with the type of test samples and the eating instructions.

5.2.7. Video analysis of oral processing characteristics

To analyse the oral processing behaviour, participants were video recorded while eating the model foods. A digital camera (Panasonic SDR-H90) was positioned in front of the participant on a tripod, and participants were instructed to look straight into the camera while eating the hydrogels. Participants were aware that their oral processing behaviour would be analysed, such as number of chews and eating rate, and were given the option to swallow the samples or indicate the moment they felt the urge to swallow by raising their hand and expectorate the sample in provided containers. Videos were analysed frame-by-frame using Observer XT 12 software (v 12.5, Noldus Information Technology, The Netherlands). A coding scheme was created to identify the first bite, number of chews and point of swallowing, as adapted from previous studies (Lasschuijt *et al.* 2017; Forde *et al.* 2013). A chew was defined as the point in time when the jaw was at the lowest position during a masticatory cycle (closing action). From

these behaviours, the eating duration was determined as the time between first bite and swallowing, identified as the first main swallow at the end of the rhythmic rotary chewing movements. The chewing frequency was calculated by dividing the number of chews for each sample by the total eating duration of this hydrogel (Chen and Lolivret 2011; Forde *et al.* 2013; Laguna *et al.* 2016b; Laguna and Sarkar 2016).

All videos were coded by a trained researcher, with a second observer analysing at least 15 % of the videos in parallel to assess the accuracy of the coding scheme and the performance of the coders. The performance of the researchers assessing the videos was validated using a reliability analysis, showing at least 85 % agreement.

5.2.8. Eating capability measurements

Tongue pressure was measured using the Iowa Oral Performance Instrument (IOPI®, Medical LLC, Redmond, Washington, USA (Ono *et al.* 2009; Laguna *et al.* 2015). Participants were instructed to place the plastic bulb sensor in the centre of the oral cavity between their tongue and the hard upper palate, and press these surfaces together with their maximum strength. The maximum tongue-palate pressure was recorded in kPa.

The maximum biting force was measured using force sensors and a multimeter connected through a bread board, a device previously used by Flanagan *et al.* (2012). The force sensor was sandwiched between two adhesive silicone disks (diameter 1.5 cm, thickness 0.3 cm), which in turn were wrapped in wrapping foil for hygienic reasons. Participants were instructed to bite down separately on the sensor for a couple of seconds using their front incisors, left

molars and then their right molars. The minimum resistance was independently recorded by the multimeter for the front incisors, left molars and right molars and subsequently converted into N (Laguna *et al.* 2015; Flanagan *et al.* 2012). Both measures of tongue pressure and bite force were measured in triplicate for each participant.

Eating capability (EC) has been defined as "the physical, physiological and cognitive capabilities of an individual consuming food" (Laguna *et al.* 2016b). In previous studies, a composite EC score was used that consisted of grip strength, manual dexterity, and oro-facial muscular capability (tongue pressure and bite force) (Laguna *et al.* 2016b; Laguna *et al.* 2015). It was determined that the most important factors in determining the EC score were related to the oro-facial muscular capabilities, therefore for this study only tongue pressure and bite force were included. The EC composite score was calculated using the following equation:

$$EC = \left(\frac{TP}{TP_{max}}\right) + \left(\frac{\frac{BF_L}{BF_{L,max}} + \frac{BF_F}{BF_{F,max}} + \frac{BF_R}{BF_{R,max}}}{3}\right) \quad (5.1)$$

where, TP represents the tongue pressure and BF the biting force measured for each participant. The subscript max indicates the highest value measured in the strongest participant for that particular variable, L is the bite force from the left side molars, F from the front incisors and R from the right side molars. Thus, the maximum EC score that could be obtained was 2-points.

5.2.9. Statistical analysis

All statistical analyses were performed using SPSS (IBM® SPSS® Statistics, v24, SPSS Inc, Chicago, USA). Results are presented as mean ± standard deviation

(SD) and alpha was set at $p < 0.05$, unless stated otherwise. Analysis of variance (ANOVA) was applied to determine statistically significant differences between samples for the fracture mechanics and eating behaviour, and between participants for the bite force parameters. Least significant differences were calculated by Bonferroni's *post-hoc* test. Pearson's correlations were calculated to study the relationships between food material properties, oral processing behaviour and participants' eating capabilities.

5.3. Results and discussion

5.3.1. Fracture properties

The force-distance curves of the three hydrogels upon puncturing with a Volodkevitch bite jaw probe can be seen in **Fracture 5.1**. It shows the typical penetration curves, with the increasing deformation of the sample upon increased applied load up to the point of fracture as the probe penetrates and ruptures the sample. The hydrogel 3*κ*C required an applied force of 8.29 ± 0.96 N, whereas the sample containing alginate beads, 2.4*κ*C0.2CaA$_{300}$, required only half that to rupture (3.67 ± 0.88 N). Interestingly, the sample containing alginate, 1.5*κ*C0.5NaA, was structurally weaker and required a force of an order of magnitude lower than the native *κ*C hydrogel to puncture the gel (0.57 ± 0.14 N). The fracture forces for the three hydrogels were determined to be significantly different ($p < 0.05$).

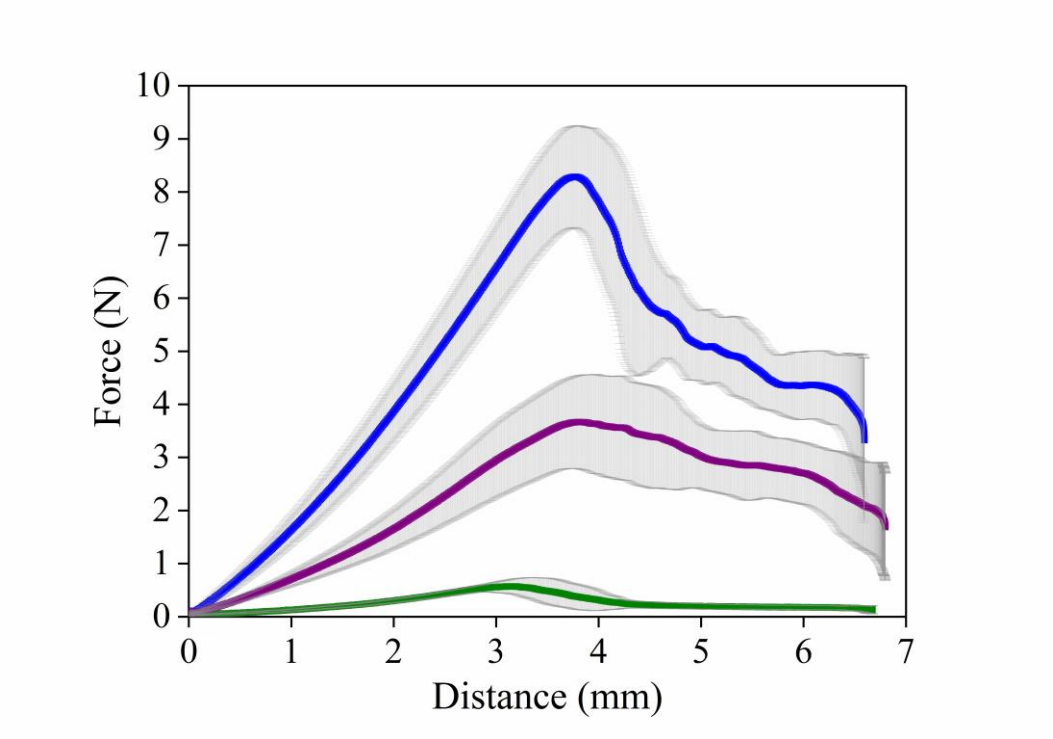

Figure 5.1. Mean (± SD) force over distance curve of the hydrogels obtained from puncture tests with a Volodkevitch probe (first bite), with 3*κ*C (▲), 1.5*κ*C0.5NaA (◆) and 2.4*κ*C0.2CaA$_{300}$ (⬡).

This indicates that *κ*C formed a strong continuous network, whereas the CaA beads disrupted this network indicating they were not inherently connected to the *κ*C matrix. The presence of NaA weakened the *κ*C network even further, causing disruption of the strong *κ*C matrix. This weakening of *κ*C gels in presence of alginates as measured with puncture tests was in agreement with results from the same gels during compression tests (previously studied by (Krop *et al.* 2019) and (Laguna and Sarkar 2016)).

5.3.2. Lubrication properties

Figure 5.2 shows the friction force as a function of entrainment speed for the bolus filtrate of the three hydrogels. From previous studies it was determined that the relevant oral processing speeds, such as the speed of the tongue, ranged from

20-50 mm/s (Krop *et al.* 2019; Steele and Van Lieshout 2009). Therefore, we have focussed here on the mixed lubrication regime. The hydrogel bolus samples were prepared using artificial saliva, and therefore the friction force of the artificial saliva was used as a control.

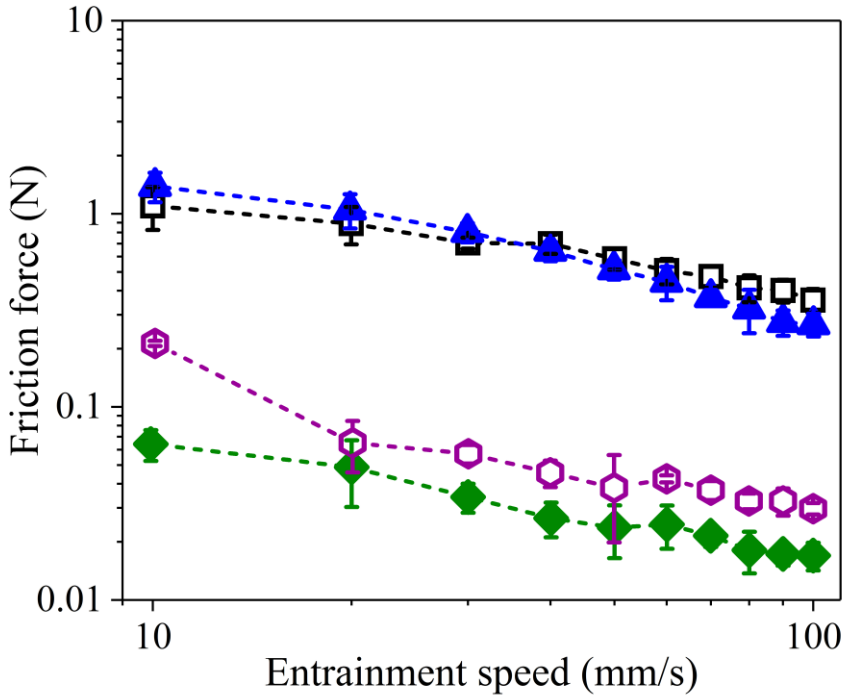

Figure 5.2. Mean friction force (± SD) of 3*κ*C (▲), 1.5*κ*C0.5NaA (◆) and 2.4*κ*C0.2CaA$_{300}$ (⬡) gel bolus filtrates, after simulated oral processing with artificial saliva (□), at 37 °C as a function of entrainment speed in the mixed lubrication regime.

It can be seen that the friction force curve of the simulated bolus filtrate of the 3*κ*C hydrogel is similar to that of artificial saliva ($p > 0.05$, except at μ = 70, 90, 100 mm/s), whereas the 1.5*κ*C0.5NaA and 2.4*κ*C0.2CaA$_{300}$ samples had a significantly lower friction force as compared to artificial saliva ($p < 0.001$). It is worth pointing out here that the larger gel particles were removed by filtration before the oral tribology measurements. The 3*κ*C hydrogel broke down into

significantly larger particles than the other two after simulated oral processing (> 500 μm), and thus were most likely removed during the filtration process resulting in friction forces more similar to those of artificial saliva. Interestingly, the 1.5κC0.5NaA and 2.4κC0.2CaA$_{300}$ hydrogel boli did not have a significant difference in friction force over the measured entrainments speeds in the mixed regime ($p < 0.001$). The reduced friction force of 1.5κC0.5NaA and 2.4κC0.2CaA$_{300}$, compared to 3κC, could be explained by the entrainment of a viscous layer of the alginate-based systems (hydrogel bolus filtrates with artificial saliva) between the PDMS contact surfaces due to the smaller broken down hydrogel particles (Gong and Osada 2001; Krop *et al.* 2019). For 2.4κC0.2CaA$_{300}$, as theoretically predicted by Hertz theory in our previous study (Krop *et al.* 2019), the alginate beads will most likely be deformed during entrainment due to the high pressures generated in the PDMS-PDMS contact zone. Therefore, the lubrication was attributed to the entrainment of the alginate polymer in continuum rather than the intact beads, as well as leaching out of a layer of water from these beads that might act as a surface separator, reducing the friction values.

5.3.3. Eating capability

The EC values of tongue pressure and the different measurement locations of bite force for all participants is shown in **Table 5.1**. The measured tongue pressure values were in line with the results from previous studies on young healthy participants (Alsanei and Chen 2014; Alsanei, Chen and Ding 2015; Laguna *et al.* 2016a).

Table 5.1. Eating capability measures of the 28 included participants.

Gender	Age	Tongue Pressure (kPa)		Bite Force (N) Left Side Molars		Bite Force (N) Front Incisors		Bite Force (N) Right Side Molars	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
Female	24	12.3	1.2	85.1	2.2	79.4	5.7	62.8	1.4
Female	32	10.0	1.0	164.2	17.4	142.3	11.8	124.7	27.8
Female	34	27.7	1.5	89.9	2.2	58.2	1.6	104.0	15.1
Female	22	42.0	3.0	73.8	5.2	62.3	0.0	63.0	19.1
Female	27	47.0	4.4	91.7	10.9	78.9	13.4	91.3	7.6
Male	25	54.3	7.5	106.9	15.8	61.0	4.1	98.4	7.9
Female	26	44.3	3.8	148.9	18.3	108.7	29.5	174.1	20.7
Male	28	61.0	1.0	121.0	4.5	85.5	8.5	105.2	12.5
Female	36	64.0	4.6	83.7	6.4	76.4	12.2	116.0	11.3
Female	31	37.3	7.5	198.7	15.1	151.6	5.3	230.7	7.5
Male	31	57.0	1.7	180.0	7.3	75.9	3.2	126.7	23.9
Female	27	47.3	8.3	196.1	3.5	127.9	10.8	186.4	10.3
Female	25	53.7	6.7	164.9	3.4	138.9	5.1	172.2	9.6
Male	52	63.0	1.7	147.0	5.2	103.9	8.5	143.7	8.3
Male	36	62.7	2.1	165.8	19.8	123.3	8.0	122.1	3.3
Female	25	51.7	7.0	247.8	3.1	110.8	24.6	191.4	44.2
Female	31	59.3	2.3	244.8	14.6	121.4	8.6	117.6	12.6
Female	24	53.0	10.1	198.9	3.1	144.5	7.6	210.0	14.6
Female	29	40.0	3.6	286.4	7.8	154.9	9.3	272.8	13.3
Male	24	56.0	8.5	106.9	1.7	160.5	4.2	256.7	5.3
Female	23	60.7	5.5	207.7	14.4	73.0	19.7	227.5	5.8
Female	23	65.3	2.9	148.8	19.2	132.9	2.5	172.0	12.7
Female	32	60.0	1.0	230.3	45.7	99.9	12.2	206.4	12.0
Male	28	52.3	5.8	237.9	11.8	151.7	27.6	236.8	16.4
Female	26	81.3	2.9	123.4	2.7	79.5	31.8	108.7	25.7
Female	22	52.3	1.5	273.6	4.4	147.6	25.3	268.6	12.9
Male	23	53.7	1.5	297.2	3.3	166.7	17.8	272.6	10.7
Male	33	46.0	2.6	336.0	9.0	284.6	5.7	280.6	16.1
Panel Mean		50.6	15.5	177.1	72.2	117.9	48.1	169.4	68.5

The bite force values were comparable to studies on healthy participants by Fernandes *et al.* (2003) and Laguna *et al.* (2016a) who used a similar measurement device. However, on average the values were slightly higher in the current study, possibly due to the positioning of the test sensors or the interpretation of the instructions by the participants. Interestingly, there was no correlation between tongue pressure and any of the bite force measurements, with $p > 0.1$ (see **Table 5.2a**). This is in line with the study by Laguna *et al.* (2016a) on participants from a similar age group, highlighting that such correlations between oro-facial muscle forces only exist in older adults with limited overall oral capabilities (Laguna *et al.* 2015).

Table 5.2. Correlation matrix of the eating capability measurements (a) and oral processing behaviours (b) for the 28 participants with 3 replicates (n = 84), and significant values indicated in green: $p < 0.01$.

(a)	Tongue pressure	Bite force, left side molars	Bite force, front incisors	Bite force, right side molars
Tongue pressure	1			
Bite force, left side molars	0.07	1		
Bite force, front incisors	-0.08	0.69	1	
Bite force, right side molars	0.11	0.77	0.71	1

(b)	Number of chews	Oral residence time	Chewing rate
Number of chews	1		
Oral residence time	0.94	1	
Chewing rate	0.35	0.07	1

To group the panellists according to their overall eating capability (tongue pressure and bite force), the EC composite scores were calculated using equation 5.1. **Figure 5.3** shows the histogram of the distribution of the EC scores of all participants. Based on the plot, two groups of panellists can be identified on the extreme ends of the plot, with eighteen observations in each group: a low EC group (< 1.0) and a high EC group (> 1.3). The age distribution was similar in both groups, and both groups consisted of male and female participants. The remaining 48 values in the middle had an EC score between 1.0 and 1.3 ($1.0 \leq$ EC score ≤ 1.3). From an eating capability perspective, the participants in this study were however rather homogeneous.

