Unravelling the unravelled

Microfibrillated cellulose as texture modifier
in liquid and semi-solid foods

Annelies Blok

Propositions

1. Microfibrillated cellulose (MFC) can be used as texture modifier in liquid and semi-solid foods.
   (this thesis)

2. Spray-drying microfibrillated cellulose in the presence of maltodextrin facilitates its use in foods without compromising its functional properties.
   (this thesis)

3. Flexitarianism is an ineffective way to reduce global meat consumption.

4. It is a flaw of the current review system that the review process takes longer than the writing process.

5. Any organisation that claims to be sustainable should completely reimburse commuting by public transport.

6. Working fewer hours makes people more productive.

Propositions belonging to the thesis, entitled

Unravelling the unravelled: Microfibrillated cellulose as texture modifier in liquid and semi-solid foods

Annelies E. Blok
Wageningen, 21 October 2022

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Happiness is the greatest form of success.

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General Introduction

1.1. Healthy food for human and planet

As the number of overweight and obese individuals continues to rise (World Health Organization, 2022), so do consumers' awareness and concern about the healthiness of the foods they eat. In addition, the future of our planet is of growing concern (Grunert *et al.*, 2014), which has generated a demand for food products that are healthy for humans and are produced environmentally friendly. Since fat is the most energy dense nutrient, an extensive range of low-fat or light products has been developed to support weight loss and management. However, fat has a considerable effect on the rheological and sensory properties of foods. The loss of consistency and sensory texture as a result of fat reduction is often compensated for by addition of food thickeners. Consumers' interest and knowledge about food ingredients has increased rapidly over the last couple of years and nowadays consumers are more critical of the way food is produced, formulated and processed (Meijer *et al.*, 2021). Sustainability concerns have moreover promoted consumers' awareness of the origin of food (ingredients) and the ways they are produced. This has raised interest in valorisation of waste streams as starting material to produce new food ingredients. Microfibrillated cellulose (MFC) is an example of a food ingredient that can be prepared from agricultural waste streams (Lavoine *et al.*, 2012). MFC is prepared from cellulose, and hence acts as a non-digestible dietary fibre that does not provide energy upon consumption. MFC has viscosifying properties and could therefore be a promising low calorie texture modifier that can be obtained from waste streams. The research described in this thesis investigated the effects of MFC on sensory, rheological and tribological properties of liquid and semi-solid foods, and compared these to other frequently used food thickeners.

1.2. Microfibrillated cellulose

1.2.1. Cellulose

Cellulose is the most abundant renewable polymer on earth. It is the main building block of plant cell walls due to its high tensile strength, but can also be produced by bacteria, algae and fungi (Wüstenberg, 2014). Cellulose consists of linear unbranched chains of β-D-glucose with a high degree of polymerisation. The large number of hydroxyl groups promotes the formation of hydrogen bonds with adjacent cellulose molecules, resulting in the formation of crystalline structures comprising 36 cellulose chains (Chinga-Carrasco, 2011). These so-called 'elementary fibrils' aggregate and bundle into microfibrils with lengths in the nanometre scale. In turn, microfibrils assemble into cellulose fibres (macrofibrils) with lengths up to several micrometres and diameters up to 400 nm (Wüstenberg, 2014) (Figure 1.1). As a result of its high molecular weight and crystalline structure, untreated cellulose is essentially insoluble in water. Apart from highly crystalline regions, cellulose contains less ordered amorphous regions in which cellulose is more loosely packed. These regions are more sensitive to hydrolysis due to increased accessibility of hydroxyl groups.

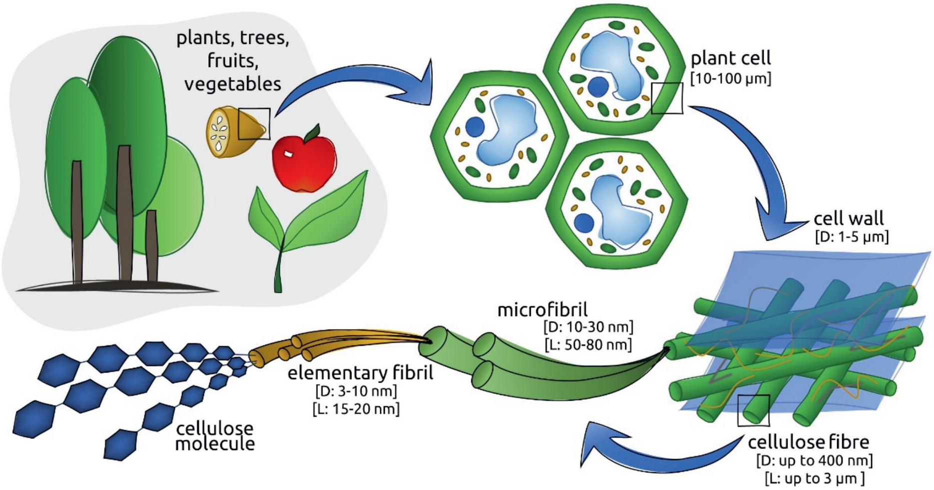

Figure 1.1. *Schematic overview of the origin and (micro)structure of cellulose and the corresponding length scales.*

While herbivores can degrade cellulose with the help of ruminal microorganisms, humans cannot digest cellulose due to a lack of cellulose-hydrolysing enzymes (*e.g.* cellulase). Cellulose therefore serves as a non-digestible dietary fibre in the human diet, where it can impart several health benefits such as reduction of appetite and risk of cardiovascular disease, colorectal cancer and diabetes (Dhingra *et al.*, 2012; Evans, 2020). Cellulose can be considered an environmentally friendly material, as it is renewable and biocompatible. It is estimated that annually 100-1,000 billion tonnes of cellulose are produced in nature through photosynthesis (Coffey *et al.*, 2006). The majority of cellulose is obtained from wood pulps, of which more than 97% is used by the paper industry (Wüstenberg, 2014). Perennial and annual plants compose an alternative source of cellulose, as most plant materials have relatively high cellulose content (30-90%) (Marchessault & Sundararajan, 1983). Agricultural waste streams and by-products can therefore be revalorised and used for the extraction of cellulose and other fibrous material. Examples of plant waste materials suitable for deriving cellulose include crop residues such as stems, leaves, stalks and husks, and fruit or vegetable peel and pulp. The widespread availability of cellulose-rich waste materials makes it a cheap starting material for preparation of cellulose derivatives, such as methylcellulose, microcrystalline cellulose (MCC), cellulose whiskers and microfibrillated cellulose (MFC). Preparation of cellulose derivatives generally aims at deagglomeration of fibrillar structures to enhance their compatibility with water. The large range of functionalities displayed by these cellulose derivatives raised industrial interest and promoted their use in foods.

1.2.2. From cellulose to microfibrillated cellulose (MFC)

One way of improving the water compatibility of cellulose, and thus its applicability in foods, is by physical treatment. The hierarchical structure of cellulose fibres can be deconstructed into its elementary constituents by mechanical treatment while preserving the chemical structure of the cellulose. This process was developed in 1977, but was not published and patented until 1983 (Herrick *et al.*, 1983; Turbak *et al.*, 1983a). According to these patents, high shear forces can transform micrometre-sized cellulose from wood pulp suspensions into nano-scale microfibril aggregates (Figure 1.2). The mechanical treatment opens up the structure of the cellulose fibres and yields a three-dimensional web-like network that forms essentially stable dispersions in water. The acquired material has been denominated 'microfibrillated cellulose' (often abbreviated to MFC), presumably after the most predominant structure encountered in the material: microfibrils and aggregates thereof. These form disordered entangled networks via the formation of junction zones, thereby providing gel-like characteristics to the material (*i.e. G' > G''*). Concentrations of 0.3 wt% MFC in water already yield stable dispersions that exhibit pseudoplastic shear-thinning behaviour and yield stresses (Iotti *et al.*, 2011; Lowys *et al.*, 2001).

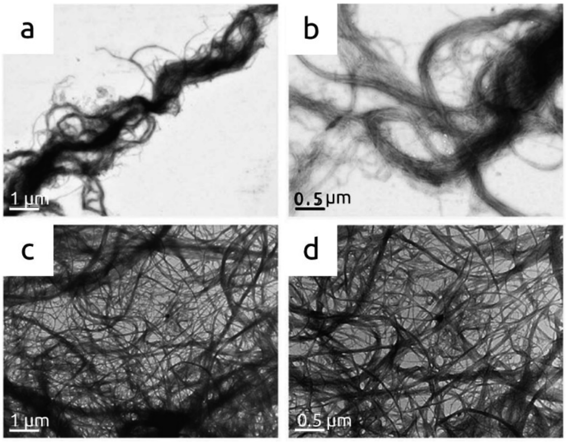

***Figure 1.2.** Transmission electron microscopy (TEM) images of native cellulose fibres (a,b) and microfibrillated cellulose dispersions (c,d) at 1 wt%, after 1,000x dilution. Image adapted from Agoda-Tandjawa et al. (2010).*

MFC consists of crystalline and amorphous regions of cellulose since its preparation does not involve chemical treatment, and the initial degree of polymerisation is retained. This highlights the main difference between MFC and cellulose whiskers (sometimes denominated nanowhiskers or nanocrystals) which are produced by hydrolysis of amorphous cellulose, yielding highly crystalline rods with lengths of several hundreds of nanometres (Siró & Plackett, 2010). Fibrillation of cellulose reduces the diameter of cellulose fibres while its length is preserved, resulting in higher aspect ratios compared to the initial material. The disintegration of cellulosic material substantially increases its surface area and thereby its reactivity and water holding capacity. It should be stressed that MFC does not only comprise entangled individualised microfibrils, but rather encompasses an inhomogeneous mixture of microfibrils, microfibrillar aggregates and occasionally entire fibres (Figure 1.3) (Chinga-Carrasco, 2011). As complete deconstruction of cellulose fibres into individual microfibrils is usually not achieved by mechanical treatment, MFC typically has a very wide size distribution.

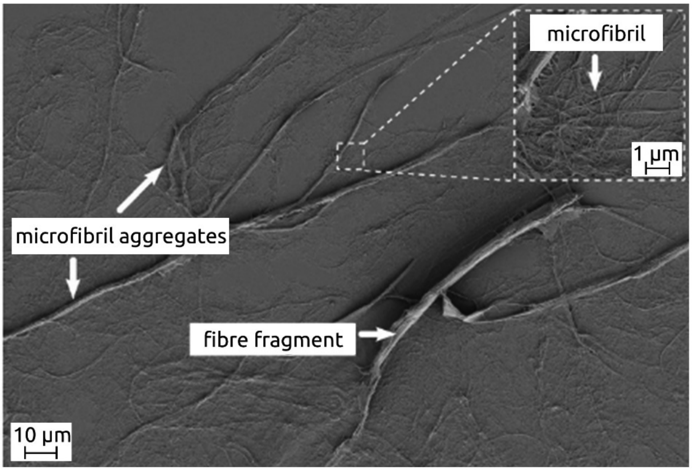

Figure 1.3. *Microfibrillation of cellulose yields material with an inhomogeneous size distribution that includes individual microfibrils, microfibril aggregates and (fragments of) fibres. Adapted from Chinga-Carrasco (2011).*

Highly purified cellulose can be used for the production of MFC. However, using minimally purified cell wall materials could be more desirable in terms of naturalness and sustainability. Depending on the source and the purification process, cell wall material contains varying amounts of residual hemicellulose, pectin and lignin. Hemicellulose can facilitate the production of MFC by acting as a steric and/or electrostatic barrier. The presence of charged hemicelluloses inhibits interfibrillar interactions and thereby enhances cellulose fibre disintegration (Lavoine *et al.*, 2012; Siró & Plackett, 2010). Pectin has been found to increase tensile strength of MFC networks by interconnecting microfibrils, thereby enhancing the material's cohesion (Siró & Plackett, 2010). Having the same chemical structure as native (untreated) cellulose, MFC can be considered an environmentally friendly material due to its renewability, biodegradability and absence of cytotoxicity and genotoxicity (Gómez *et al.*, 2016). Its water insoluble nature moreover makes MFC relatively inert toward changes in pH and temperature (Agoda-Tandjawa *et al.*, 2010), which facilitates the use of MFC in food applications. Tolerance for such environmental conditions however depends on the charge density of the microfibrillated material, as highly charged MFC is more sensitive to changes in acidity and ionic strength (Aaen *et al.*, 2019).

1.2.3. Different sources of microfibrillated cellulose

Although the first patent (1983) about MFC aimed at disentangling cellulose fibres from wood pulp, subsequent studies examined fibrillation of cell wall materials from other plant sources. MFC from wood pulp is customarily manufactured from the secondary cell wall, whereas primary cell wall material is used for the preparation of MFC from non-wood plant sources. The secondary cell wall constitutes up to 40-50% cellulose, between 25-35% hemicelluloses and 15-30% lignin (Wüstenberg, 2014). The exact composition varies depending on the type of tree, but also on its maturity, geographical location and environmental conditions. The presence of these polysaccharides affects interactions between cellulose microfibrils and ultimately influences the (degree of) fibrillation of the acquired MFC. The presence of lignin for instance hampers cellulose fibre disintegration and yields MFC with larger diameters, which can affect the functional properties of the acquired material. In contrast to the secondary cell wall of wood, the primary plant cell wall generally contains only minor quantities of lignin, if any. Instead its pectin content is significantly higher than that of wood (Nechyporchuk *et al.*, 2016). The lower lignin content and solubility of pectin facilitate cellulose disintegration, resulting in lower energy demands for fibrillation of primary cell wall material (Lavoine *et al.*, 2012; Nechyporchuk *et al.*, 2016).

Agricultural crops and their by-products can be used as a more sustainable source of raw material for the production of MFC. Therefore, researchers have examined characteristics of MFC prepared from various agricultural residues, including banana rachis (Velásquez-Cock *et al.*, 2019), sugar beet pulp (e.g. Agoda-Tandjawa *et al.*, 2010; Dinand *et al.*, 1999; Lowys *et al.*, 2001) and carrot pulp (Siqueira *et al.*, 2016). Even within the non-wood plant category, morphological and functional properties of MFC depend on the type of plant used. In addition to differences in residual hemicellulose and pectin content, degree of polymerisation (DP), cellulose crystallinity, fibre length and diameter vary depending on the type of plant material used for fibrillation. Differences in cellulose morphologies between plant varieties translate into different properties of the resultant MFC.

Specific bacteria are able to synthesise bacterial cellulose, the most well-known being *Komagataeibacter xylinus* (formerly known as *Acetobacter xylinus*) (Klemm *et al.*, 2005; Shi *et al.*, 2014). Although celluloses from bacterial and plant origin have identical molecular structures, fundamental differences exist in their supramolecular structure. Bacterial cellulose typically has a degree of polymerisation between 2,000-10,000 and high crystallinity (between 60-90%), which imparts impressive mechanical and reinforcing properties (Klemm *et al.*, 2005; Klemm *et al.*, 2011). Whereas cellulose in plants prevails in a heterogeneous mixture of compounds (*i.e.* hemicellulose, pectin and lignin), bacterial cellulose is produced as an essentially pure compound. Unlike cellulose from plants, which is bundled into fibres with diameters up to 100 µm, bacterial cellulose has diameters in the nanometer scale (Klemm *et al.*, 2005; Shi *et al.*, 2014). Bacterial cellulose therefore has a large surface area which yields properties similar to those of MFC.

1.2.4. Preparation methods for microfibrillated cellulose

The authors of the first patents describing the preparation of MFC (Herrick *et al.*, 1983; Turbak *et al.*, 1983a) specifically suggested the use of a high pressure homogeniser for converting cellulose fibres into nano-sized microfibril aggregates. Although this equipment is still frequently used for manufacturing MFC (e.g. Agoda-Tandjawa *et al.*, 2010; Henriksson *et al.*, 2007), conventional MFC production methods currently include the use of microfluidizers (e.g. Aulin *et al.*, 2009; Pääkkö *et al.*, 2007; Zimmermann *et al.*, 2004) and grinders (e.g. Abe *et al.*, 2007; Iwamoto *et al.*, 2007). One of the advantages of microfluidization is that it facilitates the production of MFC with more homogeneous fibre size. However, microfluidizers are sensitive to clogging and are therefore less suitable for materials with long fibres. Another disadvantage of this type of equipment is the large number of passes (between 10-30) required to achieve the appropriate level of fibrillation. By contrast, only 5 passes are required for fibrillation of cellulose using a grinder (Iwamoto *et al.*, 2007). As the distance between the grind stones can be adjusted, problems with clogging can be prevented. Although this method efficiently reduces the fibre diameter of the material, it is suspected that grinding also causes degradation by decreasing the fibre length. A reduction in degree of polymerisation will affect the functionality of the MFC, such as its rheological and reinforcing properties. In addition to these conventionally used techniques numerous alternative methods for MFC production have been explored in the last decades, including the use of a high-speed blender (e.g. Uetani & Yano, 2011), ball milling, cryo-crushing, extrusion, ultrasonication and steam explosion (Nechyporchuk *et al.*, 2016). It should be noted that direct comparison of different fibrillation methods is difficult, as the raw material and any optional pre-treatments have a major effect on the size of the microfibrillar fragments and the associated energy consumption. Irrespective of the mechanical treatment used, increasing the number of passes through the equipment significantly improves fibrillation of the cellulose fibres (Nakagaito & Yano, 2004; Spence *et al.*, 2011). A higher degree of cellulose delamination promotes the number of entanglements between fibres, resulting in enhanced network strength and viscosity (Aulin *et al.*, 2010).

One of the main challenges that has prevented the use of MFC in food applications is the large amount of energy that is required for its preparation. Energy inputs reaching up to 70,000 kWh per tonne MFC have been reported for fibrillation of wood pulp using homogenisation (Eriksen *et al.*, 2008). One strategy to reduce the energy consumption of the fibrillation process is by introducing a pre-treatment. Pre-treatments generally aim at reducing the level of cohesion between microfibrils, thereby facilitating structural disruption of the cellulose fibre. Oxidation of cellulose with 2,2,6,6-tetramethylpiperidine-*N*-oxyl (TEMPO) is the most commonly used pre-treatment and operates by introducing a negative charge on the cellulose molecule (Lavoine *et al.*, 2012). The repulsive forces cause the cellulose microfibrils to separate from each other, which facilitates delamination of the cell wall material. TEMPO-mediated oxidation can reduce energy consumption by a factor 100-200 and yields MFC with a narrow size distribution (Isogai *et al.*, 2011). The higher charge density of TEMPO-mediated MFC on the other hand makes the acquired material more sensitive to changes in ionic strength and pH (Aaen *et al.*, 2019). Alternatively, enzymatic pre-treatments can be performed in which enzymes hydrolyse part of the cellulose chains (Lavoine *et al.*, 2012). Such pre-treatments are particularly beneficial when MFC preparation is performed using a homogeniser or microfluidizer, as the fibre length reduction will prevent clogging of the equipment. Although MFC obtained following enzymatic pre-treatment was found to have relatively high aspect ratio (Henriksson *et al.*, 2007; Pääkkö *et al.*, 2007), the lower degree of polymerisation (DP) resulting from hydrolysis will impart inferior mechanical properties. It can be concluded that the final morphology and functionality of MFC is determined not only by the type of raw material used, but also by the number and type of purification steps, pre-treatment (mechanical/chemical) and principal (mechanical) treatment performed (Nechyporchuk *et al.*, 2016).

1.2.5. Microfibrillated cellulose in foods

Several of the patents by Turbak and colleagues highlight the potential application of MFC in foods (Turbak *et al.*, 1982, 1983b, 1984). Proposed applications include salad dressings, soups, meat products and desserts. One of the advantages of MFC is that it could facilitate reducing the energy density of foods by acting as a texture modifier in low-fat foods (Gómez *et al.*, 2016). For this purpose, Heggset *et al.* (2020) studied the effect of MFC on stability and rheology of mayonnaises with reduced oil or starch content. The authors concluded that the viscosity loss induced by a 11% oil or 50% starch reduction could be regained by addition of 0.42-0.75 wt% MFC, without compromising the stability of the mayonnaises. Golchoobi and colleagues (2016) moreover concluded that low-fat mayonnaise with 1 wt% MFC was hedonically similar to commercial low-fat mayonnaise. Addition of MFC to low-fat and full-fat ice cream improved ice cream properties, including rheological and sensory properties (Velásquez-Cock *et al.*, 2019). Higher concentrations of MFC caused increases in consistency index *K* and shear thinning behaviour, and resulted in smoother and creamier ice creams. MFC has furthermore been demonstrated to improve appearance and loaf volume of breads and to enhance water retention capacity in meat products (Ström *et al.*, 2013). Besides its texture-modifying features, MFC has been applied in foods for its emulsifying properties. Substitution of egg yolk as a surfactant by 0.05-0.1 wt% MFC resulted in stable mayonnaises that were still sensorially accepted after 4 weeks of storage (Choublab & Winuprasith, 2018). It is hypothesised that MFC acts as a Pickering emulsifier, as it adsorbs at the oil-water interface and prevents coalescence of oil droplets by steric hindrance (Winuprasith & Suphantharika, 2015). The potential of MFC as a functional ingredient in foods has been acknowledged by food industry, as evidenced by the number of patents that have been filed regarding the use of MFC in foods (Table 1.1). While several studies have examined the effect of MFC on rheological and structural properties of foods, a fundamental understanding of the tribological and sensory properties evoked by MFC is lacking. In contrast to the relationships between structural and rheological properties of foods to which MFC has been added, the relationships between structural, rheological and tribological properties on the one side and sensory perception of MFC on the other side are yet to be explored. By correlating physico-chemical properties of MFC to sensory properties, a better understanding of the factors driving texture perception of MFC could be gained.

Table 1.1. *Overview of patents filed for the use of microfibrillated cellulose (MFC) in foods.*

Type of product	**Authors**	**Patent number**
In situ preparation in foods	Turbak *et al.* (1982)	US4341807
Dressings, meat products	Turbak *et al.* (1983b)	US4378381
Dressings, meat products	Turbak *et al.* (1984)	US4464287
Bean jam	Mizuguchi *et al.* (1983a)	JP58190352
Sauces & soy soup	Mizuguchi *et al.* (1983b)	JP58190369
Retort food	Mizuguchi *et al.* (1983c)	JP58190382
Fruit fillings	Kleinschmidt *et al.* (1988)	US4774095
Dried MFC in foods	Cantiani *et al.* (2002)	US6485767
Gel foods	Oshita and Omoto (2003)	JP2004344042
Ice cream	Yano *et al.* (2016); Yano *et al.* (2014)	JP5538500, US9271514
Food preservative	Haggblom and Nordstrom (2016)	US20160088869
Plant-based beverage	Lemmers *et al.* (2017)	EP3157358
Particulate instant food	Suijker *et al.* (2017)	AU2017273661
Milk tea beverage	Koppert and Velikov (2018)	US20150335040
Edible concentrate	Suijker *et al.* (2020)	AU2018219383

1.2.6. Dehydration of microfibrillated cellulose dispersions

Despite growing evidence for the positive effects of MFC on food texture and stability and a series of patents claiming the use of MFC in foods (Table 1.1), to the best of our knowledge the commercial use of MFC in foods has so far been very limited. The use of MFC in food industry may be impaired by the fact that MFC networks are formed *in situ* at low concentrations in water (<2 wt%). The low solids content of MFC dispersions jeopardises application of MFC in foods, as its use considerably increases the water content of the final food product. The large volume occupied by aqueous MFC dispersions gives rise to inefficient transport and storage practices, which in turn increases costs. Aqueous dispersions of MFC are furthermore prone to microbial spoilage (Béguin & Aubert, 1994). Reduction of water activity of MFC dispersions could extend the material's shelf life. This has raised interest in exploring dehydration of microfibrillated materials, and several attempts have been made to dry MFC. Dehydration of MFC, however, induces irreversible binding of microfibrils and the formation of agglomerates, a process called hornification (Déléris & Wallecan, 2017). Water removal promotes the formation of hydrogen bonds with adjacent microfibrils as a result of the extensive number of hydroxyl groups of cellulose. The aggregated microfibrils are not separated upon resuspension of the dried material, leading to an overall loss in functionality compared to non-dried material (Silva *et al.*, 2021). Especially deterioration of the material's water absorption and swelling capacity are observed after reconstitution (Déléris & Wallecan, 2017). Several methods have been explored to prevent hornification, including surface modification and dehydration of MFC in the presence of other compounds. Both methods aim at preventing the formation of hydrogen bonds between cellulose molecules by steric or electrostatic repulsion. Redispersibility and rheological properties of the reconstituted material were improved (Eyholzer *et al.*, 2010; Missoum *et al.*, 2012; Yan *et al.*, 2016) and in some cases yielded properties similar to those of non-dried MFC (Butchosa & Zhou, 2014; Lowys *et al.*, 2001; Velásquez-Cock *et al.*, 2018). The use of dehydrated MFC powders instead of aqueous MFC dispersions might facilitate incorporation of MFC in foods and may therefore be more promising. So far, the majority of studies on dehydrated MFC focused on its redispersibility and functional properties, while its sensory and tribological properties in foods have never been examined. In particular, it remains unknown how the morphology of dehydrated MFC contributes to rheological, tribological and sensory properties of foods, which might currently limit its applicability.

1.3. Sensory properties of foods

Sensory properties of foods are a major determinant of food liking and are therefore highly relevant for both food manufacturers and consumers (Andersen *et al.*, 2019). Although all sensory modalities are involved in food perception (vision, taste, smell, touch, hearing), generally the focus is on appearance, flavour (combination of taste and smell) and texture. For decades, food scientists have attempted to model or predict sensory properties of foods from instrumentally determined food properties. This proves to be a challenge, especially for texture and mouthfeel of foods, and therefore sensory properties are typically assessed by a sensory panel. The type and size of the sensory panel depends on the research question and the methodology selected for sensory evaluation. Even though sensory evaluation by trained panels has traditionally been regarded as the best practice, untrained (consumer) panels have lately gained popularity as a result of their ecological validity. Several sensory evaluation methods can be performed by semi- or untrained panellists, including discrimination tests (*e.g.* triangle test, paired comparisons), scaling methods (*e.g.* magnitude estimation, rank-rating) and even some descriptive methods (*e.g.* Check-All-That-Apply, Rate-All-That-Apply, flash profiling).

1.4. Rheology in food science

During consumption the physical properties of foods change as a result of mastication, compression and saliva incorporation. Due to the change in size and mechanical properties of food, texture perception evolves continuously during this dynamic process. It is therefore believed that multiple physical factors play a role in food texture perception (Chen & Stokes, 2012). Rheology has been recognised to be of major importance in the perception of food texture, and is defined as the study of the deformation and flow of matter (Rao, 2007). Rheological properties reflect interactions at the micro- and macroscopic level and can therefore provide valuable information on the structure of foods. Various methodologies can be employed to determine a range of rheological parameters. One of the most commonly studied rheological properties is shear viscosity η, which represents the internal friction of a fluid or its tendency to resist flow (Rao, 2007). Shear viscosity can be determined using steady shear rheology (Figure 1.4a) and is described as the shear stress σ divided by shear rate $\dot{\gamma}$. The majority of foods behave as non-Newtonian shear thinning fluids, *i.e.* their viscosity decreases with increasing shear rate as a result of breakdown or reorientation of dissolved or suspended compounds. Moreover, many foods are semi-solids that to some extent display both viscous and elastic behaviour. The viscoelastic properties of foods can be characterised using dynamic shear rheology (Figure 1.4b) and the elastic and viscous components are typically expressed by storage modulus G' and loss modulus G'', respectively. In addition to shear rheology, extensional rheology can be studied to collect information about extensional or elongational viscosity η_e (Figure 1.4c). Extensional deformation occurs for instance during swallowing of food and is therefore relevant in food science.