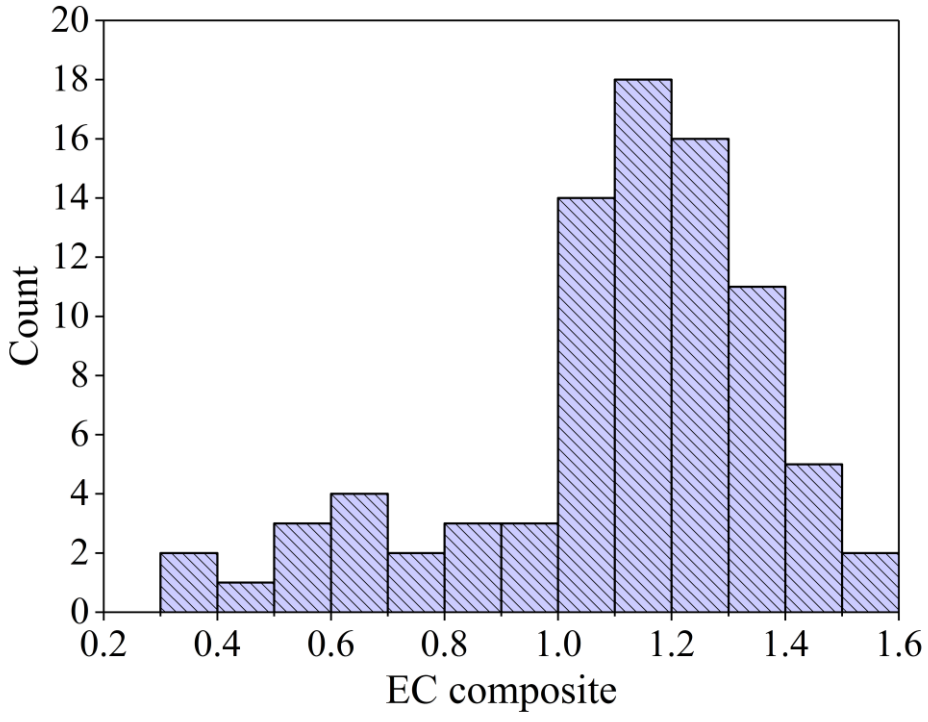

Figure 5.3. Histogram of the eating capability (EC) composite scores of the 28 participants with three replicates each (n = 84).

5.3.4. Oral processing behaviours

Figure 5.4 shows the oral processing characteristics, such as number of chews, oral residence time and chewing rate, that were derived from frame-by-frame video analysis of participants eating the hydrogels. The sample 3*κ*C had a significantly higher number of chews and oral residence time compared to the 1.5*κ*C0.5NaA and 2.4*κ*C0.2CaA$_{300}$ hydrogels ($p < 0.05$). The chewing rate did not show a significant difference between the presented samples, suggesting that chewing rate was subject to individual differences rather than the food material properties (Hiiemae *et al.* 1996).

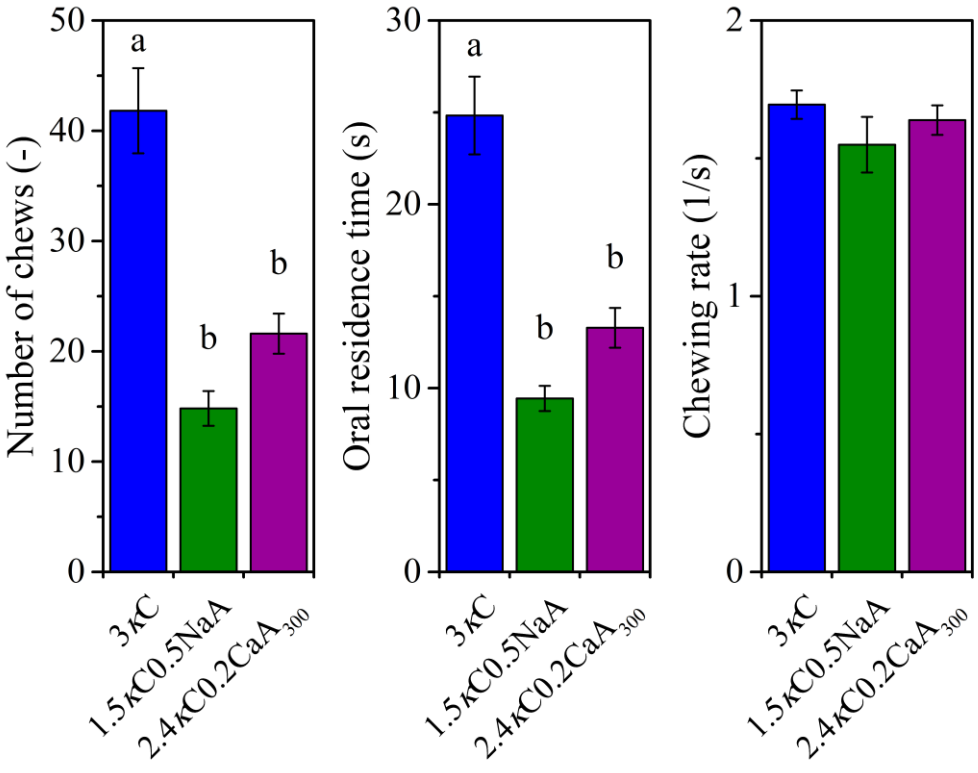

Figure 5.4. Mean values (± SEM) of the oral processing characteristics of the hydrogels obtained from video analysis (n = 28). From left to right: number of chews, oral residence time and chewing rate. Different lower case letters indicate statistically significant differences between conditions ($p < 0.05$).

The correlations between the chewing behaviours were analysed for the combined data set (see **Table 5.2b**). The number of chews and the oral residence time were strongly correlated ($p < 0.001$), meaning that food that is kept in the mouth for a longer amount of time is also chewed more. This was in line with findings from previous studies (Laguna *et al.* 2016a; Engelen, Fontijn-Tekamp and van der Bilt 2005). The number of chews also correlated with chewing rate ($p < 0.001$), but the chewing rate was not related to oral residence time ($p > 0.1$).

5.3.5. Correlations between food material properties, EC parameters and oral processing behaviours

The food material properties and EC parameters were examined for relationships (if any) with the oral processing behaviours to check whether it was the *intrinsic* capability or *extrinsic* food design factors that could best explain the oral processing strategy used for the hydrogels (**Table 5.3a and 5.3b,** respectively). As can be seen in **Table 5.3a**, the puncture force showed a strong correlation with the oral processing characteristics, *i.e.* number of chews and oral residence time ($p < 0.01$). This confirms the data from previous researchers using real food systems, where they made similar conclusions for banana, apple, biscuits (Hiiemae *et al.* 1996) and products like cheese, peanuts and carrots (Engelen, Fontijn-Tekamp and van der Bilt 2005). The friction force measurements showed a good correlation to the number of chews ($p < 0.01$ to $p < 0.05$), and the oral residence time ($p < 0.05$ to $p < 0.1$), depending on the entrainment speed. At the orally relevant speeds, 20-50 mm/s, the correlations seemed to be weaker ($p < 0.01$) than at higher speeds ($p < 0.05$), however, suggesting that slightly higher entrainments speeds might relate better to the number of chews and oral

residence time. Therefore, we propose that for the hydrogel bolus filtrates, number of chews and oral residence time are better explained by frictional properties in the mixed lubrication regime at speeds ≥ 80 mm/s, where the boli form a film separating the two PDMS surfaces (*i.e.* separating tongue and palate during *in vivo* oral processing). Additionally, we do not expect to see any correlations of the friction force in the boundary regime (speeds < 10 mm/s) to the oral residence time due to the absence of any adsorption of the hydrophilic hydrogel bolus particles to the hydrophobic tribo-surfaces (de Vicente, Stokes and Spikes 2006; Sarkar *et al.* 2019; Krop *et al.* 2019). The chewing rate did not correlate with any of the food material properties ($p > 0.1$), suggesting that it is a more inherent property linked to each individual. In addition, it is worth pointing out that where the number of chews and oral residence time showed a strong correlation (see **Table 5.2b**), and are more product specific (**Figure 5.4**), the more inherent chewing rate still increased with the number of chews as indicated by the correlation between the two. This effect was not found for the oral residence time (no correlation to chewing rate), suggesting that where chewing rate and number of chews have a link to the individual, the oral residence time does not and is mostly linked to the type of food structure being consumed.

Table 5.3. Correlation matrix of food material properties related to oral processing behaviour (a) and eating capabilities related to oral processing behaviour (b), with the levels of significance indicated in different shades of green: $p \geq 0.1$, $p < 0.1$, $p < 0.05$ and $p < 0.01$. Since the number of measurements for the food material properties and the oral processing characteristics (a) was not the same, no exact correlation values are displayed but an overall impression of the data is shown based on multiple variations of correlation analyses between the two data sets.

(a)	Number of chews	Oral residence time	Chewing rate
Puncture force			
Friction force 100 mm/s			
Friction force 90 mm/s			
Friction force 80 mm/s			
Friction force 70 mm/s			
Friction force 60 mm/s			
Friction force 50 mm/s			
Friction force 40 mm/s			
Friction force 30 mm/s			
Friction force 20 mm/s			
Friction force 10 mm/s			

(b)	Number of chews	Oral residence time	Chewing rate
Tongue pressure	0.09	0.04	0.02
Bite force left molars	0.13	0.07	0.09
Bite force front incisors	0.14	0.04	0.21
Bite force right molars	0.08	0.00	0.11
EC score	0.15	0.06	0.11

The individual EC measures did not correlate with the oral processing behaviours, nor did the EC composite scores ($p > 0.1$), see **Table 5.3b**. Together, these results suggest that the food material properties dictated the oral processing behaviour of hydrogels with different textural properties in young individuals rather than their individual EC. However, it should be noted that EC was not a limiting factor in the oral processing of the model gels used in these participants. The strength of these model gels was considerably lower than the maximum bite force and tongue pressure measured in current individuals, 8.29 ± 0.96 N or 10.83 ± 1.18 kPa for the hardest hydrogel (3κC) compared to the mean 50.6 ± 15.5 kPa for tongue pressure and 154.8 ± 68.8 N for bite force.

Additionally, the effect of EC level was checked by analysing the correlations between ECs and oral processing behaviours for the selected low EC and high EC groups separately. For the participants with an EC score < 1.0, the bite force for the front incisors and left side molars correlated with the number of chews and oral residence time ($p < 0.05$). On the other hand, the high EC group (score > 1.3) only showed correlations of the chewing rate with the bite force of the front incisors and EC ($p < 0.05$). This would suggest that participants with a low EC score compensated for this by increasing the number of chews and oral residence time, while for people with a higher EC score, and thus a combination of higher maximum bite force and higher maximum tongue pressure, the chewing rate increased.

5.4. Conclusions

Both the *extrinsic* food material properties and the *intrinsic* eating capability of the consumer are hypothesized to have an influence on oral processing behaviour.

Food material properties can be quantified by the use of instrumental as well as sensory techniques. Characteristics of an individual including age, gender and their oro-facial muscular capabilities may also affect oral processing behaviour. In this study, a panel of relatively young, healthy participants, consisting of both males and females, was recruited to investigate the importance of their eating capabilities, such as maximum bite force and tongue pressure, versus the food material properties on the oral processing strategy of three hydrogels with different textural properties and bolus tribology. It was found that the oral processing behaviour was dominated by both the instrumental fracture properties of the hydrogels and the lubrication properties of the hydrogel boli. Whilst the fracture force of the gels and the friction force of the boli in the mixed lubrication regime correlated well with number of chews and oral residence time, they did not relate to the chewing rate. Therefore, we suggest that chewing rate for hydrogels is more subject to individual differences than their physical properties. Interestingly, the number of chews and oral residence time were greater in participants with a low EC compared to high EC score, whereas individuals with a higher EC score had a higher chewing rate. In the future, this study should be replicated with different hydrogels, as well as other types of food to confirm the current findings. Also, it might be interesting to investigate relationships between oral physiological parameters specific to each individual, such as the effects of consumers' habitual salivary flow on oral processing strategy, as well as their preferred oral processing style/chewing type and their favoured type of food materials to eat (Wilson *et al.* 2018).

A key finding from this study was that in healthy, young individuals the differing material properties of the hydrogels were more important determinants

of oral processing than individual differences in eating capability. This then suggests that for hydrogels to influence outcomes such as satiation and satiety, at least in this population, manipulating the physical properties of the gels such as the chewing and oral lubrication properties may produce changes in appetite and food intake across consumers. To test these effects, the hydrogels with different material properties were employed in a preload study design, and results are discussed in **Chapter 6**.

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Chapter 6

The influence of oral lubrication on food intake: a proof-of-concept study[d]

Abstract

As overeating, overweight and obesity remain public health concerns, it is crucial to design satiety-enhancing foods that suppress appetite and lower snack intake. Existing research identifies oro-sensory targets to promote satiation and satiety, yet it remains unclear as to whether it is 'chewing' or 'oral lubrication' that might amplify satiation signals. In this study, techniques from experimental psychology, food material science and mechanical engineering have been combined to develop model foods to investigate the role of chewing and oral lubrication on food intake. Novel model gels, similar in pleasantness, were given as a preload and their effects on subjective appetite and intake of a salty snack were measured in a between-subjects design. Three mint flavoured hydrogels were engineered to vary in their texture (fracture stress) and lubrication (inverse of coefficient of friction), and a control group received mint tea. Results showed that snack intake was suppressed by 32 % after eating the low chewing/high lubricating preload as

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compared to the high chewing/low lubricating preload ($p < 0.05$), but remained unchanged after consuming the medium chewing/high lubricating preload. Hunger ratings decreased from t_1 to t_3 ($p < 0.05$), however differences between conditions were subtle and not significant. Thus, this proof-of-concept study demonstrates that manipulating oral lubrication is a promising new construct to reduce snack intake that merits future research in the oro-sensory satiety domain.

6.1. Introduction

There has been an upsurge in research efforts to design satiation- and satiety-enhancing foods that suppress appetite and prevent overconsumption. Satiation is defined as the processes leading to the termination of an eating event and satiety as the inhibition of appetite and further eating until the next meal, as described within the multifactorial concept of the 'satiety cascade' (Blundell *et al.* 2009). Both satiation and satiety responses contribute to the termination of energy intake, and therefore understanding these processes is important for designing food-based approaches to limit overeating with potential in the longer term to influence weight management (Hetherington *et al.* 2013).

Although the role of oral processing on satiation and satiety has been well established, the quantitative understanding of which dimensions of oral processing influence this has remained elusive (Hetherington and Regan 2011; Krop *et al.* 2018; Lasschuijt *et al.* 2017; Lavin *et al.* 2002). Based on a recent systematic review and meta-analysis on relating oral processing to satiety, it was demonstrated that extending the oro-sensory exposure time to foods leads to a significant reduction in self-reported hunger and food intake (Krop *et al.* 2018). Interestingly, in many, if not most of these satiety trials involving oro-sensory

cues, ‘food rheology’ (*i.e.* liquid versus solid foods, texture/thickness manipulations) has been used as a ‘gold standard’-design tool to influence the number of chews, oral residence time or eating rate, and thus, impact satiety outputs such as appetite ratings (hunger, desire to eat *etc.*), food intake and gut hormonal release (Larsen *et al.* 2016; Lavin *et al.* 2002; Hogenkamp *et al.* 2012). However, during oral exposure, food characteristics change dramatically due to lubrication by saliva as well as the saliva-food mixture that might coat the tongue and other oral surfaces that are of fundamental importance for deglutition and satisfaction (Stokes, Boehm and Baier 2013). Although oral lubrication or friction provided by food is a crucial aspect of this fundamental biological process occurring in the mouth, its’ mechanistic effects on psychological and physiological consequences implicated in altering the motivation to eat remain under-researched (Krop *et al.* 2018).

The present study was designed to address this fundamental knowledge gap using a cross-disciplinary approach. Here we report the effects of novel ‘biopolymeric hydrogel’ preloads on appetite ratings and food intake from both the ‘chewing’ and ‘oral lubrication’ perspective, of which the latter has never been used as a construct in satiety trials. For the purposes of this study, we have focussed only on the external lubrication effects, *i.e.* any lubrication induced by the food material properties and not due to saliva. The selected hydrogels had no energy content and varied in texture in two specific domains: the chewing as well as the lubrication properties. The main objective was to investigate which food design factor between chewing and lubrication might lower snack intake, and whether this is reflected in subjective appetite. The second objective was to study whether the hydrogel preload effects were variable according to eating context

(eating alone or in a group). The first hypothesis to be tested was that greater chewing would result in a lower food intake relative to lower chewing. The second hypothesis was that greater oral lubrication would reduce snack intake relative to lower lubrication. We further predicted that participants in the group setting would eat more snack compared to participants eating alone due to social facilitation, but that the preload effect would occur in both eating contexts.

6.2. Materials and methods

6.2.1. Participants

The study was performed at the University of Leeds, UK. Participants were recruited using a poster campaign around the university campus, departmental recruitment emails and emails sent to a database with people who signed up voluntarily with an interest in participating in human studies. Healthy male and female volunteers were eligible for the study, aged between 18-55 years, without any dental deficiencies or problems with chewing or swallowing, that did not have any food allergies or intolerances to the used study foods and were not taking any medications that might influence appetite or food intake. The experimental protocol of this study was approved by the University of Leeds, School of Psychology Research Ethics Committee (reference number PSC-190) and all participants signed informed consent before their participation. The aim was to recruit 60 participants, 15 in each group, to match previous studies on chewing (see Higgs and Jones (2013)). Participants were not told of the exact aim of the study, instead they were told that the aim of the study was to investigate the effect of a mint stimulus on their perception of a salty snack. Students from the School of Psychology were awarded course credits for their participation, while

other participants were entered into a prize draw with three participants being randomly selected to win a £10 shopping voucher as compensation.