Food texture is largely determined by rheological properties of foods during the first stages of oral processing (Stokes *et al.*, 2013). A large number of studies have established correlations between rheological and sensory properties of foods, for example in bread, cookies, dairy products, fruits and meats (Joyner, 2018). The vast majority of publications in the 1970's and 80's focused on the relation between perceived thickness and rheological parameters (e.g. Cutler *et al.*, 1983; Kokini *et al.*, 1977; Shama & Sherman, 1973). It is now well established that an increase in shear viscosity is reflected in higher thickness intensities, which is often accompanied by an increase in creaminess. Although creaminess is a multi-modal sensory attribute that is affected by gustatory, olfactory and tactile cues, a positive correlation between creaminess and viscosity has been reported by numerous studies (e.g. Akhtar *et al.*, 2005; Daget & Joerg, 1991; van Aken *et al.*, 2011). More viscous foods moreover give rise to increased sliminess (Brandenstein *et al.*, 2015; Lyly *et al.*, 2003), stickiness and mouthcoating texture (He *et al.*, 2016; Ross *et al.*, 2019). Even though literature relating extensional rheology to sensory properties is limited, positive correlations between extensional viscosity and stickiness and mouthcoating texture have been reported (He *et al.*, 2016).

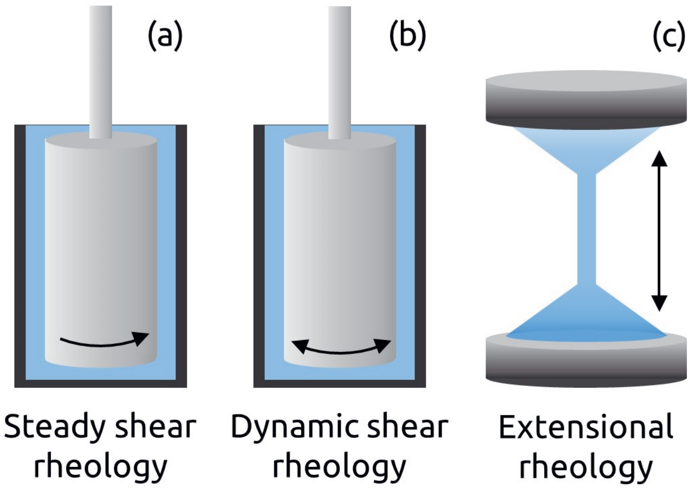

Figure 1.4. *Schematic representation of different types of rheology and how these are performed.*

1.5. Tribology in food science

While texture perception is dominated by rheology during the first stages of oral processing, tribology becomes important at later stages when the layer of food between palate and tongue becomes smaller (Stokes *et al.*, 2013). Tribology is the study of friction, lubrication and wear between two interacting surfaces that are in relative motion. The human tongue and palate can be considered as two moving surfaces that are lubricated by the food bolus during oral processing. In the last stages of oral processing the tongue and palate come into closer contact with each other, emphasising the importance of tribology in this stage of food consumption.

The main parameter used to describe tribological properties is the friction coefficient μ, which is defined as the ratio of friction force ($F_{friction}$) to the applied load (normal force F_N). The friction coefficient is typically measured as a function of sliding speed and displayed in a so-called Stribeck curve, from which three friction regimes can be distinguished (Figure 1.5) (Pradal & Stokes, 2016). The boundary regime prevails at low sliding speeds, where the two tribological surfaces are in close contact. Friction in this regime arises from direct contact between the surfaces and high friction coefficients are generally observed. Boundary friction is primarily dependent on surface properties and is only affected by the lubricant as a result of lubricant adsorption at the tribological surface. The presence and extent of such boundary surface films depends on the lubricant and the surface material (Stokes *et al.*, 2011). Upon increasing the speed the mixed regime is entered, in which a thin layer of lubricant is present between the two moving surfaces. Although the tribological surfaces are slightly more separated compared to the boundary regime, there is still some contact between the surface asperities. Therefore, the friction in this regime depends on both the surface properties and the lubricant's viscosity. The mixed regime is characterised by a decline in friction coefficient with increasing speed until a minimum is reached. This is the intersection between the mixed and hydrodynamic regime and represents maximum lubrication. The hydrodynamic regime occurs at high sliding speeds, in which the surfaces are fully separated by the lubricant. The friction in this regime is completely dependent on the lubricant's viscosity, and the distance between the surfaces is determined by the hydrodynamic pressure exerted by the lubricant. The thickness of the layer separating the two surfaces increases at higher entrainment speeds. The drag induced by the increasing layer thickness causes friction in this regime to increase with increasing speeds. Early stages of food oral processing are generally dominated by high oral speeds, while lower speeds prevail at later stages in which the thickness of the lubricant layer is reduced. The transition that food undergoes during consumption is thus represented by a Stribeck curve read backwards, starting with the hydrodynamic regime, followed by the mixed and boundary regime.

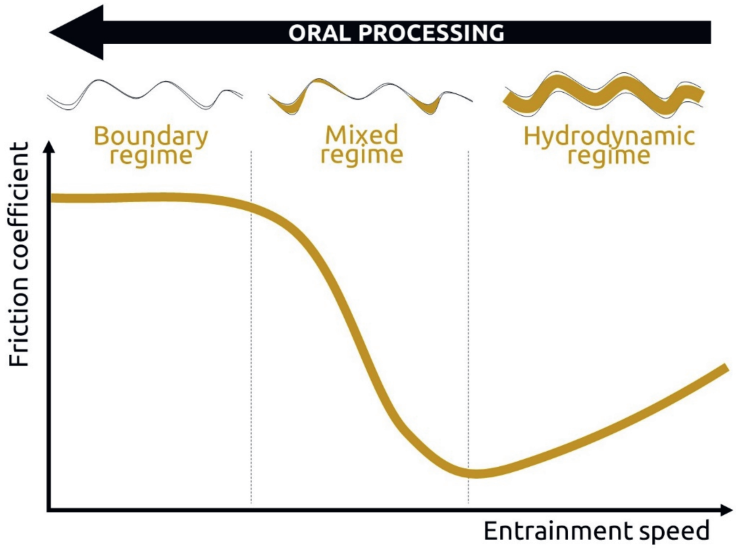

Figure 1.5. *The friction coefficient is typically plotted against the entrainment speed in a so-called Stribeck curve, in which the boundary, mixed and hydrodynamic regime can be distinguished.*

As friction is considered to be one of the physical properties contributing to food texture perception, several studies have attempted to relate tribological parameters to specific sensory texture attributes (Sarkar & Krop, 2019). For instance, positive correlations were found between friction in the mixed regime and sensory perception of stickiness (Devezeaux de Lavergne *et al.*, 2016), slipperiness or salivating (Krop *et al.*, 2019), whereas negative correlations were found with sensory smoothness (Upadhyay & Chen, 2019) and pastiness (Krop *et al.*, 2019). While these researchers used model foods to relate friction to perception, most researchers that studied actual foods addressed fat-related texture attributes (Sarkar & Krop, 2019). The majority of these studies focused on dairy products and found negative correlations between friction and creaminess (e.g. Chojnicka-Paszun *et al.*, 2012; de Wijk & Prinz, 2005; Laiho *et al.*, 2017). Laguna *et al.* (2017) concluded that full-fat and fat-free dairy products (*i.e.* milk, yoghurt, cream cheese) could be sensorially distinguished based on tribological differences between the products. Although many friction-texture correlations have been reported in recent years, the underlying physical aspects responsible for such correlations are often not elucidated. It therefore remains challenging to translate any correlations between tribological properties and texture perception to other types of foods. The fact that researchers use different tribological set-ups and that none of these has been universally acknowledged as a gold standard moreover makes it difficult to generalise the results across studies.

1.6. Aim and outline of this thesis

As evidenced by numerous patents on MFC (Table 1.1), its potential to be used as a functional food ingredient has been recognised by food industry. The majority of studies on MFC focused on its effect on the rheological properties or stability of the foods it was added to. However, a structured assessment of the tribological and sensory properties that characterise foods thickened with MFC is lacking. Moreover, it is not known how the rheological and tribological properties elicited by addition of MFC to foods contribute to the way these foods are perceived sensorially. The primary aim of this thesis was to assess the suitability of MFC as a food thickener by comparing its effect on sensory, rheological and tribological properties of liquid and semi-solid foods to other common thickeners. This thesis further aims at correlating these sensory properties to instrumental physico-chemical parameters to elucidate mechanisms underlying their perception. In addition to studying dispersed MFC fibrils similar to those described in the original patents from 1983, spray-dried MFC powders are examined in this thesis. Since the main focus is on understanding (*unravelling*) the mechanisms behind the sensory, rheological and tribological properties of microfibrillated (*unravelled*) cellulose, it may be stated that the thesis attempts *to unravel the unravelled*.

A schematic overview of the framework of this thesis is presented in Figure 1.6. **Chapter 2** examines the effect of MFC on rheological, tribological and sensory properties of foods in a fundamental way. A wide range of MFC concentrations (0.2-2.0 wt%) are used to prepare simple aqueous liquid model foods to limit the effect of potential interactions with other ingredients. These aqueous model foods are compared to iso-viscous xanthan gum solutions, as this thickener is frequently used by food manufacturers. The liquid model foods are compared for sensory (appearance, flavour, texture), rheological (shear viscosity, dynamic moduli, yield stress, extensional viscosity) and tribological properties. In a continuation of this approach, **Chapter 3** describes the application of MFC as a thickener in fat-containing semi-solid foods that are more representative of commercial products. MFC is applied in low-fat mayonnaises at several concentrations and compared to low-fat mayonnaises thickened with other commonly used thickening agents: native waxy corn starch, chemically modified corn starch and xanthan gum. In addition to sensory, rheological and tribological properties, the microstructure of the low-fat mayonnaises is studied using confocal laser scanning microscopy (CLSM). Since the high water content of MFC dispersions undermines efficient transport and storage, it was explored whether MFC can be spray-dried into dry MFC which displays similar functionality upon re-suspension as dispersed MFC fibrils. **Chapter 4** uses dehydrated MFC obtained by spray-drying MFC in the presence of maltodextrin, and examines its texture modifying properties. This chapter studies the effect of these spray-dried MFC particles on rheological, tribological and sensory properties (thickness and creaminess) of milk and instant soup. As **Chapter 2-4** assess the potential effect of MFC on texture attributes such as creaminess, **Chapter 5** aims at improving our understanding about the mechanisms underlying the perception of creaminess. An attempt is made to disentangle the relative contributions of viscosity and friction on creaminess, two physical parameters postulated to affect sensory creaminess. In addition to sensory, rheological and tribological characterisation of (model) foods thickened by MFC and comparison to other thickeners, **Chapters 2-5** explore correlations between physico-chemical food properties (rheological, tribological and microstructural properties) and sensory properties. Lastly, **Chapter 6** provides an integrated general discussion about the results obtained in **Chapters 2-5**. It moreover reflects on the methodologies used throughout the thesis and discusses future applications of MFC.

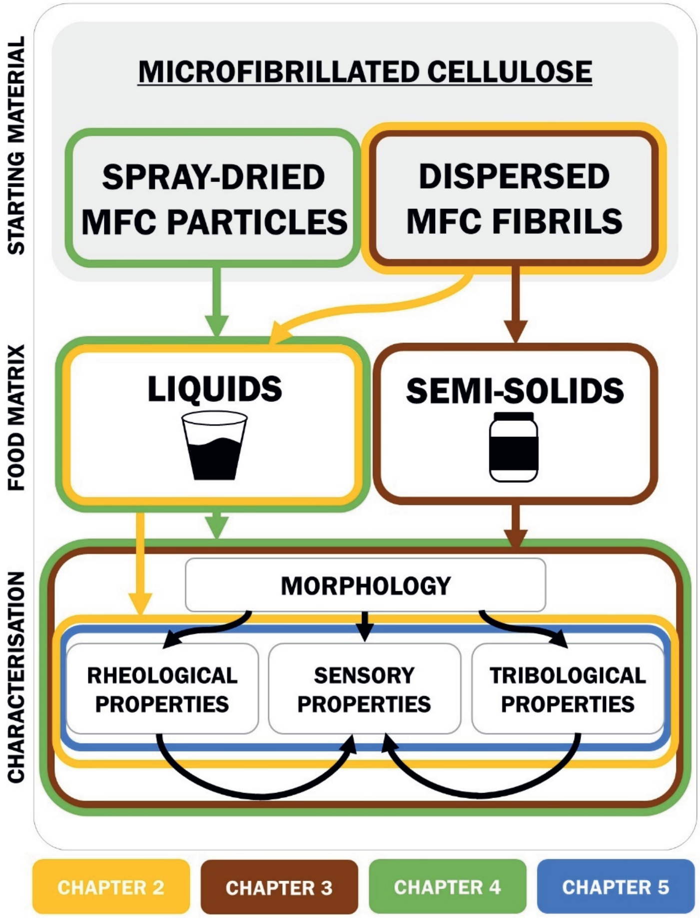

Figure 1.6. *Schematic overview of the framework of this thesis. This includes the different types of microfibrillated cellulose (dispersed vs spray-dried MFC), different types of products and parameters studied in this thesis.*

2

Comparing rheological, tribological and sensory properties of microfibrillated cellulose dispersions and xanthan gum solutions

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Food Hydrocolloids (2021), 121: 107052

Abstract

Utilisation of plant waste materials contributes to sustainable food production and allows preparation of functional ingredients from natural bio-materials. Microfibrillated cellulose (MFC) from plant waste materials such as citrus peels has been suggested to have potential as ”clean label” thickener. This study compared rheological (shear and extensional rheology, hysteresis, yield stress), tribological and sensory properties of MFC dispersions (0.2-2.0 wt%) to xanthan gum (XG) solutions (0.04-4.3 wt%) and linked sensory characteristics to instrumental properties. Concentrations of MFC and XG were chosen so that shear viscosities of MFC dispersions and XG solutions were similar over a large range of shear rates. XG had higher extensional viscosity at high deformation rates than MFC. XG had higher yield stress than MFC at similar shear viscosity. Yield stress increased linearly with increasing concentrations for XG, while it increased exponentially for MFC. Seventy-three consumers evaluated the appearance, flavour, and mouthfeel of all samples using the Rate-All-That-Apply (RATA) method. Sensory differences between MFC and XG were generally larger at higher concentrations. MFC dispersions were less transparent and had more intense cardboard flavour than XG solutions of comparable shear viscosity. At high thickener concentrations, XG solutions were perceived as glossier, stickier, slimier and more mouthcoating than MFC dispersions of similar shear viscosity. Sticky, slimy and mouthcoating perception were correlated with extensional viscosity at higher deformation rates. We conclude that MFC can thicken foods similar to XG while avoiding undesired texture sensations such as mouthcoating, sliminess and stickiness. The flavour and dispersibility of MFC need to be improved further before it can be applied as thickener in foods.

2.1. Introduction

A large variety of thickening agents is used in foods to increase viscosity, as a stabiliser or to improve texture and mouthfeel perception. Xanthan gum (XG) is one of the most widely used hydrocolloid thickeners because of its temperature and pH stability, pseudoplastic rheological properties and its ability to stabilise emulsions (García-Ochoa *et al.*, 2000). XG is used for instance in salad dressings, confectionery, tooth paste and gluten-free baked goods. It is furthermore used to increase the viscosity of foods for dysphagia patients, *i.e.* individuals that have difficulty swallowing fluids and foods (Althaus, 2002). XG is a water-soluble, natural biopolymer produced by the bacteria *Xanthomonas campestris*, that can thicken foods already at low concentration (García-Ochoa *et al.*, 2000). However, liquids thickened with XG have been described as adhesive (Ong *et al.*, 2018b), slimy (Gössinger *et al.*, 2018) and sticky (Yamagata *et al.*, 2012). Sliminess and stickiness are generally disliked sensory attributes in liquid foods (Pellegrino & Luckett, 2020; Saluja & Stevenson, 2019), which calls for the use of alternative thickeners.

One type of hydrocolloid that has recently been applied as a thickener in foods is microfibrillated cellulose (MFC). Microfibrillated cellulose is a type of nanocellulose produced by mechanically disintegrating cellulose originating from *e.g.* wood (Saito *et al.*, 2006; Spence *et al.*, 2010; Stenstad *et al.*, 2008; Taipale *et al.*, 2010), sugar beets (Agoda-Tandjawa *et al.*, 2010; Dinand *et al.*, 1999), carrots (Siqueira *et al.*, 2016) or fruits (Habibi *et al.*, 2009; Jongaroontaprangsee *et al.*, 2018; Pelissari *et al.*, 2014; Winuprasith & Suphantharika, 2013). In order to manufacture MFC, cellulose is subjected to homogenisation at high shear, thereby moderately degrading the original structure of the cellulose (Gómez *et al.*, 2016; Lavoine *et al.*, 2012; Turbak *et al.*, 1983a). As a consequence, a highly expanded network of aggregated cellulose microfibrils is formed that has a large surface area. MFC forms gels at low concentrations (Iotti *et al.*, 2011; Lowys *et al.*, 2001; Pääkkö *et al.*, 2007), displays shear-thinning behaviour (Iotti *et al.*, 2011; Pääkkö *et al.*, 2007) and has improved water retention capacity (Gómez *et al.*, 2016; Lavoine *et al.*, 2012). The process of producing MFC has first been patented in 1983 by Turbak *et al.* (1983a). The functional and physicochemical properties of MFC have since then been explored in a broad range of foods including bread dough (Ström *et al.*, 2013), hamburgers (Ström *et al.*, 2013), ice cream (Velásquez-Cock *et al.*, 2019), and mayonnaises (Choublab & Winuprasith, 2018; Golchoobi *et al.*, 2016; Heggset *et al.*, 2020). In addition to its high abundance and biodegradability, cellulose is a promising raw material to produce thickeners since it can be obtained from by-products and waste materials from agricultural crops. Depending on the treatment and source of cellulose, properties such as fibre length, aspect ratio, degree of crystallinity and amounts of residual hemicellulose or lignin of MFC vary (Lavoine *et al.*, 2012).

Despite the large amount of patents on the application of MFC in foods since its invention in the 1980s (e.g. Kleinschmidt *et al.*, 1988; Koh & Hayama, 1997; Koppert & Velikov, 2018; Lemmers *et al.*, 2017; Weibel, 2001; Yano *et al.*, 2016), research on the sensory properties of fluid foods thickened with MFC is limited. Golchoobi and co-workers concluded that addition of MFC to low-fat mayonnaises did not negatively affect hedonic ratings of taste, colour, odour, texture and acceptability (Golchoobi *et al.*, 2016). Choublab & Winuprasith reported that hedonic ratings of appearance, texture, flavour and overall acceptability of mayonnaises emulsified by MFC decreased as the concentration of MFC increased (Choublab & Winuprasith, 2018). Velásquez-Cock *et al.* (2019) recently demonstrated that addition of MFC to ice cream improved the texture compared to ice cream without MFC.

To summarise, the sensory properties of liquids thickened with MFC have so far not been compared to other biopolymers and linked to rheological and tribological properties. This study examined whether MFC can possibly be used as a thickener in foods and thereby replace currently used thickeners such as xanthan gum, while retaining sensory and functional properties of the foods. The aim of this study was to compare rheological, tribological and sensory properties of MFC dispersions to iso-viscous XG solutions, and to link the sensory properties of these model foods to rheological and tribological properties. Concentrations of MFC dispersions and XG solutions were selected so that shear viscosities (flow curves) matched over a wide range of shear rates. Shear and extensional viscosity, hysteresis, yield stress, friction and sensory properties were determined.

2.2. Materials & Methods

2.2.1. Sample preparation

Preparation of microfibrillated cellulose dispersions

Six dispersions of citrus fibre (HERBACEL® AQ® Plus, HerbaFood, Germany) differing in concentration (0.2, 0.3, 0.5, 1.0 1.5 and 2.0 wt%) were prepared in Milli-Q water. All samples were adjusted to pH ~ 4 using 1M food-grade HCl (Sigma-Aldrich, USA). The citrus fibre powder was first suspended in deionised water and thoroughly mixed using a L5M-A Silverson laboratory mixer (Silverson Machines Ltd., UK) with a 1 mm screen hole at 3,000 rpm for 10 min and afterwards passed twice through a high-pressure homogeniser (Microfluidizer M-110S, Microfluidics™, USA) with a z-shape geometry (ø 87 μm) operating at a pressure of 1200 bar.

All samples were sterilised at 125°C for 15 min in a steam steriliser autoclave in 500 mL flasks. Following this procedure microfibrillated cellulose was obtained consisting of fibres with a diameter of 3-4 nm, which is the size of elementary cellulose fibrils in primary cell walls (Chinga-Carrasco, 2011). The length of individual MFC fibrils can extend up to several micrometers, typically up to 10 μm (Agoda-Tandjawa *et al.*, 2010; Hayden *et al.*, 2019; Nomena *et al.*, 2018) and these fibrils form an attractive network.

Preparation of xanthan gum solutions

Six solutions of xanthan gum (Jungbunzlauer, Switzerland) differing in concentration (0.04, 0.10, 0.21, 2.0, 3.4 and 4.3 wt%) were prepared by dissolving XG powder in water at room temperature and stirring for at least 30 min. Xanthan gum (XG) concentrations were selected so that their shear viscosities matched those of the six MFC dispersions over a large range of shear rates. XG solutions were freshly prepared on the day of use.

Preparation of samples for instrumental measurements and sensory evaluations

MFC dispersions and XG solutions for sensory evaluations (Table 2.1) were prepared by addition of 7.5 wt% sugar, 0.1 wt% strawberry flavouring (Jo-La, Bharco Foods, the Netherlands) and 0.05 wt% red colourant (Rayner's, Healthy Food Brands, United Kingdom). The samples with the three highest concentrations of XG and MFC were prepared using a Thermomix® (Thermomix® TM5, Vorwerk, Germany) as this facilitated dissolution of the ingredients due to the high viscosity of these samples. Samples were freshly prepared each day and stored in the refrigerator at 4°C until 1h before use.

Table 2.1. *Concentrations of microfibrillated cellulose (MFC) and xanthan gum (XG) in the samples (before addition of sugar (7.5 wt%), strawberry flavouring (0.1 wt%) and red colourant (0.05 wt%)).*

Sample name	wt% MFC	Sample name	wt% XG
MFC-1	0.2	XG-1	0.04
MFC-2	0.3	XG-2	0.10
MFC-3	0.5	XG-3	0.21
MFC-4	1.0	XG-4	2.0
MFC-5	1.5	XG-5	3.4
MFC-6	2.0	XG-6	4.3

2.2.2. Rheological characterisation

Shear rheology

Rheological properties were determined using a concentric cylinder (CC17/Ti, Anton Paar, Austria) in a rheometer (MCR 302, Anton Paar, Austria). The gap size of the concentric cylinder geometry was 700 μm, which means that the gap of the concentric cylinder was more than 10 times larger than the length of individual fibrils (Agoda-Tandjawa *et al.*, 2010). After loading the sample to the concentric cylinder, samples were left for 5 min to allow for structural recovery. Except for extensional viscosity and oscillation measurements, rheological properties were determined using XG solutions and MFC dispersions to which sugar, colourant and flavouring had been added.

Shear viscosity was measured in duplicate at 35°C as a function of increasing shear rate from 1 to 1000 s^{-1} in 50 logarithmic steps. For hysteresis measurements, shear stress was measured at 35°C as a function of shear rate by first increasing the shear rate from 1 to 1000 s^{-1}, followed by decreasing the shear rate from 1000 to 1 s^{-1}. Measurements were performed in duplicate (XG solutions) or triplicate (MFC dispersions). Relative hysteresis areas were determined from the stress-strain curves as the difference in area under the curve (AUC) between the upward and downward curve divided by the AUC of the upward curve. In oscillation experiments, *G'* and *G"* were measured in duplicate at constant oscillation (1 Hz) at 35°C as a function of logarithmically increasing shear strain from 0.01-100% (1-10,000% for XG-4, XG-5 and XG-6). Values of *G'* and *G"* were determined at 1% shear strain, and yield stress was determined as the stress applied at the intersect of *G'* and *G"*.

Extensional rheology

Extensional viscosity of MFC dispersions and XG solutions was measured with a custom-built filament stretching rheometer, similar to the one described earlier (Huisman *et al.*, 2012; Kibbelaar *et al.*, 2020; Louvet *et al.*, 2014). A rheometer (MCR 300, Anton Paar, Austria) was used as the building block of the device. A speed controllable (*v*) cylindrical geometry (ø 5 mm) was used as the upper geometry and a Peltier substrate (P-PTD 200, Anton Paar, Austria) was used to impose the desired temperature to the sample. Extensional measurements were performed at 35°C. A small sample of 40 μL was initially placed between the two circular end plates (initial bridge height L_0 = 2.5 mm) which are moved apart at a constant velocity of 0.1 mm/s until the bridge breaks. Such low velocity was selected to ensure that the break up is only due to the surface tension. The evolution of the liquid bridge was recorded with a fast camera (Phantom V7) allowing frame rates up to 10,000 frames/s. The camera was coupled to a microscope tube lens, with an objective up to 12x magnification (Navitar, NY, USA) and a spatial resolution of 3 μm per pixel. The profile of the neck diameter was automatically followed in time with a homemade MATLAB routine. To avoid evaporation during the measurement, the set-up was placed in a homemade humidity chamber (80% RH). The injection of a tuneable humid air flow in the chamber allowed to suppress evaporation during measurements. Each sample was measured three times to assure repeatability of the extensional properties. The extensional viscosity was extracted from the thinning dynamics (see Supp. Material, Figure S1) using $\eta_s = 2^{n-1}n^n\varphi_0(n)\gamma[\frac{dh_{min}}{d\tau}]^{-1}$, where $\varphi_0(n)$ depends on the power law exponent (*n*) which is determined from the thinning dynamics and which is consistent with the exponent determined from the shear rheology (tabulated in Doshi & Basaran, 2004; Doshi *et al.*, 2003; Suryo & Basaran, 2006).