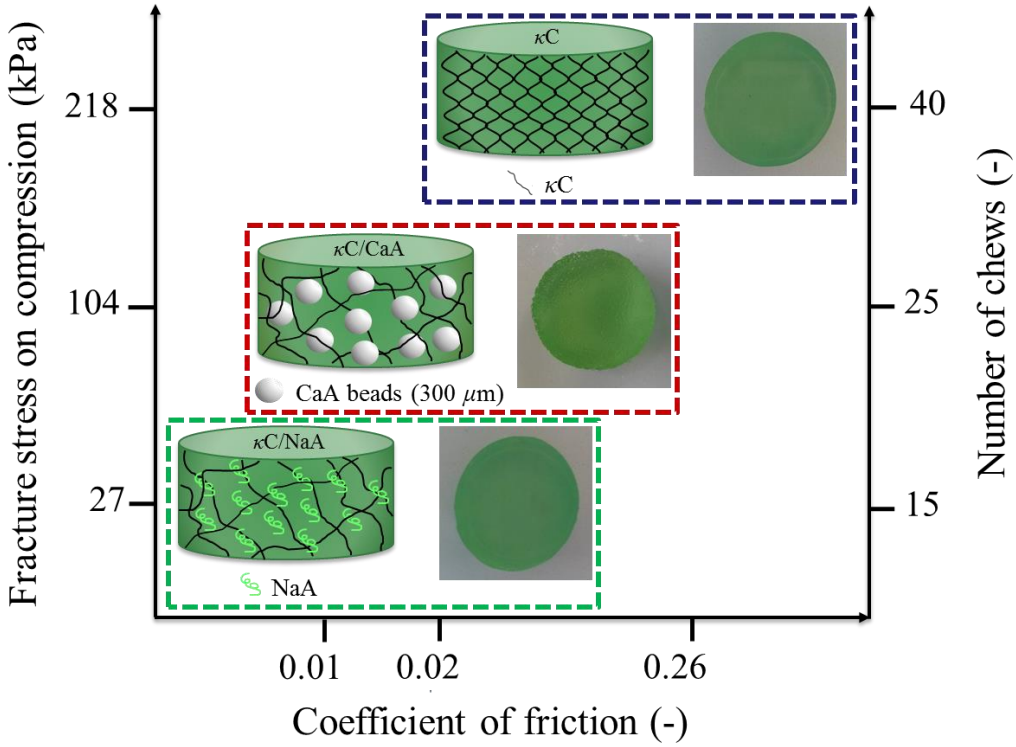

Figure 6.1. Schematic representation of the three different preload gels made up of a *κ*C gel matrix alone or with the addition of sodium alginate (NaA) or calcium alginate beads (CaA) to create distinct chewing and lubrication properties. These properties were based on instrumental characterisation by texture analysis (fracture properties) and tribology (the inverse of the coefficient of friction at 50 mm/s is a measure of oral lubrication at orally relevant speeds), as well as characterisation of the oral processing behaviour (number of chews) using frame-by-frame video analysis.

6.2.1. Experimental design

The study followed a between-subjects design where participants were assigned to one of four conditions. According to availability, participants were allocated to group or alone test sessions and within this randomly assigned to the preload condition. In the different conditions, participants received one of four preload

hydrogels with different chewing and oral lubrication properties (see **Figures 6.1 and 6.3**). To study the effect of social interactions, testing sessions took place either individually or in groups of five to six people.

6.2.2. Study foods

A standardised lunch was given to all participants prior to the start of the study. The lunch consisted of a cheese sandwich, apple, an oatmeal flapjack and *ad libitum* water. The sandwich was prepared using two slices (186 kcal/80 g) of Kingsmill medium sliced 50/50 bread (Allied Bakeries, UK), 12 g (84 kcal) Flora buttery margarine (Unilever, UK) and 32 g (133 kcal) grated British medium cheddar cheese. A Braeburn apple was washed and cut in slices, and 100 g (47 kcal) was weighed out. The sandwich and apple were presented with an individually wrapped flapjack slice (159 kcal/37 g). All products were purchased at a local supermarket. Participants were instructed to consume all the foods provided, containing 609 kcal in total. For the *ad libitum* snack, ready salted crisps (Walkers Snack Foods Ltd., UK) were provided (526 kcal/100 g).

For the preloads, novel mint flavoured hydrogels were selected based on their different chewing and lubrication aspects as characterised in our previous work (Krop *et al.* 2019a), see **Figure 6.1**. The differences in chewing and lubrication were achieved by varying the concentration of different gelling agents, i.e. *κ*-carrageenan (*κ*C) and sodium alginate (NaA), or by introducing calcium alginate beads (CaA) to create textural complexity. The 3*κ*C represents a 3 wt% *κ*-carrageenan hydrogel with high chewing and low oral lubricating properties; 1.5*κ*C0.5NaA represents a mixed 1.5 wt% *κ*-carrageenan and 0.5 wt% Na-alginate hydrogel with low chewing and high lubricating properties; and

2.4κC0.2CaA$_{300}$ denotes 2.4 wt% κ-carrageenan with a layer of 0.2 wt% Ca-alginate beads, 300 μm in diameter, with medium chewing and high lubricating properties (Krop *et al.* 2019a). The hydrogels were presented in bite-size round pieces (diameter 25 mm, height 10 mm) in small, shot-glass type plastic cups. Samples were standardised by volume, and weighed about 5-6 g each (3κC: 5.8 ± 0.4 g, 1.5κC0.5NaA: 5.3 ± 0.3 g and 2.4κC0.2CaA$_{300}$: 5.8 ± 0.3 g). The hydrogels were unsweetened, but flavoured with peppermint aroma and coloured with green food colouring to increase acceptability, and contained less than one kcal. Peppermint tea (Pure Peppermint, Twinings, UK), purchased at a local supermarket and coloured with the same food colouring as the gels, was used as a control preload matching the peppermint flavour and green colouring. The tea was presented in the same cups and filled up to the same height as the hydrogel samples.

6.2.3. Characterisation of the hydrogels

The instrumental properties of the hydrogels were characterised as related to the chewing and the lubrication aspects using texture analysis and tribology, respectively (**Figure 6.1**). The sensory properties were analysed using descriptive analysis (**Figures 6.2a and b**). More in-depth details on the methodology and results have been published elsewhere (Krop *et al.* 2019a).

Uniaxial single compression tests were performed on the hydrogels with a TA-TX2 Texture Analyser Micro Systems Ltd., Surrey, UK) using a cylindrical probe (diameter 59 mm), attached with a 50 kg load cell. The tests were carried out at room temperature at a constant speed of 1 mm/s and the deformation level

was set at 80 % strain. Measurements were performed in triplicate on at least four different preparation days, and mean values of fracture stress were calculated.

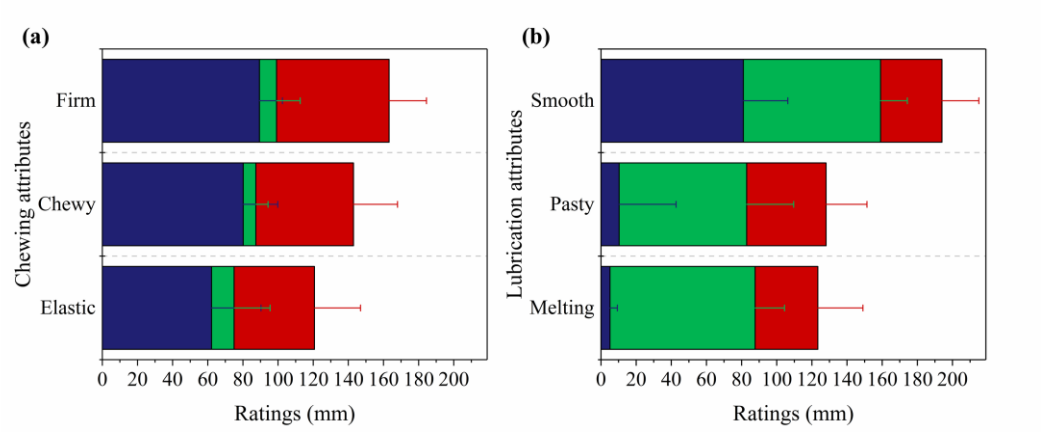

Figure 6.2. The sensory properties (mean ± SD) of the same hydrogels were evaluated using descriptive analysis as related to either chewing (a) or oral lubrication (b), with 3*κ*C (■), 1.5*κ*C0.5NaA (■), 2.4*κ*C0.2CaA$_{300}$ (■). Data was adapted from Krop *et al.* (2019a).

Tribology measurements were performed on the hydrogels after simulated oral processing in presence of artificial saliva using a Mini Traction Machine (MTM2, PCS Instruments, London, UK). The smooth steel surfaces in this device, commonly used in engineering disciplines, were replaced by a polydimethylsiloxane (PDMS) ball and disc set-up at 37 °C to mimic the oral surfaces (surface roughness, R_a < 50 nm) (Laguna *et al.* 2017). The rolling speed was reduced from 1000 to 1 mm/s at a load of 2N, using a slide-to-roll ratio (SRR) of 50 %, and the coefficient of friction in the mixed lubrication regime (50 mm/s) was measured in triplicate.

A panel of 11 participants (4 male, 28.8 ± 5.5 years old) selected sensory attributes for the hydrogels related to chewing and oral lubrication after three training sessions and rated their intensities on a 100 mm visual analogue scale (Krop *et al.* 2019a).

6.2.4. Study procedure

A schematic overview of the timeline and study procedure can be found in **Figure 6.3a**. On the day of testing, participants were instructed to eat their normal breakfast and attend the lab at lunchtime between 12:00-13:00 h. All participants were asked about their age, self-reported body mass index (BMI), health and dietary preferences, and tested for eating restraint using the Dutch Eating Behaviour Questionnaire (DEBQ) (van Strien *et al.* 1986). In addition, participants were provided with one of the novel preloads used in this study (3*κ*C gel), and were asked for their liking and preparedness to eat similar stimuli for the purposes of this study. Then, the standardised lunch was served to control for participants' hunger, and panellists.

Participants were asked to return to the lab 3 h after lunch for the snack, and instructed not to eat or drink anything besides water between sessions. Next, participants completed the pre-preload (t_1) appetite questionnaire, by rating their level of hunger, fullness, desire to eat, appetite, thirst, nausea, desire to eat something sweet and desire to eat something salty on a 100 mm visual analogue scale (VAS), anchored from 'not at all' to 'extremely'. After the appetite ratings, participants were offered the preload stimuli and 50 mL of water. Males received five units and females four to account for the difference in body size, and therefore the oral cavity, between men and women. Participants were instructed to finish the mint stimuli within 10 minutes by consuming the first mint stimulus followed by a sip of water until all mint stimuli and water were consumed. Afterwards, the perceptions of the mint stimulus were evaluated (VAS ratings), followed by another appetite questionnaire (t_2). Then, the participants were

offered a snack of 100 g *ad libitum* ready salted crisps (pre-weighed amount, 526 kcal), as shown in **Figure 6.3a**. To distract the participants form the true nature of the study, they rated their desire to eat and pleasantness of the crisps after a first bite. After that, participants were instructed to eat a normal sized snack, eat as much as they liked within 15 minutes until they felt comfortably full, and to rate their sensory perception of the crisps. Immediately after the snack, participants re-rated appetite (t_3) and answered a final debrief questionnaire, which invited participants to consider the true purpose of the study.

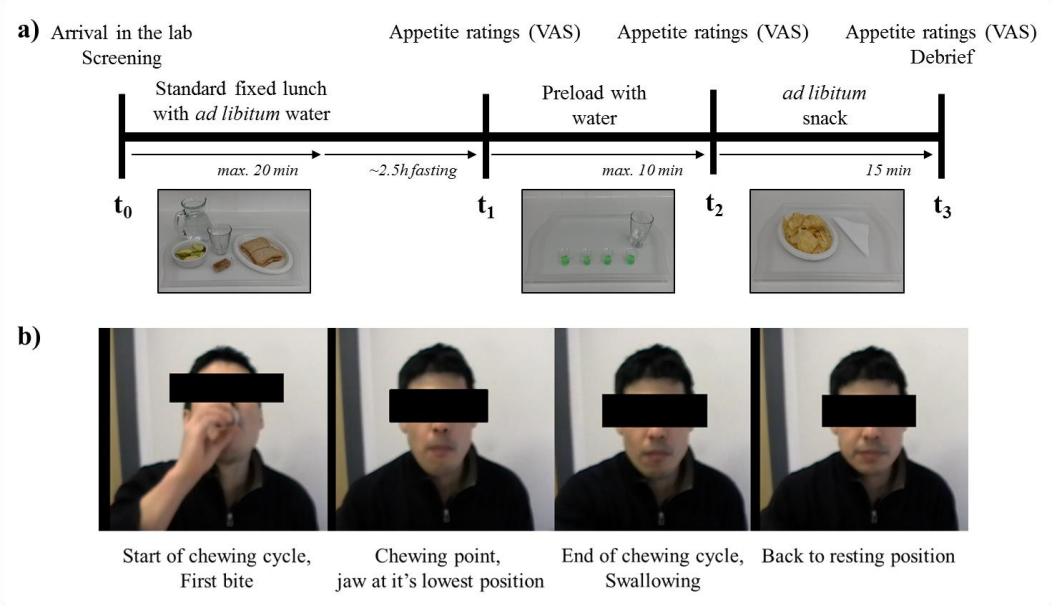

Figure 6.3. Timeline and study procedures of each experimental phase with appetite ratings scored on visual analogue scales (VAS) as a function of time (a). Frame-by-frame video analysis of oral processing behaviour (eating duration from first bite to swallowing and number of chews) (b).

6.2.5. Oral processing characteristics

To analyse the eating behaviour and make sure participants followed the study protocol, a small selection of the participants were asked permission to video record them while eating the preload model food (n = 21). A digital camera

(Panasonic SDR-H90) on a tripod was positioned in front of the participant, and participants were instructed to look straight into the camera while eating the preloads. Videos were analysed using The Observer XT 12 software (v12.5, Noldus Information Technology, The Netherlands). A coding scheme was created to analyse the chewing behaviour, including number of chews (**Figure 6.1**) and eating duration, adapted from previous studies (Laguna *et al.* 2016). A chew was defined as the moment the jaw was at the lowest level during a masticatory cycle (closing action) and eating duration as the time between first bite and swallowing, identified as the first main swallow at the end of the rhythmic rotary chewing movements (**Figure 6.3b**). From these characteristics, the chewing frequency could be calculated by dividing the number of chews by the total eating duration (Forde *et al.* 2013; Laguna *et al.* 2016).

6.2.6. Statistical analysis

All statistical analyses were performed using SPSS (IBM® SPSS® Statistics, v24, SPSS Inc, Chicago, USA). Results are presented as mean ± standard error of mean (SEM), and significance level was set at $p < 0.05$ (2-tailed), unless stated otherwise. Differences between conditions were tested by independent factorial analysis of variance (ANOVA) for food intake and repeated measures to assess condition effects on appetite ratings, followed if appropriate by a *post-hoc* Bonferroni correction for multiple comparisons. Pearson's product moment correlations were calculated to assess the relationship between the different preload conditions and hunger ratings at the three different time points.

6.3. Results

6.3.1. Participants' characteristics

In total, 59 participants completed the study. Before the start of the study, the participants' liking and their preparedness to eat the novel preload foods (3κC gel) were recorded. The mean liking for the test food was 35 ± 23 mm and all participants indicated they were willing to eat the model foods as part of this study. After data collection was completed, four participants were excluded from the analysis due to the following reasons – three participants ate less than 12.5 g of the snack, which is less than half the size of a normal portion, indicating that these participants had not complied with the instruction to eat a normal snack; one participant consumed all of the provided snack, and thus, exhibited the 'cleaning-the-plate' effect suggesting that eating was influenced simply by availability. Thus, the data for 55 participants (16 male, 39 female) were analysed (see **Table 6.1**). Participants ranged in age from 18 to 45 years (mean 26 ± 1 years) and BMI from 18 to 33 (mean 23 ± 0.4 kg/m^2). Eating restraint from the DEBQ showed that three males (> 2.89) and six females (> 3.39) were restrained eaters (van Strien *et al.* 1986), with a mean score of 2.17 ± 0.2 for males and 2.59 ± 0.1 for females.

6.3.1. Effect of oral processing on snack intake

The difference in oral processing between the three preloads, as characterised by video analysis, instrumental analysis (fracture properties and tribology) and sensory panel, is shown in **Figures 6.1 and 6.2**. The 3κC hydrogel showed a high fracture stress (218 kPa) on compression as well as a high number of chews (**Figure 6.1**), indicating that it is a hard gel that has to be chewed more.

Table 6.1. Number of participants in the different preload conditions (in bold) and eating contexts, as well as the mean (± SEM) age and BMI values for the different participant groups.