2.2.3. Tribological characterisation

Friction properties of the samples with added sugar, colourant and flavouring were characterised using an MCR 302 rheometer equipped with a ball-on-three-pins set-up (T-PTD-200, Anton Paar, Austria). A glass ball and polydimethylsiloxane (PDMS) pins were used. One mL of sample was transferred to the sample holder. Tribological tests consisted of three runs of 10 min, each run preceded by a resting period of 5 min. A normal force of 1 N was applied during runs and resting periods. In each run rotational sliding speeds were logarithmically increased from 0.0001 – 2200 rpm (equivalent to $4\cdot10^{-5}$ – 10^3 mm/s). Friction coefficients were obtained as the ratio of the frictional force divided by the normal load. Tests were conducted in triplicate at 35°C and data from the second run of each test were used for analysis. Pins were replaced after each replicate of the set of samples, to limit the effect of wear on the tribo-pair. Prior to measuring the samples, PDMS surfaces were run-in by (i) one run with 1 mL demineralised water and (ii) one run with 1 mL MFC-1.

2.2.4. Sensory evaluation

Participants

Dutch participants between 18-35 y were recruited from the surroundings of Wageningen. Pregnant or breastfeeding women, smokers and individuals with food allergies or intolerances to food colourants or flavourings were excluded from participation. Moreover, individuals with general or oral health problems, mastication or swallowing disorders or without normal smell and taste function were excluded. A total of *n*=73 subjects (12 male, 61 female; mean age 21.5 y; mean BMI 21.7 kg/m^2) participated in the study. Participants completed a general questionnaire before starting the sensory evaluation. Participants signed an informed consent and received financial reimbursement after completion of the test session.

Rate-All-That-Apply (RATA) method

All 12 samples were evaluated by *n*=73 participants in one test session of 60 min. Samples were monadically presented in random order and were evaluated using the Rate-All-That-Apply method (RATA). Participants were provided a list of sensory attributes of which the attributes that are applicable for the sample needed to be selected (Ares *et al.*, 2014). Subsequently, participants rated the intensity of the selected attributes on a 9-point scale (anchored *low* to *high*). Attributes were selected from a list of 17 attributes, which were divided over three categories: appearance, flavour and texture (Table 2.2). Two example questions were provided to the participants in order to become acquainted with the sensory method and to familiarise participants with the samples. Two of the actual samples (MFC-1 and XG-6) were provided to answer the example questions. Definitions of the attributes were sent to the participants by email several days prior to the test session. Participants were asked to study the attribute definitions prior to the test session and to refrain from eating and drinking one hour before the test session.

Participants were seated in individual sensory booths with standard white light. Samples (15-20 mL) were presented in random order in 30 mL transparent plastic cups labelled with random 3-digit codes. Participants were asked to use a spoon to taste the samples and were given the possibility to expectorate samples after evaluation. Crackers and water were provided for palate cleansing after evaluation of each sample. Data was collected in Dutch using Qualtrics software (Qualtrics, USA).

2.2.5. Data analysis

Results from sensory evaluation were reported as mean values with standard error. Sensory attributes that were not selected by participants were treated as an intensity value of 0. Two-way repeated measures ANOVAs (fixed factors: viscosity level, thickener type, viscosity level:thickener type interaction; random factor: participant) were performed on each attribute and Bonferroni post-hoc tests were performed to determine significant differences between samples (Kuznetsova *et al.*, 2017; Lenth, 2019). Principal Component Analysis (PCA) was performed with 95% confidence ellipses for the twelve samples (Kassambara & Mundt, 2020; Lê *et al.*, 2008). Multiple Factor Analysis (MFA) was performed to determine correlations between rheological and tribological properties and sensory attributes (Lê *et al.*, 2008). Data was analysed using RStudio (version 3.5.2) and a significance level of α = 0.05 was used.

Table 2.2. *Attributes used to evaluate microfibrillated cellulose (MFC) dispersions and xanthan gum (XG) solutions, their definitions and examples of products high in intensity of the respective attribute.*

Attribute	Definition	Examples of products
Appearance		
Glossy	A glossy, shiny appearance.	Olives, icing, custard
Red colour	The intensity of the red colour.	Strawberry, tomato
Slimy	The sample is thick, slippery and cohesive.	Gelatin pudding, oysters, raw egg white
Smooth	The texture of the sample is smooth and homogenous; absence of lumpiness and graininess.	Custard, milk, water (smooth) Cottage cheese (not smooth)
Thick	The thickness of the sample; the degree to which the sample flows.	Greek yoghurt (thick) Water (not thick)
Transparent	The degree to which it is possible to see through the sample.	Water (transparent) Milk (not transparent)
Flavour		
Cardboard/paper flavour	The degree to which the sample tastes like cardboard or paper; stale.	n.a.
Strawberry flavour	The degree to which the sample tastes like strawberry.	Strawberries, strawberry smoothie
Sweet taste	The intensity of the sweetness.	Sugar, strawberry lemonade
Texture		
Creamy	The degree to which the sample gives a silky, rich, full mouthfeel.	Ice cream, whipped cream
Melting	The degree to which the sample becomes thin and fluid and distributes itself in the mouth.	Ice cream, chocolate
Mouthcoating	The feeling that a layer of the sample remains behind in the mouth and palate (after swallowing).	Mayonnaise
Pulpy	The sample has a pulpy, mushy structure; the texture of the sample is fibre-like.	Apple sauce, orange juice with pulp
Slimy	The sample is thick, slippery and cohesive in the mouth.	Gelatin pudding, oysters, raw egg white
Smooth	The texture of the sample is smooth and homogenous; absence of lumpiness and graininess; the sample flows easily in the mouth.	Custard, milk, water (smooth) Cottage cheese (not smooth)
Sticky	The degree to which the sample sticks to the palate and teeth.	Honey, marshmallow
Thick	The thickness of the sample; the amount of force needed to make the sample flow or deform in the mouth.	Greek yoghurt (thick) Water (not thick)

2.3. Results & Discussion

2.3.1. Rheological properties

Concentrations of MFC and XG were selected so that shear viscosities matched over a large range of shear rates, which is demonstrated in Figure 2.1. At high MFC (1.0, 1.5 and 2.0 wt%) and XG concentrations (2.0, 3.4 and 4.3 wt%), shear viscosities of both thickeners matched over a broad range of shear rates (1-1000 s^{-1}). At low thickener concentrations (MFC-1/2/3 and XG-1/2/3) high shear viscosities (10-1000 s^{-1}) of MFC dispersions and XG solutions were similar, whereas small differences are observed at low shear viscosity (1-10 s^{-1}) which might be due to flow instabilities (see also Supp. Material, Figure S2). All MFC dispersions and XG solutions displayed shear thinning behaviour. As expected, shear and extensional viscosities increased with increasing thickener concentrations (Lundahl *et al.*, 2018; Martín-Alfonso *et al.*, 2018; Moberg *et al.*, 2014). At low concentrations of MFC and XG (0.2-0.5 wt% MFC; 0.04-0.21 wt% XG), shear viscosities were notably lower than extensional viscosities. This finding reflects earlier work, in which extensional viscosities of MFC dispersions were much higher than their corresponding shear viscosities (Moberg *et al.*, 2014). Consequently, Trouton ratios (η_{ext}/η_{shear}) of the samples with the lowest viscosities (MFC-1/2/3 and XG-1/2/3) were considerably larger than 3, emphasising the elastic nature of the dispersions and solutions. In the current study, shear and extensional viscosities became more similar as thickener concentrations increased. Systems with higher concentrations of MFC or XG behaved more like yield stress fluids. For such yield stress fluids, in which no to little polymer stretching takes place, the extensional and shear viscosity are expected to overlap (Louvet *et al.*, 2014). At lower concentrations of MFC and XG, at which respectively polymer reorientation or stretching occurs, a more pronounced difference between shear and extensional viscosity is observed as expected. Samples thickened with MFC and XG both displayed extensional thinning behaviour over a large range of deformation rates, as has been reported previously (Lundahl *et al.*, 2018; Martín-Alfonso *et al.*, 2018; Waqas *et al.*, 2017). However, for XG solutions extensional viscosity reached a plateau at the highest deformation rates (> 100 s^{-1}). Higher extensional viscosity has been linked to reduced bolus elongation and increased cohesiveness, and might therefore facilitate swallowing and reduce the risk of asphyxiation as a consequence of bolus disintegration (Brito-de la Fuente *et al.*, 2017; Hadde & Chen, 2019; Hadde *et al.*, 2019; Tobin *et al.*, 2020). Just before the break-up of the XG solution filaments, the viscoelastic behaviour of XG becomes evident by the formation of thin elastic threads that are usually observed for flexible polymers (Deblais *et al.*, 2018). Correspondingly, an increase in the apparent extensional viscosity is observed (Figure 2.1) which is comparable to flexible polymers. This underlines a major difference between MFC and XG, which is the fact that XG can undergo polymer stretching while MFC cannot.

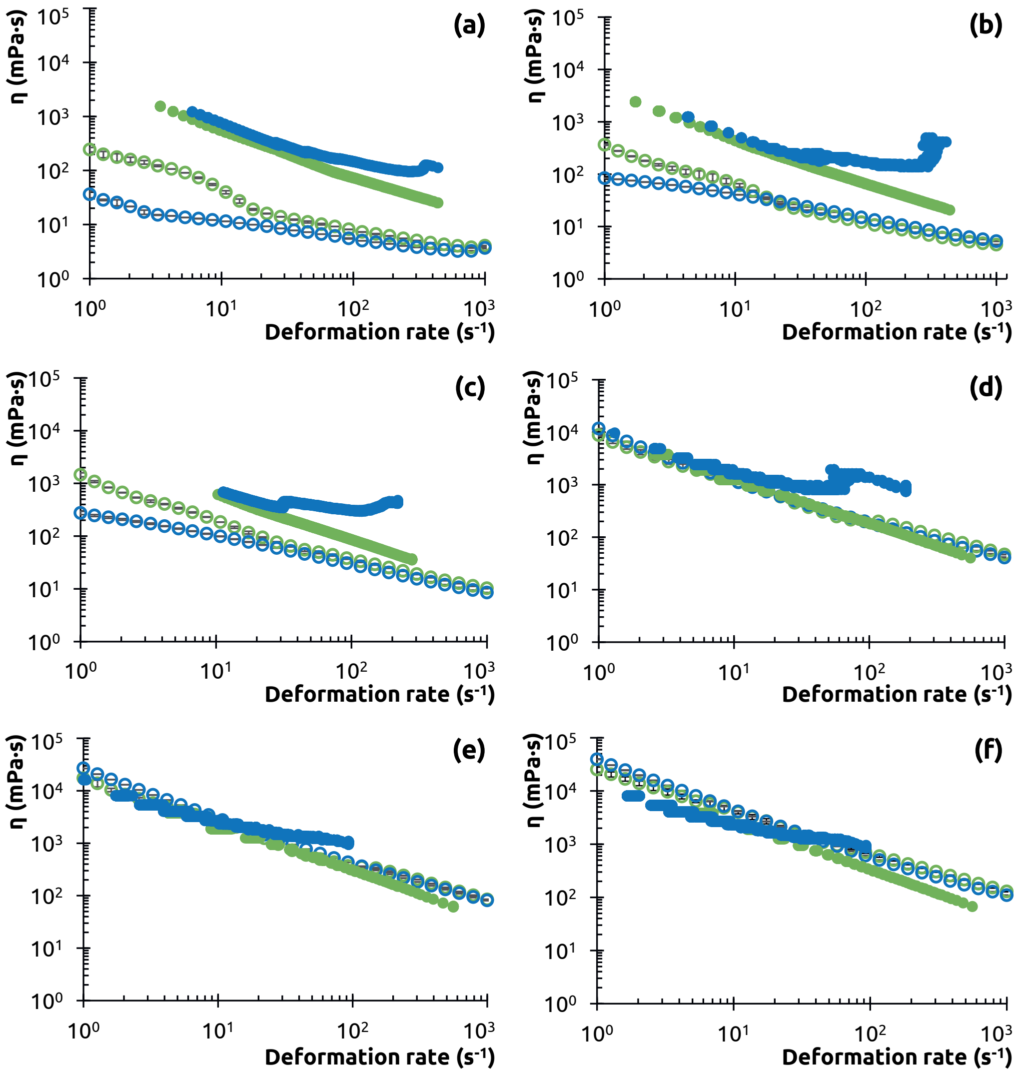

Figure 2.1. *Shear (η_{shear}; open symbols) and extensional viscosity (η_{ext}; closed symbols) of (a) MFC-1 (0.2%) and XG-1 (0.04%); (b) MFC-2 (0.3%) and XG-2 (0.10%); (c) MFC-3 (0.5%) and XG-3 (0.21%); (d) MFC-4 (1.0%) and XG-4 (2.0%); (e) MFC-5 (1.5%) and XG-5 (3.4%); (f) MFC-6 (2.0%) and XG-6 (4.3%). Green symbols represent microfibrillated cellulose (MFC), blue symbols represent xanthan gum (XG).*

As anticipated, viscoelastic moduli (*G'* & *G"*) and yield stress increased with increasing concentrations of MFC and XG (Table 2.3, Figure 2.2) (Agoda-Tandjawa *et al.*, 2010; Iotti *et al.*, 2011; Lowys *et al.*, 2001; Lundahl *et al.*, 2018; Martín-Alfonso *et al.*, 2018; Ross *et al.*, 2019). Higher thickener concentrations promote the formation of stronger networks, thus resulting in higher viscoelastic moduli and yield stresses. Yield stress of XG-1, XG-2 and XG-3 could not be determined experimentally, as *G"* was larger than *G'* and an intersect between *G"* and *G'* was absent in the range of shear strains measured. XG solutions displayed higher yield stress compared to iso-viscous MFC dispersions (*i.e.* yield stress of XG-4>MFC-4; XG-5>MFC-5; XG-6>MFC-6), which might be attributed to differences in network formation between both biopolymers. Dissolved polymer systems such as XG solutions exhibit more ductility towards applied shear stresses than particulate networks formed by rigid microfibrils in MFC dispersions, resulting in higher yield stresses that need to be overcome to induce flow. The yield stress of XG solutions increased linearly with increasing XG concentration, which is congruent with earlier work (Hannote *et al.*, 1991; Song *et al.*, 2006). Yield stress of MFC dispersions on the other hand increased following a power law with an exponent of 2, supporting earlier results (Agoda-Tandjawa *et al.*, 2010; Tatsumi *et al.*, 2002).

Table 2.3. *Mean (± SD) storage (G') and loss (G") moduli of microfibrillated cellulose (MFC) dispersions and xanthan gum (XG) solutions at 1% strain (10% shear strain for XG-4, XG-5, XG-6).*

	***G'* (Pa)**		***G"* (Pa)**	
	MFC	*XG*	*MFC*	*XG*
1	3.1 ± 0.3	0.0 ± 0.0	0.6 ± 0.1	0.3 ± 0.0
2	6.9 ± 0.5	0.2 ± 0.0	1.1 ± 0.1	0.5 ± 0.0
3	19.3 ± 0.2	0.5 ± 0.1	2.7 ± 0.0	0.8 ± 0.0
4	189.7 ± 40.5	20.2 ± 0.0	26.7 ± 5.9	7.2 ± 0.0
5	573.5 ± 105.3	44.0 ± 0.4	76.2 ± 16.3	13.3 ± 0.1
6	996.2 ± 21.8	62.1 ± 0.6	142.1 ± 1.7	17.4 ± 0.2

Table 2.4. *Absolute and relative hysteresis area of microfibrillated cellulose (MFC) dispersions and xanthan gum (XG) solutions (mean ± SD). Relative hysteresis areas were calculated as the difference in AUC between upward and downward stress-strain curve, divided by the area under the upward curve.*

	Absolute hysteresis area (s^{-1}·Pa)		**Relative hysteresis area (%)**	
	MFC	*XG*	*MFC*	*XG*
1	48 ± 15	3 ± 5	2.2 ± 0.7	0.2 ± 0.3
2	44 ± 17	-7 ± 1	1.4 ± 0.5	-0.2 ± 0.0
3	67 ± 24	-33 ± 1	1.1 ± 0.4	-0.6 ± 0.0
4	1344 ± 123	428 ± 43	4.3 ± 0.4	1.4 ± 0.1
5	519 ± 115	2729 ± 59	0.8 ± 0.2	4.2 ± 0.1
6	2726 ± 1228	6852 ± 440	2.8 ± 1.1	8.0 ± 0.5

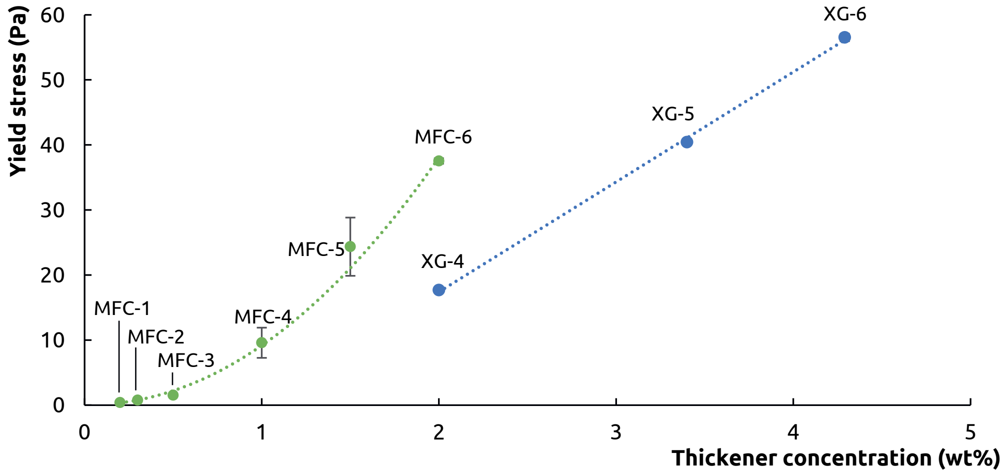

Figure 2.2. *Mean yield stress of microfibrillated cellulose (MFC) dispersions (green) and xanthan gum (XG) solutions (blue) as a function of thickener concentration. The three low-viscous XG solutions (XG-1 (0.04 wt%), XG-2 (0.10 wt%) and XG-3 (0.21 wt%)) did not display yielding behavior, as no intersect between G' and G" was observed. Error bars represent standard deviations. Dashed lines are displayed to guide the eyes and represent an exponential fit for MFC (with a power law exponent of ~2) and a linear fit for XG.*

In accordance with literature, hysteresis was observed for MFC dispersions (Agoda-Tandjawa *et al.*, 2010; Martoïa *et al.*, 2015; Schenker *et al.*, 2018) and XG solutions (Alghooneh *et al.*, 2018; Ghannam *et al.*, 2019; Silva & Lucas, 2018) (Table 2.4). Shear viscosities at low shear rates were higher for the upward curve (*i.e.* increasing shear rates) than the downward curve (*i.e.* decreasing shear rates). Hysteresis of MFC dispersions occurred between 1-20 s^{-1}, whereas hysteresis of XG solutions occurred at a larger range of shear rates (1-500 s^{-1}; Supp. Material, Figure S3). This time-dependent viscosity effect is attributed to a change in the structure of MFC dispersions and XG solutions under shear flow. XG molecules and MFC microfibrils are presumably randomly oriented in the absence of shear, whereas these orient themselves and align upon application of shear, thereby reducing resistance towards the flow field. Higher MFC and XG concentrations resulted in larger absolute hysteresis areas, which is in line with previous work (Schenker *et al.*, 2018; Silva & Lucas, 2018). More concentrated systems generally exhibit larger hysteresis areas, as relatively more time is required for stronger systems to return to their original state. Absolute hysteresis was generally larger for samples thickened with MFC compared to XG, except for samples with the highest viscosities (MFC-5 and MFC-6). Relative hysteresis areas ($(AUC_{upward}-AUC_{downward})/AUC_{upward}$) became larger as XG concentration increased, implying relatively more structural breakdown at higher XG concentrations. In contrast, thickener concentration did not influence relative hysteresis areas of MFC dispersions, which confirms previous work in which the relative hysteresis of MFC dispersions was found to be independent of the solids content (Schenker *et al.*, 2018).

It should be noted that shear viscosity, hysteresis, tribological and sensory properties were determined with MFC dispersions and XG solutions containing 7.5 wt% sugar, 0.1 wt% strawberry flavouring and 0.05 wt% red colourant. In contrast, oscillation and extensional rheology measurements were performed with MFC dispersions and XG solutions without these ingredients. We acknowledge that the addition of these ingredients, especially the 7.5 wt% sugar, might have caused a change in the rheological properties of the samples. These compositional differences need to be taken into account when comparing the extensional viscosity with the shear viscosity (Figure 2.1). We have compared the shear viscosity of samples with and without these ingredients and conclude that the addition of these ingredients has only a negligible effect on shear viscosity, and we therefore do not expect any large effects on the other rheological properties either (Supp. Material, Figure S4). Furthermore, instead of focusing on replacing one thickener by another, future research could study partial replacement of XG by MFC in MFC-XG mixtures and their effect on rheological, tribological and sensory properties of foods.

2.3.2. Tribological properties

Mean friction coefficients of MFC dispersions and XG solutions as a function of sliding speed are displayed in Figure 2.3. Friction coefficients of XG solutions decreased with increasing XG concentration, whereas this was not observed for MFC dispersions. The effect of viscosity on friction in the boundary and mixed regime has been described before (de Vicente *et al.*, 2005, 2006; Selway *et al.*, 2017; Stokes *et al.*, 2013). On the other hand, our results show no clear reduction in friction for increasing concentrations of MFC. This confirms recent work by Kinoshita *et al.* (2020), who concluded that MFC does not form a tribofilm on the tribological surface and postulated that MFC can only reduce friction by physical rolling or sliding mechanisms. Moreover, as larger MFC flocs can be formed under shear (Karppinen *et al.*, 2012; Saarikoski *et al.*, 2012), MFC flocs might have formed in the current study that were too large to enter the gap between the tribo-surfaces. It is thus likely that the MFC microfibrils were excluded from the contact region and friction properties of the continuous phase were measured instead, which mainly consisted of water, sugar and possibly the non-fibrous fraction of the cellulosic material used to prepare the MFC. Indeed, supernatant of centrifuged MFC dispersions (*i.e.* water-insoluble material was removed) showed similar friction properties as non-centrifuged MFC dispersions, supporting our hypothesis that the water-insoluble microfibrils did not enter the tribological gap (data not shown). Possibly, friction properties of MFC dispersions could be measured by using a different tribological set-up or tribo-pair. It is expected that a viscosity effect similar to that observed for XG solutions will occur for increasing concentrations of MFC.

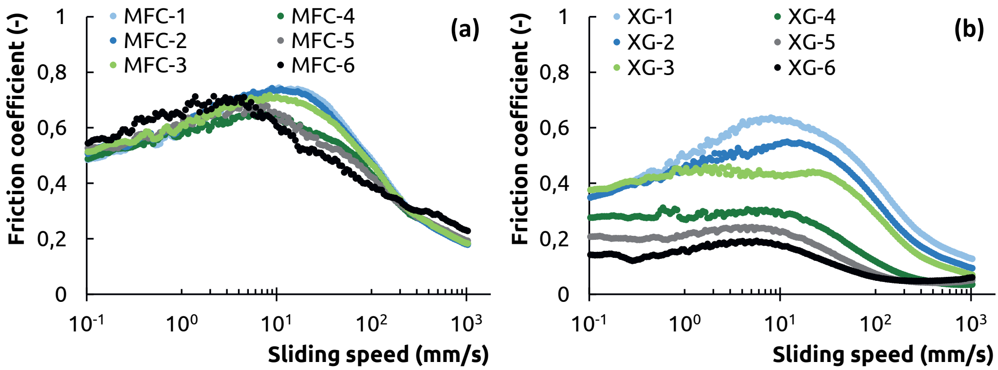

Figure 2.3. *Mean friction coefficients (triplicates) as a function of sliding speed of the six microfibrillated cellulose (MFC) dispersions (a) and xanthan gum (XG) solutions (b). It is hypothesised that MFC microfibrils were excluded from the tribological gap due to their size, and the friction curves thus represent the continuous aqueous phase of the MFC dispersions.*

2.3.3. Sensory properties of MFC dispersions and XG solutions

Mean intensities of appearance, flavour and texture attributes of the twelve samples obtained by RATA (*n*=73 participants) are shown in Table 2.5. To summarise the results of the sensory evaluation, Figure 2.4 shows the Principal Component Analysis bi-plot positioning the twelve samples in the sensory space.

Effect of thickener concentration on sensory perception

A significant main effect of thickener concentration on all sensory attributes was found (Table 2.5). This is reflected in the first dimension of the PCA bi-plot, which is related to the concentration of thickener in the samples and explains 37.9% of the variance between the samples.

Appearance - For both biopolymers, an increase in thickener concentration resulted in decreased transparency, red colour intensity and smoothness. The decrease in transparency at higher thickener concentrations has been reported by others (Kim *et al.*, 2017), and was accompanied by a reduction in colour intensity. Thickness and sliminess on the other hand increased as the thickener concentration increased. This was expected since higher thickener concentrations resulted in higher shear viscosities (Figure 2.1) and viscosity is related to visual thickness (Christensen & Casper, 1987) and sliminess (Brandenstein *et al.*, 2015).

Flavour - Strawberry flavour and sweetness intensity were reduced as thickener concentration increased. This was expected, since the flavour and taste intensity of liquid foods decrease with increasing viscosity or hydrocolloid concentration (Cook *et al.*, 2002; Gössinger *et al.*, 2018; Hollowood *et al.*, 2002; Kim *et al.*, 2017; Malone *et al.*, 2003b; Matta *et al.*, 2006). Cardboard flavour on the other hand is presumably an intrinsic property of the thickeners used, as its intensity increased with increasing concentrations of MFC and XG. Kim *et al.* (2017) reported that addition of xanthan-based thickeners to water resulted in increased starchy and nutty flavour.

Texture - Increasing the thickener concentration resulted in large differences in shear viscosity (Figure 2.1) and consequently affected all texture attributes assessed. Consistent with earlier findings, the increase in viscosity due to higher thickener concentrations resulted in increased perceived thickness (Cutler *et al.*, 1983). Since creaminess, sliminess and stickiness are correlated with perceived thickness (Bom Frøst & Janhøj, 2007; He *et al.*, 2016; Lyly *et al.*, 2003; Morris *et al.*, 1984; Upadhyay *et al.*, 2020), the intensity of these attributes increased as a consequence of increasing thickener concentration. In accordance with the present results, others determined that thickener concentration and viscosity were positively correlated with mouthcoating (Kim *et al.*, 2017), pulpiness (Brandenstein *et al.*, 2015), sliminess (Brandenstein *et al.*, 2015) and stickiness (Ross *et al.*, 2019) of hydrocolloid-thickened beverages. Melting was the sole attribute representing a dynamic sensory experience, *i.e.* the change in thickness over time. More viscous samples were perceived as more melting, which is presumably due to the fact that these samples exhibit larger degrees of oral breakdown. As the majority of the samples was liquid and was in the mouth for only a short time, the oral exposure time might have been too short to properly assess dynamic attributes such as melting.