	Total	*Male*	*Female*	*Age (years)*	*BMI (kg/m^2)*
Hard/Low lubricating (3*κ*C)	**13**	5	8	29 ± 3	22.6 ± 0.5
Individual	8	5	3		
Group	5	0	5		
Soft/High lubricating (1.5*κ*C0.5NaA)	**13**	3	10	23 ± 2	21.8 ± 0.8
Individual	7	2	5		
Group	6	1	5		
Medium/High lubricating (2.4*κ*C0.2CaA$_{300}$)	**15**	3	12	27± 2	22.7 ± 0.9
Individual	10	2	8		
Group	5	1	4		
Control (Mint tea)	**14**	5	9	26 ± 1	25.0 ± 1.0
Individual	9	3	6		
Group	5	2	3		
Total	**55**	**16**	**39**	**26 ± 1**	**23.0 ± 0.4**
Individual	34	12	22		
Group	21	4	17		

The coefficient of friction from the tribology measurements, however, was relatively high (μ = 0.26), indicating that it has low lubricating properties. The opposite was found for the preload 1.5*κ*C0.5NaA, with low fracture stress (27 kPa) and correspondingly low number of chews, and low coefficient of friction (μ = 0.01), indicating it is low chewing and high lubricating. The sensory properties further corroborate this, as the chewing-related attributes, such as ‘firm’, ‘chewy’ and ‘elastic’, were rated high for 3*κ*C and low for 1.5*κ*C0.5NaA (**Figure 2a**). In addition, the 3*κ*C hydrogel scored lower on the lubrication-related

attributes, such as ‘smooth’, ‘pasty’ and ‘melting’, whereas 1.5*κ*C0.5NaA scored higher on the same attributes (**Figure 2b**). The hydrogel with beads, 2.4*κ*C0.2CaA$_{300}$, showed fracture stress (104 kPa), number of chews and chewing attribute ratings between the 3*κ*C and 1.5*κ*C0.5NaA samples, and therefore was characterised as medium chewing. The coefficient of friction of 2.4*κ*C0.2CaA$_{300}$ was similar to that of the 1.5*κ*C0.5NaA hydrogel, and therefore characterised as high lubricating, however 2.4*κ*C0.2CaA$_{300}$ scored rather intermediate on the lubrication-related sensory attributes as well.

The amount of snack eaten was significantly different after the four preload conditions ($p < 0.01$), with snack intake suppressed by 32 % after the soft/high lubricating mint stimulus (1.5*κ*C0.5NaA, 37 ± 3 g) compared to the hard/low lubricating stimulus (3*κ*C, 59 ± 6 g), see **Figure 6.4a**.

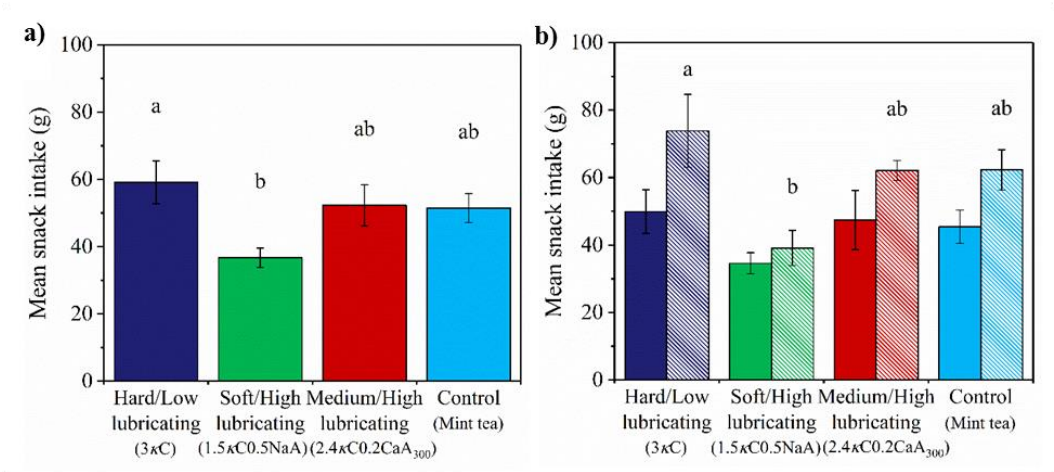

Figure 6.4. Mean (± SEM) snack intake after the four preload conditions (a) and mean (± SEM) snack intake split between the individual sessions (solid fill, n = 34) and group sessions (diagonal lines, n = 21) (b), with 3*κ*C (■), 1.5*κ*C0.5NaA (■), 2.4*κ*C0.2CaA$_{300}$ (■) and mint tea (■). Different lower case letters indicate statistically significant differences between conditions ($p < 0.05$).

The overall snack intake also differed between session types ($p < 0.01$), with the intake in the group sessions being higher (59 ± 4 g) than when eating alone (44 ± 3 g), as was expected due to the social setting. **Figure 6.4b** shows the difference in snack intake between conditions separated by session type (alone or in a group). No interaction effects were found between condition and session type ($p = 0.604$). Also, the effect of gender was analysed but main effects and interactions were not significant, consistent with previous research (Hetherington and Regan 2011). Therefore all subsequent analyses were reported for the group as a whole: male and female, and individual and group sessions together.

6.3.2. Effect of oral processing on subjective appetite ratings

Hunger ratings did not differ by condition, nor was there a significant condition by time interaction. However, the hunger ratings did change over the different time points (t_1 - t_3, see **Figure 6.5a**), with a significant decrease over time ($F(2, 102) = 14.87$, $p < 0.001$). Post-hoc tests revealed that hunger at t_3 was significantly lower than at t_1 and t_2. There was no significant difference between ratings at t_1 and t_2. Similar effects were found for desire to eat ($F(2, 102) = 14.15$, $p < 0.001$) and appetite ($F(2, 102) = 14.34$, $p < 0.001$), see **Figures 6.6a and 6.6b**. The fullness ratings, on the other hand, mirrored those of the hunger, desire to eat and appetite ratings showing a significant time effect ($F(2, 102) = 11.97$, $p < 0.001$), where fullness ratings at t_3 were significantly higher than t_1 and t_2 ratings (see **Figure 6.5b**). There was no significant effect of condition on thirst ratings (**Figure 6.5c**), nor was there any interaction effect of condition by time. However, there was an effect of time alone ($F(2, 96) = 31.62$, $p < 0.001$). Post

hoc tests revealed that t_3 thirst was higher than at t_1 and t_2. Thirst ratings were also lower at t_2 compared to t_1 at the start of the second session.

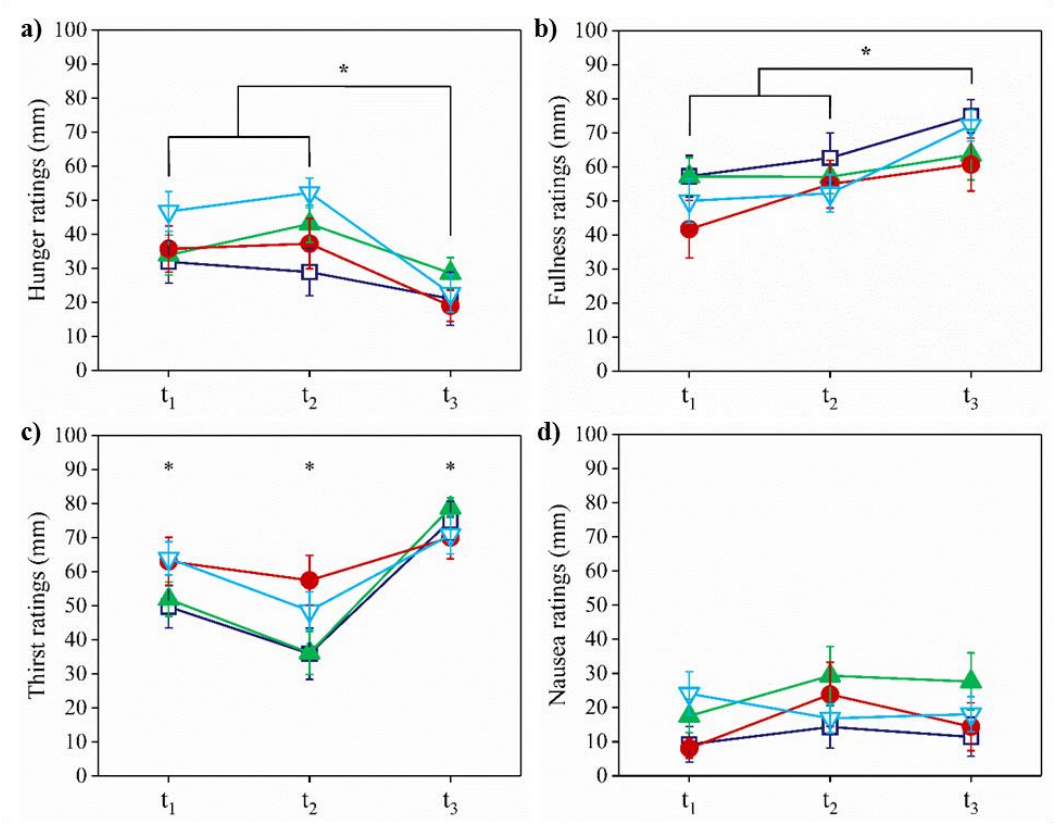

Figure 6.5. Mean (± SEM) hunger (a), fullness (b), thirst (c) and nausea (d) ratings over time for the four preload conditions, with 3*κ*C (□), 1.5*κ*C0.5NaA (▲), 2.4*κ*C0.2CaA$_{300}$ (●) and mint tea (▽), and t_1 before preload, t_2 immediately after preload and t_3 immediately after the snack. Asterisks (*) indicate statistically significant differences ($p < 0.05$).

There were no interaction effects between conditions and time points, and there was no effect of condition on desire to eat something sweet or desire to eat something salty. However, desire to eat something sweet ($F(2, 96) = 4.52$, $p < 0.05$) and desire to eat something salty ($F(2, 96) = 33.28$, $p < 0.001$) did significantly change over time (**Figures 6.6c and 6.6d**). To make sure none of the preloads invoked a stronger feeling of nausea, due to the novelty of the model foods or the presence of the hydrocolloids in the preloads, nausea was rated over

time as well (**Figure 6.5d**). There was no significant main effect of preload condition or time point, nor was there any interaction effect of condition vs time.

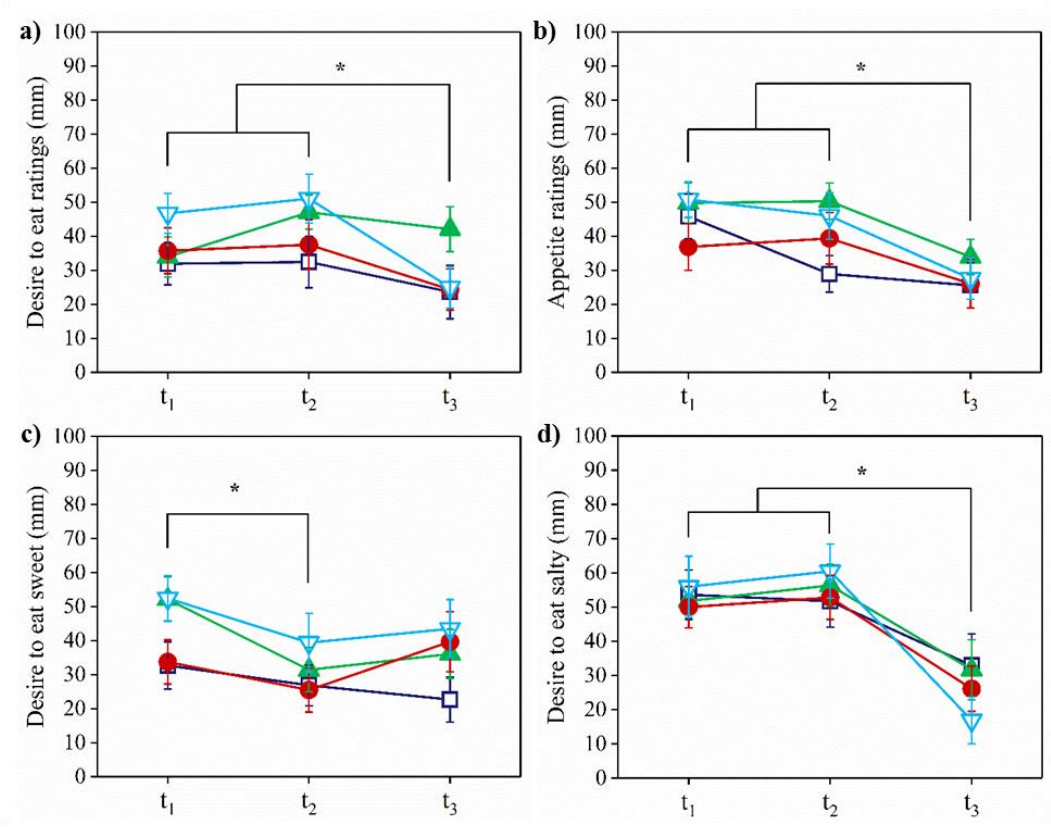

Figure 6.6. Mean (± SEM) desire to eat (a), appetite (b), desire to eat something sweet (c) and desire to eat something salty (d) ratings over time for the four preload conditions, with 3*κ*C (□), 1.5*κ*C0.5NaA (▲), 2.4*κ*C0.2CaA$_{300}$ (●) and mint tea (▽), and t_1 before preload, t_2 immediately after preload and t_3 immediately after the snack. Asterisks (*) indicate statistically significant differences ($p < 0.05$).

6.3.3. Perception of the study foods

The pleasantness, strength of the mint flavour, sweetness or chewiness of the preload foods were rated on 100 mm VAS. One-way ANOVA indicated that pleasantness, mint flavour and sweetness did not differ between the preload conditions. However, the chewiness of the preload samples was significantly different ($F(3, 51) = 31.30$, $p < 0.001$). The post hoc test indicated that the mint

tea (control sample) was not perceived as chewy at all (mean 3 ± 2 mm), the 1.5κC0.5NaA was significantly more chewy (mean 37 ± 8 mm) than the mint tea, and 3κC (mean 77 ± 7 mm) and 2.4κC0.2CaA$_{300}$ (mean 67 ± 6 mm) were the most chewy.

6.3.4. Correlations

Pearson correlations between chewiness of the preload and the snack intake showed that they were not related (r = 0.056, *p = 0.687*). Food intake between the different preloads also did not correlate with the perceived pleasantness (r = -0.132, *p = 0.338*) or any potentially induced nausea after eating the preload foods (r = -0.189, *p = 0.168*).

6.4. Discussion

The present study investigated whether model hydrogels with varying chewing and oral lubrication properties had a significant influence on self-reported appetite measures, such as hunger, fullness and desire to eat, as well as the intake of a snack. It was hypothesized that more chewing would lead to lower food intake, as reported in previous studies (Krop *et al.* 2018). Interestingly, results showed that snack intake was only lowered after the consumption of the soft/high lubricating preload sample (1.5κC0.5NaA) compared to the hard/low lubricating preload (3κC), suggesting that it was not the chewing but the lubricating properties that governed subsequent intake of a salty snack. Sensory ratings for the different preloads did not reveal a significant difference in terms of pleasantness, strength of mint flavour or sweetness, and therefore these characteristics could not account for the suppressed food intake after the soft/high lubricating preload (1.5κC0.5NaA). Nevertheless, there was no significant

difference between the soft/high lubricating preload (1.5*κ*C0.5NaA) and medium/high lubricating preload (2.4*κ*C0.2CaA$_{300}$). Therefore, it seems unlikely that the instrumental measure of coefficient of friction alone can explain the mechanism of lubrication behind the lower snack intake after the soft/high lubricating preload (1.5*κ*C0.5NaA). Previous research analysing the hydrogel preloads using a sensory panel found that the soft/high lubricating preload (1.5*κ*C0.5NaA) was rated more 'smooth, 'pasty' and 'melting' compared to the medium/high lubricating preload (2.4*κ*C0.2CaA$_{300}$), see **Figure 6.2b**, though they were rated similarly for other indices of lubrication (Krop *et al.* 2019a). In particular, this pastiness was defined as 'a sensation of the presence of wet/soft (immiscible) solids in the mouth', which could result in a certain amount of mouth coating. Such mouth-coating aspects of sodium alginate (1.5*κ*C0.5NaA) have previously been reported as related to a mouth moistening and hydrating property (Cook *et al.* 2017), which in turn might lead to a lower snack intake. To make sure this mouth-coating did not lead to any lingering feelings of nausea, the nausea ratings were analysed and no significant differences in subjective nausea were found between conditions after the consumption of any of the preloads.

Besides the difference in snack intake between 1.5*κ*C0.5NaA and 3*κ*C, snack intake after the hard/low lubricating (3*κ*C) and the control sample (no chewing/low lubricating) did not show a significant difference, indicating that it was not the chewing properties that determined snack intake after the preload. This does not support previous research, which showed that a higher level of chewing did indeed reduce food intake (Lasschuijt *et al.* 2017; Lavin *et al.* 2002; Krop *et al.* 2018). This might be explained by the short exposure time of 10 minutes and the low amount of elicited chewing in this period, indicating that the

total chewing time may not have been sufficiently long enough to influence food intake. Future research incorporating more hydrogel pieces into the preload to increase overall chewing time may find a more pronounced effect on food intake. However, there are other studies that confirm no impact of chewing on food intake (Julis and Mattes 2007).

In addition, it was found that snack intake was greater in a group setting compared to eating alone, confirming the hypothesis that social interactions during a snack increases food intake (Redd and de Castro 1992).

The effect size was considered relatively small, which is also consistent with previous research investigating oro-sensory stimulation (Hetherington and Regan 2011). This may be related to a small effect of chewing or lubrication during oral processing, or the amount of preload gels (four or five units per participant) was rather small. The novelty of the preload hydrogels and their generally low rated pleasantness were a consideration in providing a limited amount of the preload foods (Pliner and Hobden 1992), as well as not wanting to prevent any further food intake due to the volume of the preload. The present study also found that preload foods with varying chewing and oral lubrication properties did not significantly influence self-reported appetite measures, such as hunger, fullness and desire to eat, indicating that one preload did not lead to higher or lower self-reported appetite ratings than any of the other preloads. In addition, a decrease in hunger, desire to eat, appetite and desire to eat something salty, and an increase in fullness ratings were observed over time in the following snack intake in all preload conditions. Thus, this confirmed that the participants consumed the snack until satiety was reached.

6.5. Limitations

Limitations of the study include the lack of a fully factorial design with model foods representing hard/high lubricating and soft/low lubricating properties. In a future replication study, hydrogels with these qualities could be developed to improve the matrix for comparisons. In addition, the *in-vivo* oral lubrication effects of the preloads, *i.e.* the lubrication contributed by the bio-lubricant saliva (internal) versus hydrogels (external), were not checked whereas the chewing properties were measured by video analysis of the chewing behaviour. Furthermore, the sample size was smaller than planned and this limits extrapolation from this study; future studies should use a larger sample. Also, the use of ready salted crisps as a salty snack in the current study may have influenced the results. Liking for the crisps may have overshadowed the chewing and oral lubrication effects of the preloads. A larger effect may have been found had we included a sweet snack since intake is influenced by individual food preferences. On the other hand, increasing variety by providing both salty and sweet snacks might stimulate appetite and increase intake (Rolls *et al.* 1981), and overpower any effects due the preloads.

An independent between-subjects design was used in the present study to facilitate easier panel recruitment and flexibility. Better results might have been obtained with a within-subjects design where the random noise would be minimized (Stone and Sidel 2004). However, a within-subjects design would have resulted in increased familiarity with the preload hydrogels, and would be associated with increased expected satiation and satiety.

6.6. Conclusions

The aim of this study was to investigate whether chewing and lubrication during oral processing, manipulated by hydrogel preloads, had an influence on snack intake and self-reported appetite ratings. Results from this proof-of-concept study demonstrated that snack intake was reduced following the soft/high lubricating preload relative to the hard/low lubricating preload, which was not predicted. The mechanism by which oral lubrication, rather than chewing, played a prominent role in reducing subsequent food intake of a salty snack, was associated with the sensory 'mouth-coating' aspects of the preload; however, exact biological cross-talk between mouth-coating, tactile perception and mechano-receptor stimulated satiation, as well as the role of food material-saliva interactions in both satiation and satiety, demand systematic future studies.

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Chapter 7

General discussion[e]

7.1. Summary of the main thesis results

Understanding the interface between the physical properties of foods, the intrinsic features of the consumer and how consumers respond to oro-sensory manipulations both advances knowledge in the field and leads to potential strategies for appetite control. This is especially urgent and relevant given the current obesogenic environment, where an increasingly sedentary lifestyle is matched by an excessive intake of food leading to a positive energy balance. The broad aim of this thesis was to explore the domain oral processing using different food structures with the potential for appetite control and reduced food intake. If successful, these studies may lead to the development of satiety-enhancing foods to address the dual concerns of excess food intake and risk of obesity. An overview of the chapters in this thesis and their outcomes is shown in **Figure 7.1**, from the development of the model foods, their physical properties and sensory perception to the use of these foods with consumers differing in eating capability and ultimately their use in a short-term satiety study.

[e] Part of this chapter is published as Sarkar, A. and E. M. Krop. 2019. Marrying oral tribology to sensory perception: a systematic review. *Current Opinion in Food Science,* **27**, pp.1-10.

Based on the perceived knowledge gap in the literature on the effects of all aspects of oral processing on satiety (**Chapter 2**), this PhD project has advanced the knowledge base by moving beyond the chewing-related aspects of oral processing to external lubrication. In order to test both the effects of chewing and oral lubrication, a set of model food systems were developed and their properties characterised instrumentally and sensorially (**Chapters 3 and 4**). Using the mechanical engineering technique of tribology, it was hypothesised that the lubrication processes in the mouth can be quantified and potentially linked to the sensory perception of lubrication-related attributes using descriptive sensory analysis. The strategy to test this hypothesis involved the engineering of different hydrocolloid systems, with a range of concentrations and ratios, and by tuning the interactions between the structural elements so that they affect the oral processing behaviour (**Chapter 3**). The hypothesis was supported and the hydrogels with high chewing and lubricating properties were identified (**Chapter 4**). It was demonstrated that for young adults, it was not the person's eating capabilities but the food's material properties that influenced the oral processing behaviour, such as number of chews (**Chapter 5**). Then, using the identified model hydrogels from **Chapters 4 and 5** as preload food systems, the effects on food intake and appetite ratings were measured (**Chapter 6**). New insights were generated on the reduction of food intake by the lubricating gels rather than the chewy gels.

This discussion chapter reflects on the novelty of the key findings obtained in each chapter of this thesis and considers the major study parameters. Finally, the implications of the current findings are discussed and recommendations for future work are made.

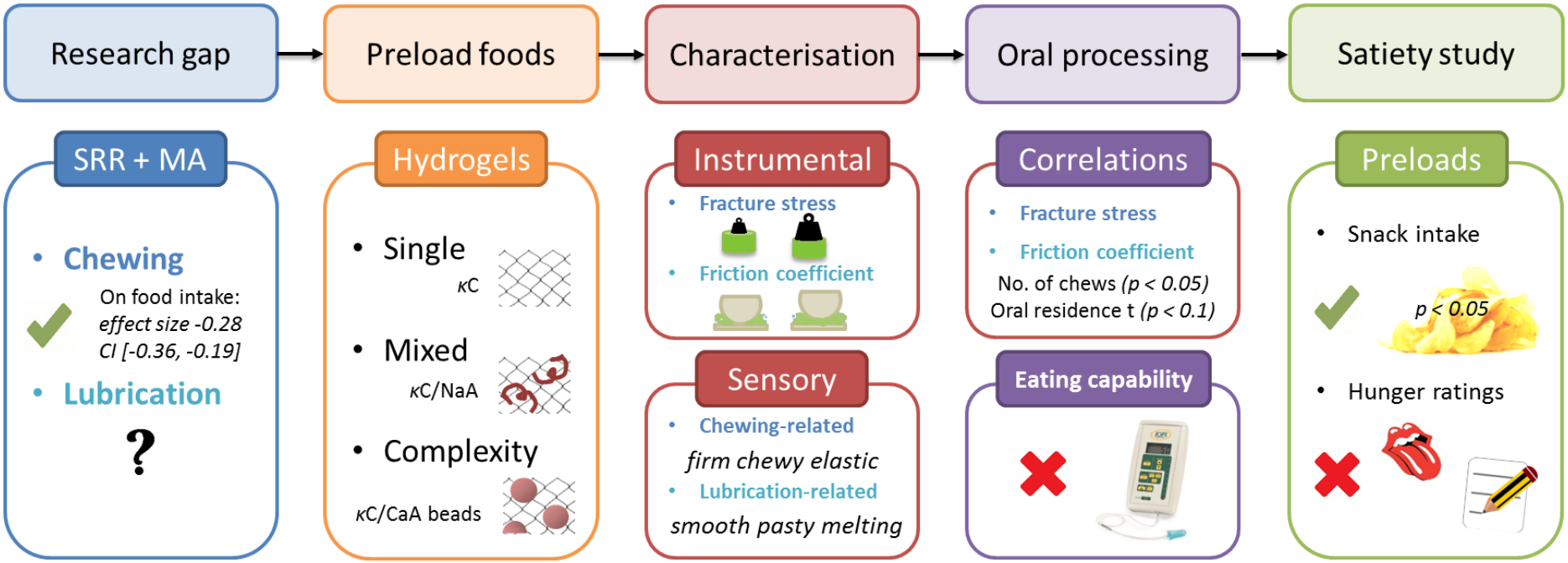

Figure 7.1. Thesis summary: linking model foods' instrumental, sensory and oral processing properties to food intake, focussing on chewing and external lubrication using hydrogel preload foods.

7.2. Novelty of this thesis

In **Chapter 2**, a systematic review and meta-analysis on the influence of oral processing on satiation and satiety revealed a clear knowledge gap in the literature. For the first time, key words related to oral lubrication, friction and tribology were used as part of the oral processing search terms, and linked to appetite ratings and food intake measures. Little to no literature was found investigating the effects of oral lubrication as an integral part of oral processing, using a standardised test-design to allow for comparisons between studies.

In **Chapter 3**, a wide range of hydrogels with different hydrocolloids, concentrations and mixing ratios were prepared and the fracture properties were analysed instrumentally. Previous studies on model gels focussed mainly on emulsion-filled gels, where the focus was on the fat-related properties, *e.g.* Devezeaux de Lavergne *et al.* (2016) and Liu *et al.* (2015). In this study, no fat was added to the model systems to study the instrumental and sensory properties, which required the development of new methodologies to measure lubrication-related aspects of oral processing in non-fat food products. Based on the fracture properties of these hydrogels and their potential suitability for human consumption, a selection was made. In **Chapter 4**, the instrumental friction properties (tribology) and sensory lubrication attributes were determined for this selection of hydrogels. By correlating the various properties, it was found that instrumentally determined friction coefficients could be used in order to predict sensory perception, specifically in non-fat hydrogels. In addition, it was found that creating simulated bolus samples by breaking down the hydrogels in the presence of artificial saliva, provided realistic samples that could then be compared to the findings from sensory

analyses. More importantly, the method development involving gel bolus fragment samples and gel bolus filtrates highlighted the importance of the thin-film aspects in tribology, and indeed the instrumental tribological behaviour of gel bolus filtrates were correlated with lubrication-related sensory attributes.

An important aspect of this thesis was to design the instrumental properties of the hydrogels in such a way that would result in different oral processing strategies in the same person. However, different individuals can have widely varying oral capabilities, *i.e.* the size of the oral cavity and maximum bite force. Therefore, we wanted to make sure any differences in the oral processing strategy we measured was due to the different hydrogels and not due to the individual eating capabilities (ECs) of the consumer. In **Chapter 5**, we showed the importance of the food material properties over individuals' ECs in young adults, where the differences in EC did not indicate any correlation to the number of chews, the oral residence time or eating rate.

Finally, using a proof-of-concept study we demonstrated the effect of oral lubrication on short-term satiation (snack intake), as well as the importance of considering lubrication as an aspect of oral processing for food perception and satiety development (**Chapter 6**), which had not been considered before.

7.3. Discussion points and thesis' implications

7.3.1. Considerations of the preload foods: hydrogels

This thesis concerns model hydrogels based on different hydrocolloid systems, such as *κ*-carrageenan (*κ*C), locust bean gum (LBG), sodium alginate (NaA) and calcium alginate (CaA) beads. These hydrocolloids were selected based on their ability to form stable, non-thermo-reversible gels at body temperatures of 37 °C

that were suitable for human consumption (**Chapter 3**). Aside from the selected hydrocolloid systems, many more polysaccharide options exist, such as agar, gelatine, gellan, pectin, xanthan gum, *etc*, as well as proteins such as whey protein, that can form gels. By creating systems with different concentrations and mixtures, different textural and tribological properties might be obtained. Certain hydrocolloids were not considered further in this study, as they are known to possess certain characteristics unsuitable for the purposes in this thesis, *e.g.* gelatine which shows melting behaviour at orally relevant temperatures. This melting behaviour would further complicate the sensory experience, particularly when comparisons are made with systems that do not melt at body temperatures (Devezeaux de Lavergne *et al.* 2016). Based on a series of preliminary studies, other hydrocolloid systems were excluded as they did not form stable gels that would be pleasant to eat. Pectin gels for example showed a brown discolouration, especially at higher concentrations (see **Figure 7.2**), which might be difficult to conceal from the panellists, and xanthan gum was unable to form stable gels convenient for handling. For this thesis, *κ*C was selected for its suitability and LBG and NaA were used to change structural properties of the *κ*C matrix. As this was the first study to look at both the instrumental and sensory lubrication characteristics of solid hydrogel systems, the expected properties were unclear at this early stage.

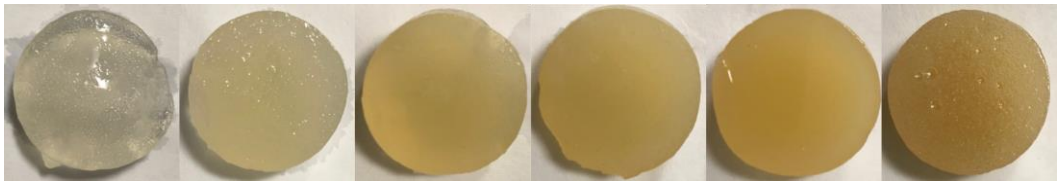

Figure 7.2. Images of pectin hydrogels (concentration range 1.0-6.0 wt%).

In addition, the κC gels were layered with CaA beads of two different sizes (300 and 1000 μm), with the aim to achieve a level of inhomogeneity resulting in a different lubrication perception in the mouth. However, from the sensory descriptive analysis (**Chapter 4**), it was found that the CaA beads were not necessarily noticed during consumption. Incorporating the beads directly into the κC network, instead of as a separate layer, would potentially improve the sensory experience. Different CaA concentrations and bead sizes were not explored further. Instead of larger particles, which we expected would have increased lubrication properties due to particles bursting and releasing liquid during oral processing, smaller particles might be able to create the so-called "rolling" effect with improved perceived smoothness.

7.3.2. *In vitro* compared to *in vivo* oral tribology

The surfaces in the mouth relevant for oral lubrication measurements consist of the tongue, palate, teeth and mucosa (Sarkar *et al.* 2019). To measure the oral lubrication processes instrumentally, it is important to use a set-up that closely mimics the actual oral surfaces. Although an attempt was made to change the hydrophobic surface properties of the PDMS tribopairs by coating them in artificial saliva before analysing the bolus hydrogel properties (**Chapter 4**), other studies have shown the ineffectiveness of this procedure considering the high contact angle of PDMS (108°), where almost any saliva might just slip off rather than adsorb (Sarkar *et al.* 2017). Therefore, the PDMS tribopairs might not have been the best representatives of the hydrophilic oral surfaces, making it more difficult to find any potential correlations with the sensory lubrication attributes. Although PDMS is commonly considered a gold-standard for tribological testing in food science, the Young's modulus of smooth PDMS surfaces is actually two orders of magnitude

higher than that of the human tongue (Dresselhuis *et al.* 2008a). Using the Hertz contact theory, the contact pressure during testing can be determined, which using PDMS surfaces is one order of magnitude lower than the pressure measured in healthy adults (Sarkar *et al.* 2019). In order to better mimic the oral conditions, lower loads (below 0.1 N) or materials with lower Young's modulus should be considered. In addition, the tongue surface is not smooth, but consists of numerous filiform papillae, mainly at the front of the mouth, as well as larger mushroom shaped fungiform and other papillae that trigger taste perceptions (Sarkar *et al.* 2019). The rather smooth PDMS surfaces are thus quite dissimilar from the surface of the human tongue. Future studies might benefit from improved tribological surfaces before any more definitive correlations to sensory perceptions can be made.

In this thesis, the sensory texture attributes were targeted for their connection to chewing and oral lubrication. During the first phase of oral processing, the focus is more on the chewing related texture attributes, such as chewiness, hardness, brittleness and elasticity. In the later phases, the sensory attributes are more influenced by the rheological and tribological properties. After swallowing, some food residues can remain in the mouth adhered to the oral surfaces forming an oral coating. These food residues are often perceived as fatty, creamy and smooth, or dry, rough and gritty, summarised as after-feel attributes. Moreover, these after-feel attributes are better described by the tribological properties of the food residues and their interactions with saliva and the oral surfaces, rather than the rheological bulk properties of the food (Prakash, Tan and Chen 2013). Since the hydrogels used in this thesis did not contain any fat, the after-feel properties were considered less relevant at the start of the project.

However, the ‘pasty’ properties revealed in the hydrogels containing NaA were theorised to be a result of their mouth-coating capabilities (**Chapter 4**), showing the importance of dynamic sensory evaluations.

Besides the instrumental tribology measurements to determine oral friction, it is important to study the link with *in vivo* oral lubrication processes. In this thesis, the chewing behaviour for the different hydrogels was analysed using video recordings and frame-by-frame analysis of targeted chewing behaviours, such as number of chews and chewing duration. As the in-mouth friction behaviour cannot be visually studied outside of the mouth, this poses a research challenge that still needs to be tackled. Previous studies have used *in vivo* fluorescence methods to quantify fatty deposit layers on the tongue (Camacho *et al.* 2014; Camacho *et al.* 2015), however, due to the absence of fat in the hydrogels this method was less suitable for use in this thesis. Alternatively, the mouth-coating properties of non-fat products could be determined from the protein content on the oral surfaces. Due to the food-saliva interactions, the salivary film usually present in the mouth may be broken down at various rates causing differences in sensory perception (Selway and Stokes 2013). By measuring the mucin content of the salivary film at different stages of oral processing, it may be possible to link the *in-vivo* mouth-coating properties to the sensory lubrication attributes. Other researchers have focussed on a classification system for individuals that show a preference for a specific eating style, *i.e.* ‘chewers’, ‘crunchers’, ‘smooshers’ and ‘suckers’ (Wilson *et al.* 2018). This classification system shows a potential application in further lubrication studies, where the eating style might indicate something about the friction behaviour of certain foods or a participants’ preference for foods with certain lubrication properties. Using more advanced techniques, such as tracking of the

jaw's movements using a 3D electromagnetic system (Wilson *et al.* 2016), chewing behaviour might be analysed in more detail which might be linked to their eating behaviour and also the oral lubrication properties. Overall, little is known about how to analyse the mouth-coating properties of non-fat products. Therefore, there is a need to develop new methodologies to measure the *in vivo* mouth-coating properties, and standardise them for both fat and non-fat food products.

7.3.3. Correlations between tribology and sensory perception

Quantitative relationships between μ at particular entrainment speeds and specific sensory attributes evaluated by panellists have attracted significant research attention in both model foods (emulsions, emulsion gels and hydrogels) and real foods (milk, yoghurts, custards, cream cheese, chocolate and bread). From a systematic review into published literature, 38 studies were identified correlating instrumental tribology data and sensory attributes (see **Table 7.1**). Most previous studies looking at the correlations between tribology and sensory perception have concentrated on model foods or real food products with a fatty component, resulting in a focus on attributes such as 'creamy', 'fatty' and 'smooth'. However, these attributes were found to be less relevant in products not containing any fat.