Table 2.5. *Mean intensities (± SE) of appearance, flavour and texture attributes obtained from RATA with n=73 participants. Samples in the same row containing the same letter are not significantly different from each other. Statistically significant main effects of thickener concentration or thickener type (microfibrillated cellulose (MFC) vs xanthan gum (XG)) or their interaction are indicated by asterisks (n.s. = not significant; * p < 0.05; ** p < 0.01; *** p < 0.001).*

	MFC-1	MFC-2	MFC-3	MFC-4	MFC-5	MFC-6	XG-1	XG-2	XG-3	XG-4	XG-5	XG-6
Appearance												
Glossiness	6.0 ± 0.3^{ab}	5.5 ± 0.3^{ab}	5.6 ± 0.3^{ab}	5.1 ± 0.3^{bc}	4.3 ± 0.3^{cd}	3.4 ± 0.3^{d}	5.5 ± 0.3^{ab}	5.8 ± 0.3^{ab}	4.2 ± 0.3^{cd}	6.3 ± 0.2^{a}	6.3 ± 0.2^{a}	5.9 ± 0.3^{ab}
Red colour intensity	6.7 ± 0.2^{a}	6.3 ± 0.2^{ab}	5.1 ± 0.2^{cd}	4.6 ± 0.2^{de}	3.6 ± 0.2^{f}	3.5 ± 0.2^{f}	6.5 ± 0.2^{a}	6.7 ± 0.2^{a}	6.5 ± 0.2^{a}	5.6 ± 0.2^{bc}	4.6 ± 0.2^{de}	4.4 ± 0.2^{e}
Sliminess	0.9 ± 0.2^{de}	1.2 ± 0.2^{de}	2.7 ± 0.3^{c}	6.2 ± 0.3^{ab}	5.9 ± 0.4^{b}	5.8 ± 0.4^{b}	0.1 ± 0.0^{e}	0.4 ± 0.1^{de}	1.2 ± 0.2^{d}	5.9 ± 0.3^{b}	7.1 ± 0.2^{a}	7.1 ± 0.2^{a}
Smoothness	7.8 ± 0.2^{a}	7.5 ± 0.2^{a}	6.9 ± 0.3^{ab}	5.6 ± 0.3^{c}	5.1 ± 0.3^{cd}	4.1 ± 0.4^{d}	8.0 ± 0.2^{a}	8.0 ± 0.2^{a}	5.8 ± 0.4^{bc}	5.2 ± 0.4^{cd}	4.3 ± 0.4^{d}	4.2 ± 0.4^{d}
Thickness	1.0 ± 0.2^{fg}	1.5 ± 0.2^{f}	2.9 ± 0.2^{e}	7.0 ± 0.2^{c}	8.2 ± 0.1^{ab}	8.5 ± 0.1^{a}	0.3 ± 0.1^{g}	0.4 ± 0.1^{g}	1.2 ± 0.2^{f}	5.6 ± 0.2^{d}	7.6 ± 0.1^{bc}	8.0 ± 0.1^{ab}
Transparency	1.5 ± 0.2^{c}	1.2 ± 0.2^{cd}	1.0 ± 0.2^{cde}	0.6 ± 0.1^{def}	0.4 ± 0.1^{ef}	0.3 ± 0.1^{f}	7.5 ± 0.2^{a}	7.1 ± 0.2^{a}	4.4 ± 0.3^{b}	0.6 ± 0.1^{def}	0.7 ± 0.2^{def}	0.6 ± 0.1^{def}
Flavour												
Cardboard flavour	1.9 ± 0.3^{c}	2.0 ± 0.3^{bc}	3.0 ± 0.4^{bc}	4.5 ± 0.4^{a}	5.1 ± 0.4^{a}	5.2 ± 0.4^{a}	0.4 ± 0.1^{d}	0.3 ± 0.1^{d}	0.3 ± 0.1^{d}	2.0 ± 0.3^{bc}	2.4 ± 0.3^{bc}	3.0 ± 0.3^{b}
Strawberry flavour	4.8 ± 0.3^{abc}	5.0 ± 0.3^{ab}	4.5 ± 0.3^{bc}	3.4 ± 0.3^{de}	2.9 ± 0.3^{def}	2.8 ± 0.3^{ef}	5.5 ± 0.3^{a}	5.4 ± 0.3^{ab}	5.3 ± 0.2^{ab}	3.8 ± 0.3^{cd}	2.6 ± 0.3^{ef}	2.2 ± 0.3^{f}
Sweetness	6.1 ± 0.2^{ab}	5.7 ± 0.3^{ab}	5.2 ± 0.2^{b}	4.2 ± 0.3^{c}	3.5 ± 0.3^{cde}	3.4 ± 0.3^{cde}	6.4 ± 0.2^{a}	6.5 ± 0.2^{a}	5.6 ± 0.2^{ab}	4.0 ± 0.3^{cd}	3.2 ± 0.2^{de}	2.7 ± 0.2^{e}
Texture												
Creaminess	0.2 ± 0.1^{b}	0.3 ± 0.1^{b}	0.9 ± 0.2^{b}	2.0 ± 0.3^{a}	2.5 ± 0.4^{a}	2.9 ± 0.4^{a}	0.1 ± 0.1^{b}	0.2 ± 0.1^{b}	0.6 ± 0.2^{b}	2.5 ± 0.3^{a}	2.3 ± 0.3^{a}	2.5 ± 0.4^{a}
Melting	0.8 ± 0.2^{c}	0.8 ± 0.2^{c}	1.0 ± 0.2^{bc}	2.3 ± 0.3^{a}	2.0 ± 0.3^{ab}	2.3 ± 0.3^{a}	0.6 ± 0.2^{c}	0.8 ± 0.2^{c}	1.4 ± 0.3^{abc}	1.6 ± 0.3^{abc}	1.1 ± 0.2^{bc}	1.5 ± 0.3^{abc}
Mouthcoating	0.9 ± 0.2^{e}	1.1 ± 0.2^{e}	1.8 ± 0.2^{de}	2.8 ± 0.3^{d}	4.0 ± 0.3^{c}	4.6 ± 0.3^{c}	0.8 ± 0.2^{e}	1.2 ± 0.2^{e}	1.4 ± 0.2^{e}	4.9 ± 0.3^{bc}	5.8 ± 0.3^{ab}	6.0 ± 0.3^{a}
Pulpiness	0.0 ± 0.0^{d}	0.0 ± 0.0^{d}	0.0 ± 0.0^{d}	1.0 ± 0.2^{bc}	1.6 ± 0.3^{b}	2.8 ± 0.4^{a}	0.0 ± 0.0^{d}	0.0 ± 0.0^{d}	0.0 ± 0.0^{d}	0.5 ± 0.2^{cd}	1.0 ± 0.3^{bc}	1.1 ± 0.3^{c}
Sliminess	0.3 ± 0.1^{f}	0.4 ± 0.1^{f}	1.5 ± 0.2^{e}	3.5 ± 0.4^{d}	4.5 ± 0.4^{c}	4.5 ± 0.3^{c}	0.1 ± 0.1^{f}	0.2 ± 0.1^{f}	0.6 ± 0.1^{ef}	6.0 ± 0.3^{b}	7.5 ± 0.2^{a}	7.9 ± 0.2^{a}
Smoothness	8.1 ± 0.1^{a}	8.0 ± 0.2^{a}	7.8 ± 0.2^{a}	6.5 ± 0.3^{b}	6.6 ± 0.3^{b}	5.4 ± 0.4^{c}	8.3 ± 0.2^{a}	8.3 ± 0.1^{a}	7.9 ± 0.2^{a}	6.4 ± 0.3^{b}	6.2 ± 0.3^{bc}	5.9 ± 0.4^{bc}
Stickiness	0.4 ± 0.1^{c}	0.6 ± 0.2^{c}	0.7 ± 0.2^{c}	1.0 ± 0.2^{c}	2.2 ± 0.3^{b}	2.2 ± 0.3^{b}	0.2 ± 0.1^{c}	0.4 ± 0.1^{c}	0.5 ± 0.1^{c}	3.0 ± 0.3^{b}	4.8 ± 0.4^{a}	5.4 ± 0.4^{a}
Thickness	0.4 ± 0.1^{e}	0.5 ± 0.1^{e}	1.5 ± 0.2^{d}	4.6 ± 0.3^{c}	5.5 ± 0.2^{b}	6.5 ± 0.2^{a}	0.2 ± 0.1^{e}	0.3 ± 0.1^{e}	0.8 ± 0.1^{de}	4.9 ± 0.3^{bc}	6.7 ± 0.2^{a}	6.9 ± 0.2^{a}

Table 2.5 (continued). *Mean intensities (± SE) of appearance, flavour and texture attributes obtained from RATA with n=73 participants. Samples in the same row containing the same letter are not significantly different from each other. Statistically significant main effects of thickener concentration or thickener type (microfibrillated cellulose (MFC) vs xanthan gum (XG)) or their interaction are indicated by asterisks (n.s. = not significant; * p < 0.05; ** p < 0.01; *** p < 0.001).*

	Thickener concentration		**Thickener type**		**Interaction effect**	
Appearance	*F*-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
Glossiness	$F(5,789) = 9.3$	***	$F(1,789) = 29.1$	***	$F(5,789) = 23.1$	***
Red colour intensity	$F(5,787) = 148.3$	***	$F(1,787) = 97.5$	***	$F(5,787) = 8.7$	***
Sliminess	$F(5,788) = 342.4$	***	$F(1,788) = 1.3$	n.s.	$F(5,788) = 12.6$	***
Smoothness	$F(5,789) = 81.2$	***	$F(1,789) = 3.2$	n.s.	$F(5,789) = 2.8$	*
Thickness	$F(5,787) = 1246.9$	***	$F(1,787) = 150.6$	***	$F(5,787) = 5.9$	***
Transparency	$F(5,788) = 371.3$	***	$F(1,788) = 1121.4$	***	$F(5,788) = 213.7$	***
Flavour	*F*-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
Cardboard flavour	$F(5,789) = 61.5$	***	$F(1,789) = 247.0$	***	$F(5,789) = 2.2$	*
Strawberry flavour	$F(5,788) = 70.8$	***	$F(1,788) = 4.2$	*	$F(5,788) = 3.5$	**
Sweetness	$F(5,788) = 104.6$	***	$F(1,788) = 0.2$	n.s.	$F(5,788) = 4.1$	**
Texture	*F*-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
Creaminess	$F(5,789) = 62.0$	***	$F(1,789) = 0.9$	n.s.	$F(5,789) = 1.0$	n.s.
Melting	$F(5,789) = 12.7$	***	$F(1,789) = 9.5$	**	$F(5,789) = 3.0$	*
Mouthcoating	$F(5,789) = 148.3$	***	$F(1,789) = 36.1$	***	$F(5,789) = 11.1$	***
Pulpiness	$F(5,788) = 42.1$	***	$F(1,788) = 21.3$	***	$F(5,788) = 7.2$	***
Sliminess	$F(5,788) = 421.0$	***	$F(1,788) = 122.5$	***	$F(5,788) = 46.8$	***
Smoothness	$F(5,787) = 56.1$	***	$F(1,787) = 0.5$	n.s.	$F(5,787) = 1.3$	n.s.
Stickiness	$F(5,787) = 116.0$	***	$F(1,787) = 102.1$	***	$F(5,787) = 29.6$	***
Thickness	$F(5,789) = 651.0$	***	$F(1,789) = 3.4$	n.s.	$F(5,789) = 8.1$	***

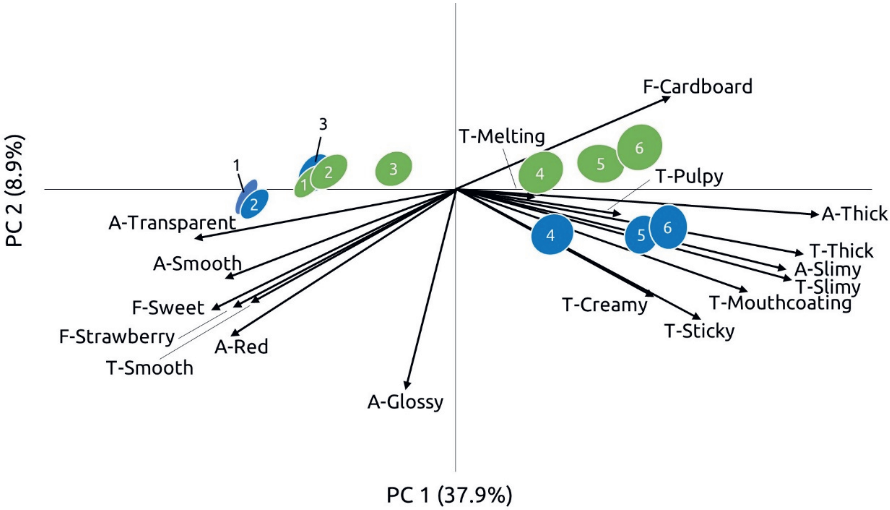

Figure 2.4. *Principal Component Analysis (PCA) bi-plot displaying loadings for the appearance, flavour and texture attributes and scores for the twelve model foods (green = microfibrillated cellulose (MFC), blue = xanthan gum (XG)) with their respective 95% confidence ellipses. Numbers in circles correspond to the sample code.*

Effect of thickener type on sensory perception

Thickener type (MFC vs. XG) had a significant effect on 11 out of 17 sensory attributes, and sensory differences between samples thickened with MFC and XG were generally larger at higher concentrations (Table 2.5). The differences between the two thickeners is reflected by the second dimension of the PCA bi-plot (Figure 2.4), which particularly separates the high viscous samples based on thickener type (MFC-4, MFC-5, MFC-6 vs. XG-4, XG-5, XG-6). Sensory attributes related to this dimension include glossy appearance, cardboard flavour and sticky texture.