In this thesis, we have shown for the first time correlations between instrumental tribology measurements and lubrication-related sensory attributes in aqueous hydrogels, not containing any fat (**Chapter 4**). The coefficient of friction (μ) in the mixed lubrication regime (50 mm/s) of the hydrogel bolus filtrate (*i.e.* gel particles > 500 μm were filtered out after simulated oral processing in presence of artificial saliva) revealed an inverse correlation with 'pasty' (R^2 = -0.80, $p < 0.05$) and a positive correlation with 'slippery' (R^2 = 0.82, $p < 0.05$) and 'salivating'

($R^2 = 0.79$, $p < 0.05$). A study by Malone, Appelqvist and Norton (2003) on hydrocolloid solutions also found a positive correlation with 'slippery' in the mixed regime (10-100 mm/s), similar to our result on solid hydrogels. However, another study on hydrocolloid-protein gel particle dispersions, without added fat, found the opposite with an inverse correlation to 'slippery' ($R^2 = -0.40$, $p < 0.05$) in the mixed regime (50 mm/s) (Chojnicka-Paszun, Doussinault and de Jongh 2014).

The direction of the correlations between μ and sensory attributes in hydrogels might be counterintuitive when comparing against those in fat-based emulsion gels. For the hydrogels, 'pastiness' was linked to the mouth-coating aspects of the samples, *i.e.* the coating of the bolus filtrates was viscous enough to separate the oral surfaces and thus μ was reduced in the hydrogels that were evaluated to be highly 'pasty'. On the other hand, 'slipperiness' was defined as the ease of sliding through the mouth during oral processing, indicating that highly slippery hydrogels were easily sliding past the oral surfaces. This resulted in the hydrogel boli not being retained within the contact surfaces, with as a consequence higher μ values. The inverse correlation found in the study by Chojnicka-Paszun, Doussinault and de Jongh (2014) seemed to be a result of the added protein gel particles. Due to the added complexity, the expected positive correlation between μ and 'slippery' was not found. Likely, the perception of 'slippery' here is influenced by some other property of the dispersion, such as the film-forming capacity, and would suggest it might behave more like foods with a fatty-aspect.

Figure 7.3 shows a schematic representation of the more recent studies in **Table 7.1**, summarizing possible existing correlations in different test foods (both model and real food systems) between lubrication-related sensory attributes as well as other relevant instrumental parameters, such as friction coefficient, viscosity and

particle size. It can be seen that three clusters were identified: 1) foods containing fat, 2) no- to low-fat containing foods, and 3) a variety of different solid model and real foods. For clusters 1 and 2, the relevant sensory attributes were 'smooth', 'creamy', 'viscous', 'astringent' and 'grainy' in the fat-related foods, whereas for the solid foods different descriptors were used. The exception being the fat-containing emulsion-filled gels, which showed some overlap with attributes found mainly in cluster 1.

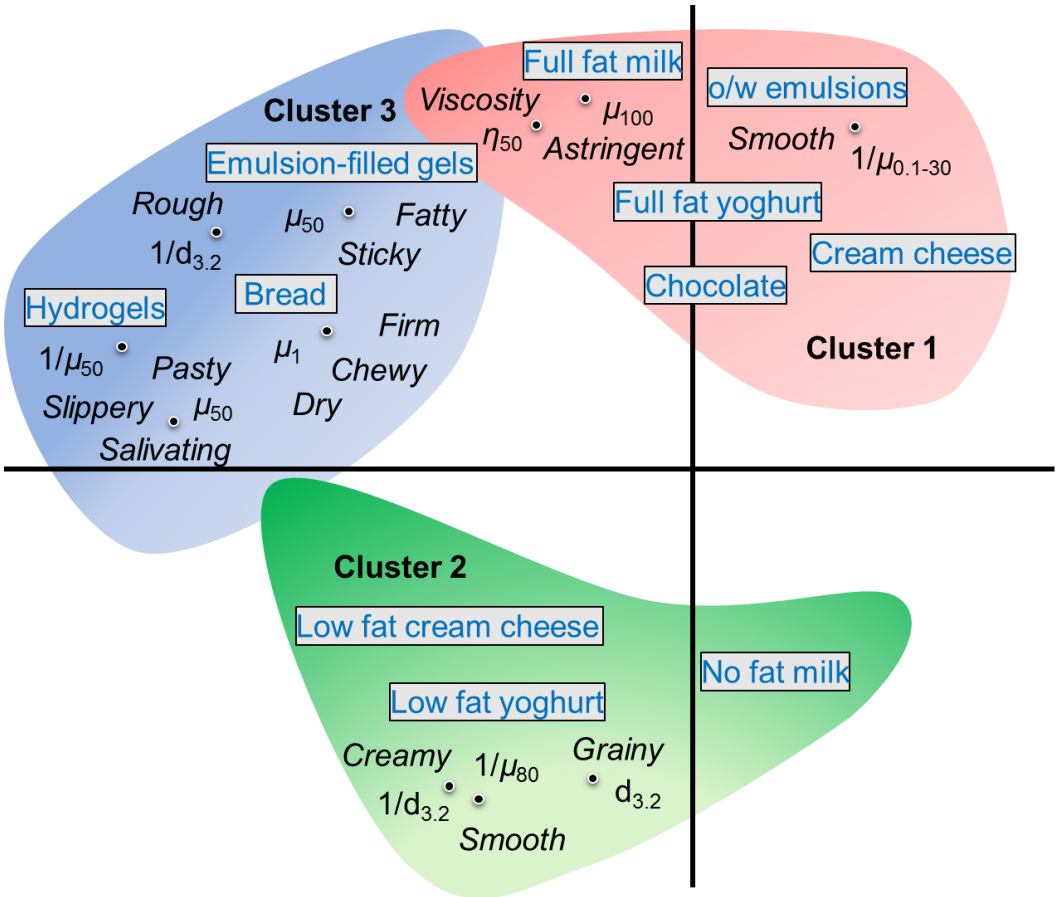

Figure 7.3. Schematic representation of qualitative clusters on correlations between instrumental and sensory parameters for different model and real food products, based on the studies published in or after 2016 reported in Supplementary Table E.1. Here μ, η, $d_{3,2}$ represent the friction coefficient, viscosity and mean particle size. The subscripts for μ and η are the speed (mm/s) and shear rate (s^{-1}), respectively.

Table 7.1. Studies that have examined the relationship between tribology and sensory properties.

		Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
Model foods	Emulsions	Hydrocolloid solutions and o/w emulsions (guar gum)	MTM (1-1100 mm/s, 3 N, steel ball on silicon disc set-up)	Artificial saliva (ions, mucin), 35 °C	Ratings compared to references	Pearson's correlations	Slippery [hydrocolloid solutions], creamy (-) [emulsions]	Mixed (10-100 mm/s)	Malone, Appelqvist and Norton (2003)
		Emulsions varying in type and amount of emulsifier, droplet size and fat type	OTC (80 mm/s, 0.5 N, 16 mm oscillation, pig's tongue surface and glass)	No saliva, 20 °C	QDA, n=8	PCA	Raw tongue, dry, rough, astringent		Dresselhuis *et al.* (2008b)*
		Protein-stabilised emulsions	OTC (10-80 mm/s, 0.5 N, 16 mm oscillation, sandblasted PDMS probes and glass surface)	Human saliva (stimulated), 20 °C	QDA, n=8		Raw tongue, dry, rough		Dresselhuis *et al.* (2007)
		Emulsions (22 and 33% fat, different emulsifiers)	Lab-built (1 Hz, 0.5 N, PCTFE ball on silicone disc)	Human saliva (stimulated), 37 °C	Triangle tests, n=20	Pearson's correlations	The bigger the difference in friction between samples, the better the discrimination	Mixed (10 mm/s)	Bellamy *et al.* (2009)
		O/w emulsions	Lab-modified TA (0.1-40 mm/s, 0.57 N, PDMS surface and steel balls)	Artificial saliva (ions, mucin, α-amylase), 28 °C	Sensory ratings: compare to reference, n=25	Pearson's correlations	Smooth (-)	Boundary, mixed (0.1-30 mm/s)	Upadhyay and Chen (2019)
	Emulsion-filled gels	Emulsion-filled gels varying in fat content (5 and 15%) and type of emulsifier (bound or unbound fat droplets)	OTC (80 mm/s, 0.5 N, 16 mm oscillation, flat-bottom PDMS probe and glass surface)	No saliva, 20 °C	DA, n=13		Fatty (-)	Mixed (80 mm/s)	Camacho *et al.* (2015)
		Emulsion-filled gels (gelatine) after simulated oral processing	OTC (10-80 mm/s, 0.5 N, 16 mm oscillation, flat-bottom PDMS probe and glass surface)	No saliva, 20 °C	QDA, n=11				Liu *et al.* (2015)

	Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
	Microbubble dispersions, o/w emulsions and protein solutions (without/with thickener or gelling agent)	OTC (10-80 mm/s, 0.5 N, 16 mm oscillation, flat-bottom PDMS probe and glass surface)	No saliva, 20 °C	Tetrad test, n=7				Rovers *et al.* (2016)
	O/w emulsions and emulsion-filled gels (gelatine)	OTC (10-80 mm/s, 0.5 N, 16 mm oscillation, flat-bottom PDMS probe and glass surface)	No saliva, 20 °C	QDA, n=12				Liu *et al.* (2016)
	Emulsion-filled mixed gels (gelatine/agar, κ-carrageenan/LBG, low/high acyl gellan) after simulated oral processing	OTC (10-80 mm/s, 0.5 N, 16 mm oscillation, flat-bottom PDMS probe and glass surface)	No saliva, 37 °C	QDA, n=12	Pearson's correlations	Sticky [*first bite*], sticky, rough, powdery, spreadable, fatty [*chew-down*], fatty, dry [*after-feel*]	Mixed (80 mm/s)	Devezeaux de Lavergne *et al.* (2016)*
Hydrocolloid foods	Liquids with additives and commercial liquid foods (mainly syrups)	Friction apparatus (chamois-coated surfaces)	No saliva, 20 °C	Ratings compared to reference	Pearson's correlations	Smooth (-)	Mixed (25 mm/s)	Kokini, Kadane and Cussler (1977)
	Polysaccharide (xanthan, locust bean gum, pectin) mixed with protein gel particles of different types, sizes and hardness	MTM (5-500 mm/s, 2 and 5 N, neoprene O-ring on neoprene disc set-up)	No saliva, 30 °C	QDA, n=11	Pearson's correlations	Slippery (-)	Mixed (50 mm/s)	Chojnicka-Paszun, Doussinault and de Jongh (2014)
	Hydrogels (κ-carrageenan) after simulated oral processing	MTM2 (1-1000 mm/s, 2 N, PDMS ball-on-disc set-up)	Artificial saliva (ions, mucin), 37 °C	DA, n=11	Pearson's correlations	Pasty (-), slippery, salivating	Mixed (50 mm/s)	Krop *et al.* (2019)*

		Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
Real foods	Milk	Commercial liquid and semi-solid foods (mainly dairy)	Tribo-rheocell accessory (0.21-0.85 mm/s, 10-45 g, chamois-coated surfaces)	No saliva, 20 °C	Ratings compared to reference	Pearson's correlations	Smooth (-)	Mixed (25 mm/s)	Kokini and Cussler (1983)
		Skimmed milk (two types of inulin)	MTM (0-750 mm/s, 5 N, nitrile ring on silicon disc set-up)	No saliva, 21 °C	QDA, n=11	Pearson's correlations	Thin-as-water (-), creamy (-)	Hydrodynamic (750 mm/s)	Meyer *et al.* (2011)*
		Skimmed and full fat milk (0.15, 0.3, 0.5, 0.7, 1.0, 2.0, 3.0, 4.0 and 6.5% fat)	MTM (5-500 mm/s, 5 N, neoprene O-ring on silicone, neoprene or Teflon disc set-up)	No saliva, 20 °C	QDA, n=10	Pearson's correlations	Creamy (-), soft/velvet (-), fat-film (-), slimy (-), watery	Mixed (10 mm/s)	Chojnicka-Paszun, de Jongh and de Kruif (2012)
		Non-fat stirred acid milk gels with and without the addition of saliva	Tribo-rheocell accessory (0.01-100 rpm, 2.1 N, double-polypropylene ball on whey protein isolate gel plates glued to base)	Human saliva (stimulated), 25 °C	DA with reference samples, n=7	Pearson's correlations	Chalky, smooth (-), gritty (-)	Boundary, mixed (0.016-100 mm/s)	Joyner, Pernell and Daubert (2014)*
		Skim (fat < 0.2%), 1% fat, whole (fat > 3.25%) milk	Tribo-rheocell accessory (0.15-750 mm/s, 1 N, double-polypropylene ball on PDMS disc)	Human saliva (stimulated), 25 °C	Paired comparison (2-AFC), n=24				Li *et al.* (2018b)
		Skim (fat < 0.2%), 2% fat, 5% fat milk	Tribo-rheocell accessory (0.15-750 mm/s, 1 N, double-polypropylene ball on PDMS disc)	No saliva, 25 °C	Spectrum, n=7	Regression analysis	Astringency	Mixed to hydrodynamic (100 mm/s)	Li *et al.* (2018a)
	Mayonnaise	Mayonnaises and custard desserts	Friction tester (rubber band on metal cylinder)	Human (stimulated) and artificial saliva (ions, mucin)	QDA, n=8	PCA and Pearson's correlations	Creamy (-), rough		de Wijk and Prinz (2005)*
		Vanilla custard desserts	Friction tester (rubber band on metal cylinder)	Human saliva (stimulated)	QDA, n=8	PCA and PLS2 (regression coefficients)	Creamy (-), fatty (-), rough		de Wijk, Prinz and Janssen (2006)*

	Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
	Vanilla custard desserts, white sauces and mayonnaises	Friction tester (rubber band on metal cylinder)	Human saliva (stimulated)	QDA, n=9	PCA and PLS2	Creamy, fatty (-)		de Wijk and Prinz (2007)*
	Mayonnaises and mayonnaise-type dressings	Friction tester (rubber band on metal cylinder)	Unspecified saliva (stimulated)	QDA, n=10	PCA and PLS2	Grainy, powdery, sticky		Terpstra *et al.* (2009)
Yoghurt	Stirred yogurts	Tribo-rheocell accessory (0.001-1000 min-1, 3 N, steel ball on styrene butadiene rubber pad)	No saliva, 10 °C	DA, n=22	PCA, Pearson's correlations and backward multiple linear regressions	Viscous (-), fatty (-), creamy (-)	Boundary (1 mm/s)	Sonne *et al.* (2014)
	Whey protein-pectin mixtures added to low-fat yoghurt matrix compared to a full-fat control	Tribo-rheocell accessory (0.001-1000 mm/s, 3 N, steel ball on styrene butadiene rubber pad)	No saliva, 10 °C	DA, n=22		Creamy (-)		Krzeminski *et al.* (2014)
	Model emulsions, white sauce, milk and yoghurt	Tribo-rheocell accessory (10 mm/s, steel wrapped in acrylonitrile butadiene copolymer films)	No saliva, 20 °C	Paired comparison (2-AFC), n=35-50			Mixed (10 mm/s)	Le Calvé *et al.* (2015)
	Milk, yoghurt, soft cream cheese	MTM (1-1000 mm/s, 2 N, PDMS ball-on-disc set-up)	Artificial saliva (ions, mucin), 37 °C	Triangle and intensity score, n=63 consumers				Laguna *et al.* (2017)
	A range of casein to whey protein ratio yoghurt systems (80:20, 70:30, 60:40 and 50:50) prepared from skim milk (no fat)	Tribo-rheocell accessory (0.001-1000 mm/s, 3 N, stainless steel ball on rubber pads)	No saliva, 10 °C	DA, n=7	Pearson's correlations	Gelatinous, aerated, lumpy, grainy, adhesive (-), creamy (-), smooth (-) [*in-mouth*], difficult to swallow (-), mouth coating (-) [*after-feel*]	Boundary (0.1 mm/s)	Laiho *et al.* (2017)*

	Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
	Yoghurts with extra protein (milk powder, whey protein concentrate) and modified starch	Lab-modified TA (0.1-10 mm/s, 0.27 N, silicone elastomer surface and steel balls)	Human (stimulated) and artificial saliva (ions, mucin, α-amylase), 25 °C	Flash profiling, n=13				Morell, Chen and Fiszman (2017)
	Pot-set yoghurts (0.1, 1.3 or 3.8% milk fat, with added gelatine, xanthan gum, carrageenan or modified starch)	Tribo-rheocell accessory (0.01-100 s^{-1}, 2 N, half-ring on surgical tape plate)	No saliva, 35 °C	QDA, n=8	Ranking of products according to the different parameters			Nguyen *et al.* (2017)
	Skim (0.1% fat) stirred yoghurt (with added inulin, pectin, β-glucan or galacto-oligosaccharides)	Tribo-rheocell accessory (0.01-100 s^{-1}, 2 N, half-ring on surgical tape plate	No saliva, 35 °C	QDA, n=8				Ng *et al.* (2018)
	Custard dessert formulations	Tribo-rheocell accessory (0.01-6.5 rad/s, 2 N, half-ring on surgical tape plate)	No saliva, 35 °C	Ranking DA, n=11, *not for all samples				Godoi, Bhandari and Prakash (2017)
Cheese	Cream cheese with varying pH, salt and fat content	Lab-built (0.7 mm/s, 100 g, pig's tongue on oesophagus shaft)	No saliva, 20 °C	DA, n=10	PLSR and mixed model ANOVA with Measurement Error Methodology	Creamy	Boundary (0.7 mm/s)	Janhoj *et al.* (2009)
	Cream cheese differing in fat content	Tribo-rheocell accessory (0.1-600 s^{-1}, 2 N, ring on surgical tape plate)	No saliva, 35 °C	TDS, n=10				Ningtyas *et al.* (2018)

	Lubricant	Tribology	Simulated oral conditions	Sensory	Statistical analyses	Correlation	Lubrication regime	Reference
Other	Two chocolate samples manufactured to the same shear viscosity	Tribo-rheocell accessory (0.001-420 mm/s, 0.5 N, steel ball on 3 polyurethane plates)	No saliva, 37 °C	Paired comparison (2-AFC), n=40		Mouth-coating (-)		Carvalho-da-Silva *et al.* (2013)
	Four milk chocolates	Tribo-rheocell accessory (0.02-750 mm/s, 3 N, stainless steel ball on PDMS plates)	Human saliva (stimulated), 40 °C	QDA, n=12				He *et al.* (2018)
	Gluten-free bread upon addition of different modified dietary fibres	Tribo-rheocell accessory (1 mm/s, 0.2 N, three steel balls on bread taped to plate)	No saliva, 20 °C	Time-intensity, n=10	Pearson's correlations	Firm, chewy, dry	Boundary (1 mm/s)	Kiumarsi *et al.* (2019)*
	Toothpaste	Lab-modified TA (0.03-20 mm/s, 0.57 N, PDMS surface and steel balls)	Artificial saliva (ions, mucin, α-amylase), 27 °C	Sensory ratings: compared to reference, n=25	Pearson's correlations	Smooth	Boundary (0.08 mm/s)	Cai, Li and Chen (2017)

In those studies that measured both instrumental and sensory lubrication data, a relationship with μ at a specific speed within the mixed regime, *i.e.* at speeds within the range of real tongue movements, has been reported (Sarkar *et al.* 2019; Steele and van Lieshout 2009). In addition, it is noteworthy that the use of artificial saliva in tribology experiments improved the strength of the relationship with sensory attributes, a role that is often ignored in the literature. Besides μ, many if not most studies in **Supplementary Table E.1** have also conducted bulk rheology and particle size analysis that have enabled better understanding of the physical reasoning behind sensory attributes, like in this thesis. For instance, besides μ in boundary to mixed regimes (30-100 mm/s), the sensory viscosity in milks was found to correlate strongly with instrumental viscosity (η) at 50 s^{-1} shear rate (Li *et al.* 2018a). And in another study, mean particle size ($d_{3,2}$) has been an important factor in understanding the reason behind higher μ values and corresponding increased sensory roughness (Liu *et al.* 2016). An increase in 'rough' perception was attributed to the higher $d_{3,2}$ values, which were much above the sensory detection threshold. In this thesis, we also considered bulk rheology (**Chapter 4**) and this was used not only to relate to sensory behaviour, but also helped to generate a Master curve particularly scaling the elastohydrodynamic regime. Again, to our knowledge, this has never been done to date.

7.3.4. Considerations of the satiety trial: study design

To test the satiating effects of the hydrogels with different chewing and oral lubrication properties, a preload paradigm was selected. Although preload studies are attractive for lab-based research, it is more realistic to study an independent variable as part of a meal rather than a small amount of food before a meal (Blundell

et al. 2009). However, due to the nature of the hydrogels, the participants' unfamiliarity with them and their low liking scores, as well as the lack of a clear meal time for these novel gels, it was more complicated to use a meal study design. In order to test the hypothesis that oral lubrication plays any role in satiety development, it was decided to use a preload study design followed by a snack to prove the concept first (**Chapter 6**).

A within-subjects repeated-measures design is most suitable for preload studies, where the participants serve as their own controls (Blundell *et al.* 2009). This serves to reduce inter-individual variability in satiety regulation, which is not controlled for in a between-subjects design. However, as we decided to use novel model hydrogels as preloads without further familiarisation, a between-subjects design was considered to be advantageous in preventing the participants from familiarising themselves with the preloads and acquiring any "learned" satiating effects. Learning about a products' satiating capacity occurs with repeat exposure (Yeomans 2012), and so within-subject design might affect the results of the preload hydrogels. Preload designs require the inclusion of control conditions, either covertly (to assess the physiological response to the preload) or overtly manipulated (to assess both the physiological and cognitive responses) depending on the tested hypothesis (Rolls and Hammer 1995). Therefore, the mint tea control condition was included to evaluate the effect of the preload study design on snack intake, while controlling for the influence of exposure to the mint flavour.

Palatability plays a major role in satiation (Blundell *et al.* 2010), so test foods should be similarly liked. For this thesis, novel model hydrogels were

developed without prior notions of their liking and satiating characteristics. They were given a peppermint flavour and green colour to avoid any (subconscious) comparisons with desert type products, as well a fruity-flavoured jelly candies. It is known that familiarity with the satiating characteristics of certain type of foods might influence their food intake, due to expectations rather than energy content (Cecil, Francis and Read 1998; Chambers, McCrickerd and Yeomans 2015). The pleasantness of the hydrogels between conditions was measured, and though scores were low, their intensity level for the different hydrogel samples was similar ($p > 0.05$, **Chapter 6**). Similarly, a snack was selected that was palatable, but not so highly palatable that intake would be high regardless of testing condition obscuring any condition effects.

Appetite ratings were recorded before and after the preloads, as well as after the *ad libitum* snack. Since ratings were not collected over a longer interval beyond the snack session, they are not truly representative of differences in satiety induced by the manipulation. In addition, we are unable to say anything in this thesis about the longer term-effects of the chewing and oral lubrication manipulations. However, due to the nature of the small preload manipulations (four to five bite size hydrogels in 10 minute time period), longer-term effects were not expected.

In addition to the appetite measures, thirst ratings were recorded. The feeling in the mouth at the start of the preload intervention (level of dryness), might have an effect on how its lubrication qualities are perceived. Thirst was not specifically standardised at the start of the study, as the liquid intake during the day varies naturally between individuals. During the standard lunch, *ad libitum* water was provided and participants were free to drink water in between sessions. The water intake between standard lunch, and the preload and snack, however, was not

logged. Nevertheless, since the thirst ratings did not differ between conditions, there is no expected effect of water intake during the day on the intake of the snack.

7.4. Future directions and concluding remarks

The main aim of this thesis was to understand the role of instrumental friction measurements and the associated oro-sensorially perceived lubrication in the fields of food science and oral processing, and show the potential of lubrication as an aspect of oral processing on short-term satiety and satiation. In this thesis we have shown the added value of oral lubrication and tribology in satiety research using non-fat hydrogels. Though tribological measurements may be improved upon further by better mimicking the oral surfaces, linking instrumental data with sensory measurements shows potential for predicting the sensory lubrication perception and the oral processing behaviour. Based on the findings in this thesis, the following opportunities can be identified for future research:

Oral lubrication properties of hydrogels: a selection of hydrogels were studied in this thesis. Further analysis of the instrumental and sensorial characteristics of other hydrocolloids, mixtures and levels of inhomogeneity might reveal additional insights in the mechanisms and contributing factors behind oral lubrication. Although we developed important insights by using artificial saliva containing ions and mucin to replicate human oral conditions at 37 °C, developing tribological measurement surfaces for the tribometer that replicate the oral surfaces better might help to achieve better correlations between instrumental friction measurements and sensory perception.

Dynamic aspects of oral processing and texture perception: texture perception in this thesis was analysed with descriptive analysis, an adapted version of Quantitative Descriptive Analysis (QDA®). Proper analysis of the sensory texture properties of model gels requires more extensive training of the panellists on the model foods, as well as the methodology (purpose, rating scales etc) and attributes, specifically related to lubrication. On the other hand, alternative sensory characterisation methodologies that require less training/time, such as flash profiling, progressive profiling, (temporal) check-all-that-apply (TCATA) *etc.* can be explored. In addition, it would be good to compare the simulated hydrogel bolus samples to real food bolus samples from participants. Instrumental analyses of the bolus, such as fracture properties, particle size and amount and quality (*e.g.* protein content) of the saliva incorporated into the hydrogel bolus, can be measured as a function of oral processing time to gain further insights on internal lubrication.

Satiety study design: a preload between-subjects design was used to measure the short-term satiety effects of manipulating the chewing and oral lubrication behaviour during a snack. Follow-up satiety experiments should focus on the effects during a different meal moment to confirm whether these results with the same gels extend beyond a snack to a test meal design. Furthermore, the effects of chewing and lubrication should be explored in real food products, to test the validity in a more natural setting. The time until the next eating occasion after the intervention meal can also be used as an additional measure to quantify satiety, *e.g.* using food diaries. Finally, in order to extrapolate the results to the longer-term satiety effects, several post-ingestive measures can be recorded, such as the gastro-intestinal response, circulating hormones and rate of gastric emptying.

Despite these reflections highlighting limitations and future studies, the overall outcome of this thesis has been to showcase the impact of systematic instrumental friction manipulations on hydrogels and the associated changes to their lubrication properties, the impact on oral processing, sensory perception and short-term satiation. The systematic approach adopted in this thesis should now be replicated and extended to real foods in order to advance our understanding and to stimulate further efforts to produce satiety-enhancing foods for different groups of consumers. Further research efforts might specifically target babies or young children to improve learned eating habits, and control their food intake better with an eye on preventing future obesity and other health related problems. On the other hand, a research focussing on helping older adults who are suffering from malnutrition due to the decreasing quantity and/or quality of saliva might help to increase their food intake and improve their enjoyment of eating.

7.5. References

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Appendix A

Supplementary information for Chapter 2

Quality assessment tool (Moore, 2012), based on:

- National Institute for Clinical Excellence (NICE). 2007. The Guidelines Manual. London, National Institute for Clinical Excellence.

- Downs and Black. 1998. The feasibility of creating a checklist for the assessment of the methodological quality of both randomised and non-randomised studies of health care interventions. *J. Epidemiology Community Health*, **52**(1), pp. 377-384.

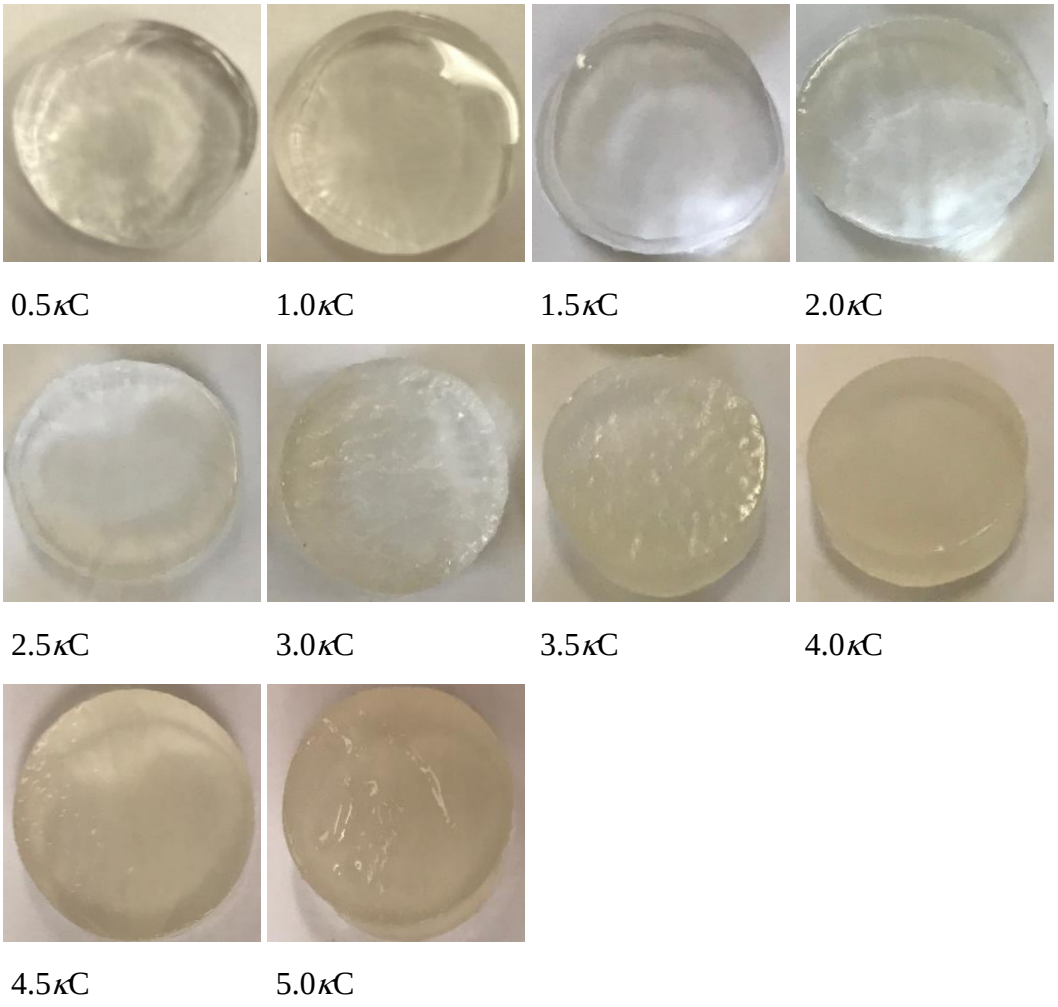
QUESTIONS	**Yes = 2 Partly = 1 No = 0**
1. Is the Qualitative/Quantitative approach appropriate? ➢ Could another approach have better addressed the research question?	
2. Is the study clear in what it seeks to do? Qualitative: ➢ Is the purpose of the study discussed? ➢ Are the research question(s) presented? ➢ Is there adequate/appropriate reference to the literature? ➢ Are underpinning values/assumptions/theory discussed? Quantitative: ➢ Is the purpose of the study discussed? ➢ Are the hypothesis presented? ➢ Are the Outcomes to be measured clearly stated?	
3. How defensible/rigorous is the research design/methodology? ➢ Is the design appropriate to the research question? ➢ Is a rationale given for using the approach?	
4. How well was the data collection carried out? ➢ Are the data collection methods clearly described? ➢ Were the appropriate data collected to address the research question?	
5. Is the context clearly described? Both: ➢ Are the characteristics of the participants and settings clearly defined? ➢ Was context bias considered? Qualitative: ➢ Has the relationship between the researcher and the participants been considered? ➢ Does the paper describe how the research was explained and presented to the participants?	

6. Was the analysis sufficiently rigorous? Qualitative: ➢ Is the procedure explicit – is it clear how the data were analysed to arrive at the results? ➢ How systematic is the analysis – is the procedure reliable/dependable? ➢ Is it clear how the themes and concepts were derived from the data? Quantitative: ➢ Were the measures used valid and reliable?	
7. Is the analysis reliable? Qualitative: ➢ Did more than one researcher theme and code transcripts/data? ➢ Did participants feedback on the transcripts/data? (if possible and relevant) Quantitative: ➢ Were the statistical tests used to assess the main outcomes appropriate?	
8. Are the findings convincing? Both: ➢ Are the findings clearly presented? ➢ Are the findings internally coherent? ➢ Are the data appropriately referenced? ➢ Is the reporting clear and coherent? Qualitative: ➢ Are extracts from the original data included? Quantitative: ➢ Have actual probability values been reported?	
9. Are the findings relevant to the aims of the study?	

10. Are the conclusions adequate? ➢ How clear are the links between data, interpretation and conclusions? ➢ Are the conclusions plausible and coherent? ➢ Have alternative explanations been explored and discounted? ➢ Does this study enhance understanding of the research subject? ➢ Are the implications of the research clearly defined? ➢ Is there adequate discussion of any limitations?	
11. How clear and coherent is the reporting of ethical considerations? ➢ Have ethical issues been taken into consideration? ➢ Are ethical issues discussed adequately – do they address consent and anonymity? ➢ Have the consequences of the research been considered; for example, raising expectations, changing behaviour?	
TOTAL	

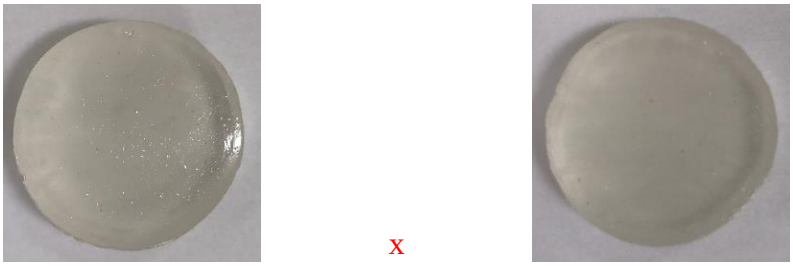
Appendix B

Supplementary information for Chapter 3



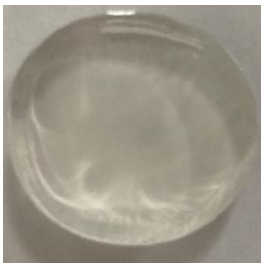
Supplementary Figure B.1. Images of single κC hydrogels with a wide range of concentrations (0.5 - 5.0 wt%).

0.7κC0.3LBG | 0.8κC0.2LBG | 0.9κC0.1LBG



1.5κC0.5LBG | x | 1.67κC0.33LBG | x | 1.8κC0.2LBG | x | 2.25κC0.75LBG | 2.5κC0.5LBG | 2.7κC0.3LBG

Supplementary Figure B.2. Images of mixed κC and LBG hydrogels with different concentrations and ratios (1.0-2.0 wt% total hydrocolloid concentrations).



x

x

x

1κC1NaA

1.4κC0.6NaA

1.5κC0.5NaA

1.8κC0.2NaA

x

x

x

x

1.5κC1.5NaA

1.8κC1.2NaA

2.25κC0.75NaA

2.7κC1.3NaA

x

1.6κC0.2CaA$_{300}$

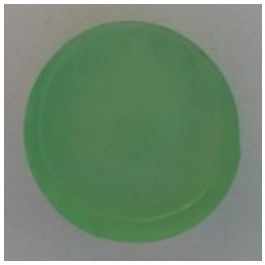
2.4κC0.2CaA$_{300}$

1.6κC0.2CaA$_{1000}$

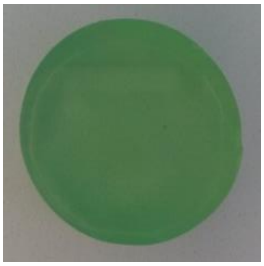
Supplementary Figure B.3. Images of mixed κC and alginate gels (NaA or CaA) with different concentrations and ratios (1.8-3.0 wt% total hydrocolloid concentrations), without and with beads (of various sizes).

Appendix C

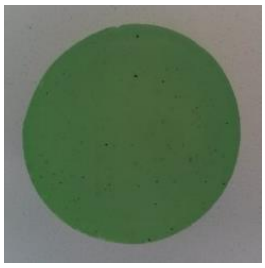
Supplementary information for Chapter 4



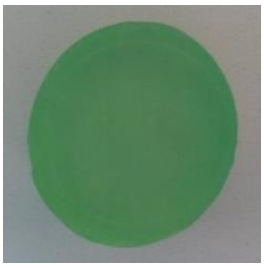
2κC



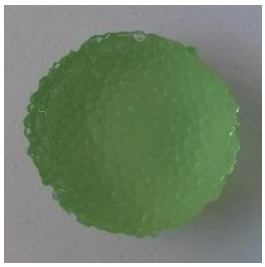
3κC



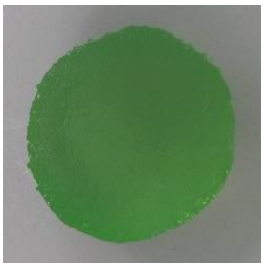
2.25κC0.75LBG



1.5κC0.5NaA



1.6κC0.2CaA$_{1000}$



1.6κC0.2CaA$_{300}$

2.4κC0.2CaA$_{300}$

Supplementary Figure C.1. Visual images of the different hydrogels.

Questionnaires:

- QDA test CompuSense

Panelist Code: _____ Panelist Name: _____

Welcome to Compusense *five* Release 5.2

Press the 'Continue' button below to begin the test.

Panelist Code: _____ Panelist Name: _____

Welcome to this test session.

Today you will be rating the texture intensity of 9 different attributes for several different gels.

Please check you received the sample number corresponding to the sample number displayed on the screen. Alert the researcher when you have finished the assessment and would like to receive the next sample.

Please cleanse your palate with some water and/or the crackers provided. Make sure there are no more gel particles in your mouth before moving on to the next sample.

Panelist Code: _____ Panelist Name: _____

Question # 1 - Sample <<Sample xx>>

Please rate the intensity of the following attributes.

Smooth

A little A lot

Firm

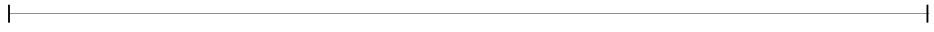
A little A lot

Elastic

A little A lot

Chewy

A little A lot



Cohesive

A little A lot

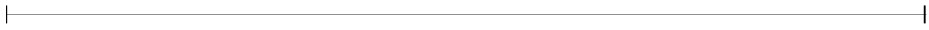
Panelist Code: _____ Panelist Name: _____

Question # 2 - Sample <<Sample xx>>

Please rate the intensity of the following attributes.

Pasty

A little A lot



Slippery

A little A lot

Salivating

A little A lot

Melting

A little A lot

Panelist Code: _____ Panelist Name: _____

Question # 3.

What is your gender?

- Male
- Female

Question # 4.

What is your age?

Age _____

Panelist Code: _____ Panelist Name: _____

THANK YOU

Appendix D

Supplementary information for Chapter 5

Questionnaires:

- Instructions for the video recordings of the chewing behaviour

- Eating Capability (EC) measurements

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Characterisation of model gels - Video Analysis

Participant Information

Participant Name: Participant Code:

Gender:

Age:

Instructions

Please look into the camera, and refrain from moving your head as much as possible. Please don't talk during the sample assessment. Place the sample in your mouth and chew as you would normally do and swallow. You will be given a warm-up sample first to familiarise yourself with the sort of samples and the test procedure.

Optionally: at the point you feel the urge to swallow, please raise your hand and then you may spit-out the sample in the provided cup and rinse your mouth with water.

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Participant Code:

Characterisation of model gels – Eating capability

Tongue Pressure

Place the tongue bulb in your mouth into the centre of the oral cavity. Compress between your tongue and hard palate as hard as you can. Repeat 3 times. The maximum pressure will be recorded on the Iowa Oral Performance Instrument.

	Tongue Pressure
1	
2	
3	

Max. Bite Force

Place the bite sensor between your left side molars and compress, as hard as you would for normal chewing procedures. Hold each measurement for a couple of seconds. Repeat 3 times. Then do the same for the front incisors and the right side molars. The minimum resistance shown on the multimeter will be recorded.

	Left Side Molars	**Front Incisors**	**Right Side Molars**
1			
2			
3			

Appendix E

Supplementary information for Chapter 6

Questionnaires

- Satiety study

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Mint stimulus study – Q1

PARTICIPANT INFORMATION

Participant Name: _____ Participant ID: _____

Age: _____ D.O.B: _____

Estimated height: _____ BMI: _____

Estimated weight: _____

Email: _____

GENERAL INFORMATION

Ethnicity: _____

What is your occupation? Employed Unemployed Retired
Housewife/husband Student

If you are a student, what course are you currently studying? _____

What year of study are you in? _____

YOUR HEALTH

How would you rate your general health?

Not at all 1 2 3 4 5 6 *Extremely*

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Do you suffer from any medical conditions (*i.e.* heart, asthma, diabetes)?

_____

Are you currently taking any medication (prescribed or over-the-counter)? YES/NO

If YES, please specify: _____

Have you taken any medication recently? YES/NO

If YES, please specify any medications taken during the past month:

_____

Do you smoke? Yes, regularly Yes, occasionally
No, given up No, never

Do you exercise regularly? YES/NO

If YES, how many times a week? One to four More than four

Generally, what sort of exercise do you do? _____

YOUR DIET

In general, how healthy would you rate your diet?

Not at all 1 2 3 4 5 6 *Extremely*

Do you usually eat breakfast? YES/NO

Do you usually eat lunch? YES/NO

Do you ever eat more in order to gain weight? YES/NO

Do you ever eat less in order to lose weight? YES/NO

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Are you a vegetarian? YES/NO

Do you have any food allergies or intolerances?

_____

Are there any specific foods you do not like or could not eat?

_____

BREAKFAST INFORMATION

When did you last eat something? _____

When did you last drink something? _____

When did you eat your breakfast? _____

What did you eat for breakfast? _____

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DIETARY RESTRAINT

Please indicate the answer that applies to you by placing "X" next to the appropriate response from the 5 options. Please answer all questions, based on your behaviour in the past three months.

	Never	Seldom	Some-times	Often	Very often
1. When you have put on weight, do you eat less than you usually do?	☐	☐	☐	☐	☐
2. Do you try to eat less at mealtimes than you would like to eat?	☐	☐	☐	☐	☐
3. How often do you refuse food or drink offered because you are concerned about your weight?	☐	☐	☐	☐	☐
4. Do you watch exactly what you eat?	☐	☐	☐	☐	☐
5. Do you deliberately eat foods that are slimming?	☐	☐	☐	☐	☐
6. When you have eaten too much, do you eat less than usual the following day?	☐	☐	☐	☐	☐
7. Do you deliberately eat less in order not to become heavier?	☐	☐	☐	☐	☐
8. How often do you try not to eat between meals because you are watching your weight?	☐	☐	☐	☐	☐
9. How often in the evenings do you try not to eat because you are watching your weight?	☐	☐	☐	☐	☐
10. Do you take your weight into account with what you eat?	☐	☐	☐	☐	☐

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Mint stimulus study – Q2

Participant ID:

Please answer the following questions by placing a **vertical mark** through the line. Regard the ends of each line as indicating the most extreme sensation you have ever experienced. Please taste the provided sample before answering the first two questions.

1. How much do you like the taste of this gel?

Not at all Extremely

2. Would you be prepared to consume this gel as part of a study?

YES NO

3. How much do you like the taste of mint tea?

Not at all Extremely

4. Would you be prepared to consume mint tea as part of a study?

YES NO

5. How much do you like the taste of a cheese sandwich?

Not at all Extremely

6. How much do you like the taste of Braeburn apples?

Not at all Extremely

7. How much do you like the taste of flapjacks?

Not at all Extremely

8. Would you be prepared to consume a cheese sandwich, an apple and a flapjack as a lunch meal?

YES NO

9. How much do you like the taste of ready salted crisps?

Not at all Extremely

10. Would you be prepared to consume a ready salted crisps as a snack?

YES NO

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Mint stimulus study

We want you to tell us about your perception of a mint stimulus on a snack.

- First, answer the questions on Questionnaire 3.
- You will be presented with a mint stimulus. Eat the first mint stimulus, followed by a sip of water. Follow each mint stimulus, by a sip of water until all the stimuli have been consumed. You have 10 minutes to eat all stimuli, as well as finish the water.
- Please answer Questionnaires 4 and 5.
- After finishing these questionnaires, a snack will be presented to you. Answer questions 1 and 2 on Questionnaire 6 with a first bite of the snack. Then, you will be allowed to eat as much as you like from the snack until comfortably full while answering the remaining two questions.
- Finally, complete Questionnaire 7 and the debrief questionnaire.

Thank you for you participation!

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Mint stimulus study – Q3

Participant ID: Time point: t_0

Please answer the following questions by placing a **vertical mark** through the line. Regard the ends of each line as indicating the most extreme sensation you have ever experienced.

1. When did you last eat?

---

2. What did you last eat?

---

3. How hungry do you feel right now?

Not at all hungry — Extremely hungry

4. How full do you feel right now?

Not at all full — Extremely full

5. How strong is your desire to eat right now?

Not at all strong — Extremely strong

6. How strong is your appetite right now?

Not at all strong ——————————————— Extremely strong

7. How thirsty do you feel right now?

Not at all thirsty ——————————————— Extremely thirsty

8. How nauseous do you feel right now?

Not at all nauseous ——————————————— Extremely nauseous

9. How strong is your desire to eat something sweet right now?

Not at all strong ——————————————— Extremely strong

10. How strong is your desire to eat something salty right now?

Not at all strong ——————————————— Extremely strong

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Mint stimulus study – Q4

Participant ID:

Please eat the first mint stimulus, followed by a sip of water. Follow each mint stimulus, by a sip of water until all the stimuli have been consumed. Make sure you finish all of the mint stimuli and the water that was presented to you. Answer the following questions by placing a **vertical mark** through the line. Regard the ends of each line as indicating the most extreme sensation you have ever experienced.

1. How pleasant is the taste of the mint stimulus?

Not at all pleasant ——— Extremely pleasant

2. How strong is the mint flavour of the mint stimulus?

Not at all minty ——— Extremely minty

3. How sweet is the taste of the mint stimulus?

Not at all sweet ——— Extremely sweet

4. How chewy is the mint stimulus?

Not at all chewy ——— Extremely chewy

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Mint stimulus study – Q5/Q7

Participant ID: Time point: t_1 / t_2

Please answer the following questions by placing a **vertical mark** through the line. Regard the ends of each line as indicating the most extreme sensation you have ever experienced.

1. How hungry do you feel right now?

Not at all hungry — Extremely hungry

2. How full do you feel right now?

Not at all full — Extremely full

3. How strong is your desire to eat right now?

Not at all strong — Extremely strong

4. How strong is your appetite right now?

Not at all strong — Extremely strong

5. How thirsty do you feel right now?

_____

Not at all thirsty — Extremely thirsty

6. How nauseous do you feel right now?

_____

Not at all nauseous — Extremely nauseous

7. How strong is your desire to eat something sweet right now?

_____

Not at all strong — Extremely strong

8. How strong is your desire to eat something salty right now?

_____

Not at all strong — Extremely strong

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Mint stimulus study – Q6

Participant ID:

We want you to tell us about your perception of the mint stimulus on a snack. In order to do this, please answer the first two questions with your first bite of the snack. Indicate your answer by placing a **vertical mark** through the line. Regard the ends of each line as indicating the most extreme sensation you have ever experienced.

1. How strong is your desire to eat the crisps?

Not at all strong ——— Extremely strong

2. How pleasant is the taste of the crisps?

Not at all pleasant ——— Extremely pleasant

Please now eat a normal-sized snack. Whilst you are eating the snack, please answer the next two questions.

3. How sweet is the taste of the crisps?

Not at all sweet ——— Extremely sweet

4. How salty is the taste of the crisps?

Not at all salty ——— Extremely salty

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Mint stimulus study – Debrief questionnaire

Participant ID:

In this questionnaire we are interested in your views of the study and your experiences as a volunteer. In order for us to learn as much as possible from the study we would appreciate you completing this questionnaire fully and honestly. All your responses will be treated in confidence.

1. What did you think the aim of the study was?

_____

2. How easy did you find it to comply with the instructions of this study?

_____

3. Had you ever tasted anything similar to the model gels?

   YES/NO

4. What did they remind you of?

_____

5. Do you like custard desserts? YES/NO

6. Do you like bubble tea? YES/NO

7. Have you seen the New Year Diet Special episode of the Channel 4 programme “Food Unwrapped”, which aired on Thursday 4th January 2018 at 20:00 BST? YES/NO

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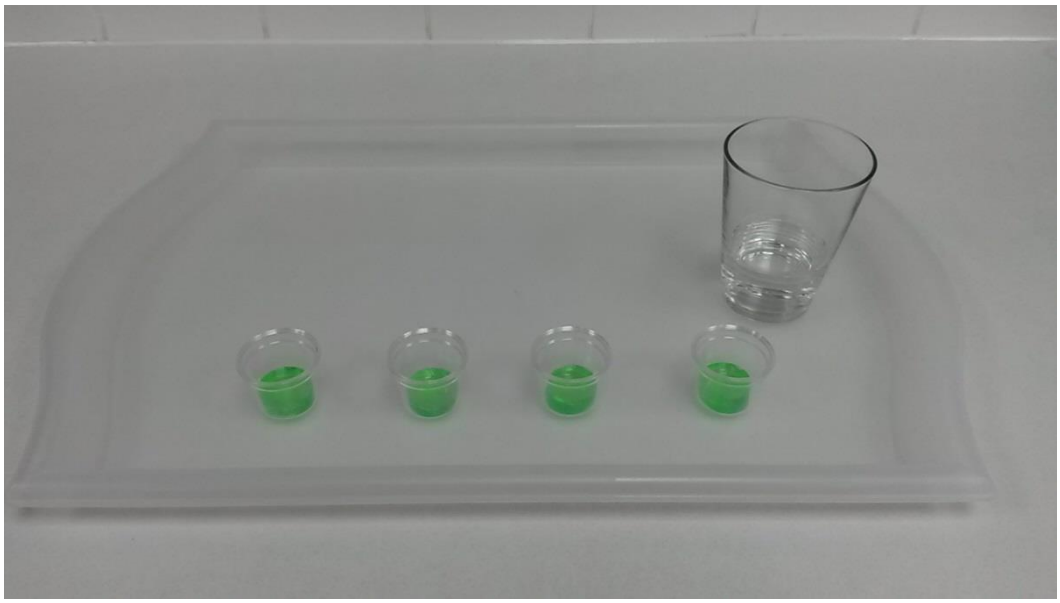
If you would like to receive a summary of the study results after the study has been completed, please leave your email address below:

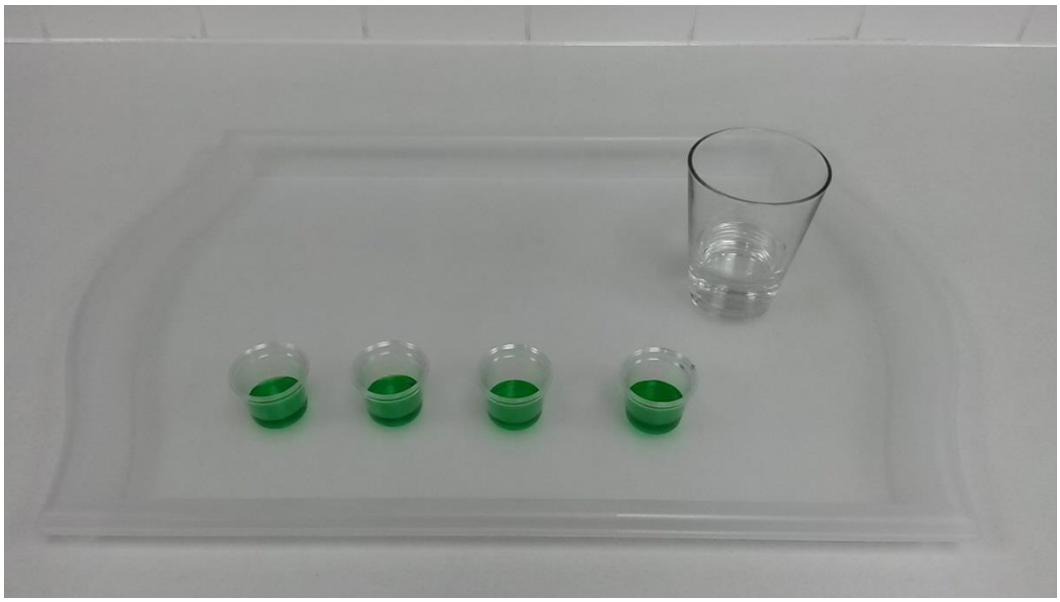
_____

Supplementary Figure E.1. Standard lunch

Supplementary Figure E.2. Snack intervention of ready salted crisps



Supplementary Figure E.3. Preload hydrogel samples



Supplementary Figure E.4. Preload control tea samples