Appearance - Thickener type had a strong effect on the transparency of the samples. MFC dispersions were considerably less transparent than XG solutions of similar shear viscosity, especially at the lowest thickener concentrations (transparency intensities of 1.5 vs 7.5). Although red colour intensities were comparable at low thickener concentrations, intensities displayed a sharper decrease with increasing MFC concentration than XG concentration. Glossy appearance has high loadings on the second dimension of the PCA bi-plot (Figure 2.4), indicating that glossy appearance is related to thickener type. Glossiness decreased with increasing MFC concentration, whereas no univocal effect of XG concentration on glossiness was observed. Since MFC forms a dispersion in water, the cellulose microfibrils might induce more light scattering than XG solutions (Hutchings, 1994). The scattering of light presumably resulted in reduced glossiness, transparency and lower colour intensity. Visual thickness of MFC dispersions was higher than that of XG solutions, although the samples had comparable shear viscosities over a large range of shear rates (Figure 2.1).

Flavour - When comparing iso-viscous MFC dispersions and XG solutions, cardboard flavour was consistently rated more intense for samples thickened with MFC. Although higher concentrations of MFC resulted in lower hedonic flavour ratings in mayonnaises (Choublab & Winuprasith, 2018), the majority of literature reports that incorporation of MFC does not lead to off-flavours in hamburgers (Ström *et al.*, 2013), mayonnaise (Golchoobi *et al.*, 2016) and ice creams (Yano *et al.*, 2016). It is hypothesised that the off-flavour perceived in the current study was caused by the heat treatment (sterilisation at 125°C for 15 min) given to the MFC dispersions in order to extend shelf life. Higher strawberry flavour intensities were observed for samples thickened with XG compared to those thickened with MFC, especially for low-viscous samples. Possibly, the strawberry flavour was suppressed by the cardboard flavour in MFC dispersions.

Texture - Significant differences between samples thickened with MFC and XG were observed in terms of mouthcoating, sliminess and stickiness. Especially at higher thickener concentration, samples thickened with XG were found to be more mouthcoating, slimy and sticky. These results confirm those from other studies reporting an effect of XG concentration on mouthcoating (Kim *et al.*, 2017), sliminess (Gössinger *et al.*, 2018) and stickiness intensities (Akhtar *et al.*, 2006; Ross *et al.*, 2019; Yamagata *et al.*, 2012) of thickened liquids. Thickening with MFC on the other hand resulted in increased pulpiness and slightly increased melting sensations. Higher pulpiness of samples thickened with MFC may be explained by the fact that the cellulose microfibrils behave like water-insoluble particles, which might be perceived as small fibres. MFC dispersions might moreover be perceived as more melting due to weaker interactions between the microfibrils compared to XG molecules. The presence of saliva in the mouth dilutes the MFC dispersions, which presumably results in reduced microfibril interactions and a rapid loss of in-mouth viscosity. Thickener type did not significantly affect thickness and creaminess intensities. This was not unexpected, as samples thickened with MFC and XG were iso-viscous and viscosity is a major contributor to creaminess (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; Bom Frøst & Janhøj, 2007).

2.3.4. Linking sensory characteristics to rheological and tribological properties

This is the first time rheological and tribological properties of MFC were compared to another thickener and linked to sensory appearance, flavour and texture attributes. The Multiple Factor Analysis (MFA) plot shows the relationships between rheological, tribological and sensory properties of the MFC dispersions and XG solutions (Figure 2.5). Several attributes are located close to each other at the left side of the plot, including smoothness (A-Smooth and T-Smooth), sweetness and strawberry flavour, implying that these attributes did not vary independently in the studied samples. These attributes are moreover negatively correlated with attributes on the other side of the plot, *i.e.* creaminess, thickness (A-Thick and T-Thick) and shear viscosity parameters. This suggests that more viscous samples were perceived as thick and creamy, but had low intensities of smoothness, sweetness and strawberry flavour, which is consistent with results from the PCA bi-plot (Figure 2.4). The fact that thickness (A-Thick and T-Thick) and creaminess are positively correlated with shear viscosity at 10, 50 and 100 s^{-1} supports previous literature (Conti-Silva *et al.*, 2018; Krzeminski *et al.*, 2013; Sonne *et al.*, 2014).

The right side of the MFA plot shows that the texture attributes mouthcoating, slimy and sticky are correlated (Figure 2.5). These results reflect those of Ross *et al.* (2019) who also found a strong correlation between stickiness and oral residue, which is considered comparable to mouthcoating in the present study. The attributes mouthcoating, slimy and sticky are moreover located close to hysteresis, yield stress and extensional viscosity. This is in agreement with the fact that XG solutions exhibited higher extensional viscosities at higher deformation rates and that these solutions were perceived to be more mouthcoating, slimy and sticky than MFC dispersions. Similarly, Lyly *et al.* (2003) reported a strong correlation between sliminess and extensibility of model beverages, and He *et al.* (2016) found that stickiness and mouthcoating were strongly correlated with extensional viscosity (r > 0.9). Interestingly, several studies postulated that mouthcoating and stickiness correlate with the degree of shear-thinning of hydrocolloids (*i.e.* flow behaviour index n) (Ross *et al.*, 2019; Szczesniak & Farkas, 1962; Vickers *et al.*, 2015; Wood, 1974). Our results show that liquids thickened with different hydrocolloids but with similar shear thinning behaviour can have different mouthcoating, stickiness and sliminess intensities. This suggests that other rheological properties, such as extensional viscosity, yield stress or hysteresis, might be associated with sensory perception of these texture attributes. It should be noted that only tribological data from XG samples was used as input for the MFA, which might explain why no correlation was found between friction parameters and sensory attributes. Furthermore cardboard flavour, pulpy and melting mouthfeel were located close to the dynamic moduli (G' and G'') of the samples, which is presumably driven by MFC samples being characterised by these sensory attributes and simultaneously having high G' and G''.

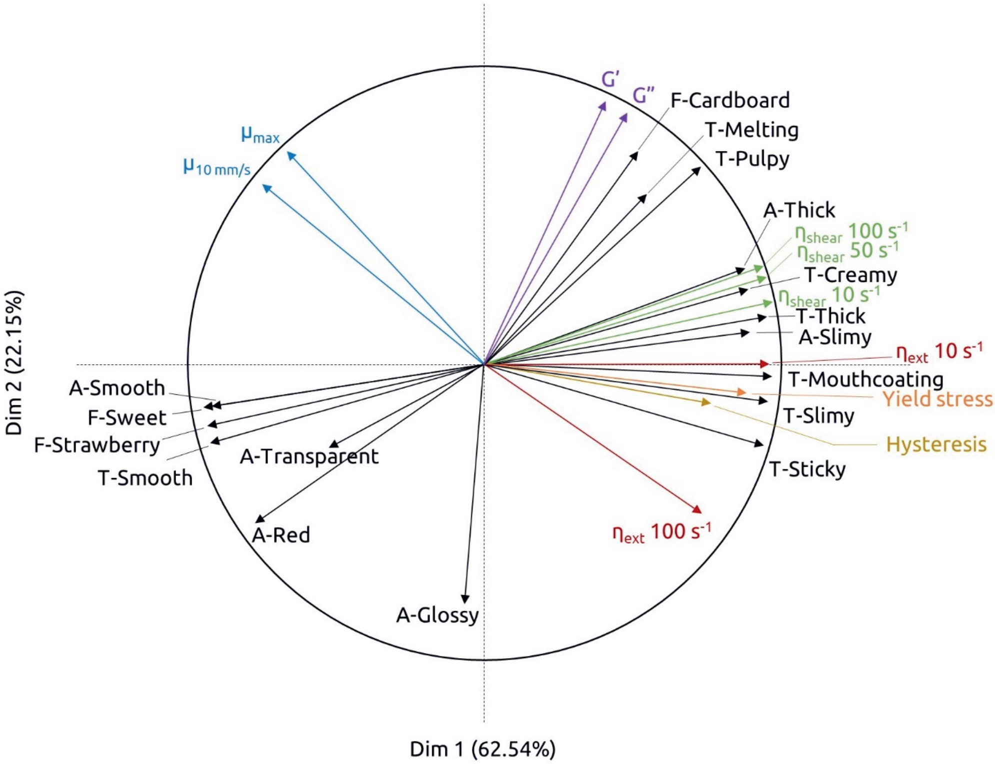

Figure 2.5. *Multiple Factor Analysis (MFA) displaying the appearance (A), flavour (F) and texture (T) attributes (in black), shear viscosity (in green), extensional viscosity (in red), tribological properties (in blue; only XG data), dynamic moduli G' and G" (purple), yield stress (orange) and relative hysteresis area (yellow) of twelve aqueous model foods thickened with either MFC or XG.*

2.4. Conclusions

This is the first study that compared and linked rheological, tribological and sensory properties of aqueous model foods thickened with different concentrations of MFC and XG. Although shear viscosities of MFC dispersions and XG solutions matched over a wide range of shear rates, viscous XG solutions exhibited higher yield stress than MFC dispersions at similar shear viscosity. Moreover, yield stress increased linearly with concentration for XG solutions whereas it increased exponentially for MFC dispersions. XG solutions displayed higher extensional viscosity at higher deformation rates, which was correlated with sensory perception of mouthcoating, slimy and sticky mouthfeel. These sensory attributes mainly prevailed in XG solutions, whereas MFC dispersions were characterised by reduced transparency and glossiness and stronger cardboard flavour. Our results furthermore show that thickener concentration affected all appearance, flavour and texture attributes assessed in this study. Since sliminess, stickiness and mouthcoating are generally disliked and can impede swallowing, MFC might offer a good alternative to XG to be used as thickening agent, for example in dysphagia management. In that case the cardboard flavour of MFC dispersions should be reduced and its dispersibility improved, to allow for easy application in liquids.

Acknowledgements

We thank Roland Gouzy for his input and the preparation of the MFC dispersions. This research was performed within the framework of the “Molecular aspects of biopolymers defining food texture perception and oral digestion” project funded by NWO (The Netherlands Organization for Scientific Research), grant number 731.017.201, Unilever and Anton Paar. The project partners (University of Amsterdam, Unilever, Anton Paar) have contributed to the project through regular discussion.

3

Influence of thickeners (microfibrillated cellulose, starch, xanthan gum) on rheological, tribological and sensory properties of low-fat mayonnaises

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Abstract

Microfibrillated cellulose (MFC) is obtained by high-shear treatment of cellulose. MFC is suitable for use as clean-label, low-calorie thickener in semi-solid foods such as mayonnaises due to its high surface area and water holding capacity. The aim of this study was to determine the effect of type and concentration of thickener on rheological, tribological and sensory properties of low-fat mayonnaises. Low-fat mayonnaises were prepared with four types of thickeners (MFC, chemically modified starch, native waxy corn starch, xanthan gum) at three concentrations. Higher biopolymer concentrations resulted in increased shear viscosities, *G'* and *G"*, yield stress and enhanced lubrication (*i.e.* lower friction coefficients). Mayonnaises with modified starch and xanthan gum generally had higher shear viscosity and yield stress compared to mayonnaises with comparable concentrations of MFC and waxy corn starch. MFC-thickened mayonnaises had highest *G'*, *G"* and boundary friction coefficients. Sensory properties of mayonnaises were determined using the Rate-All-That-Apply (RATA) method (n = 80). Addition of xanthan gum induced high sliminess and pulpiness, and low melting, creaminess and smoothness. Sensory properties of mayonnaises with MFC were generally similar to those with modified and waxy corn starch, despite differences in appearance (increased yellowness and slightly lower glossiness). Multiple Factor Analysis revealed that more shear-thinning mayonnaises were perceived as slimy. Boundary friction was negatively correlated with stickiness, while friction at the start of the hydrodynamic regime was positively correlated with melting sensations. We conclude that microfibrillated cellulose can be used as a thickener in low-fat mayonnaise as an alternative to commercially used chemically modified starch without considerably affecting its sensory texture properties.

3.1. Introduction

Microfibrillated cellulose (MFC), sometimes called nanofibrillated cellulose, is a type of nanocellulose produced by mechanical treatment of cellulose (Gómez *et al.*, 2016; Klemm *et al.*, 2011; Lavoine *et al.*, 2012). Cellulosic materials can be derived from wood and agricultural crops, including fruit and vegetables peel (Gómez *et al.*, 2016; Lavoine *et al.*, 2012). MFC has been developed and patented in the 1980s by Turbak and colleagues (Turbak *et al.*, 1983a), who used high-pressure homogenisation to obtain MFC from wood pulp fibres. The high mechanical shear applied to the cellulose dispersion causes cellulose fibres to deagglomerate and disintegrate. As a consequence MFC has a high aspect ratio and an increased surface area, resulting in high water absorption capability that facilitates the formation of stable, viscous dispersions with pseudoplastic properties at concentrations below 10 wt% (Klemm *et al.*, 2011; Lavoine *et al.*, 2012). MFC is produced without significant chemical treatment and thus contains both the crystalline and amorphous regions of cellulose. Its biodegradability, renewability and the possible use of agricultural by-products such as fruit and vegetable peels as starting material for MFC offer environmental and sustainability benefits (Lavoine *et al.*, 2012). MFC is a dietary fibre that is not absorbed or digested in the human digestive tract. It can therefore be used as a thickener or fat substitute to produce low-calorie foods (Kleinschmidt *et al.*, 1988; Tuason *et al.*, 2004; Turbak *et al.*, 1983b). Despite its potential health benefits (Gill *et al.*, 2020) the main application of MFC in food industry has been in food packaging (Gómez *et al.*, 2016). Commercialisation of MFC has been challenging due to the energy-consuming production process and the associated high production costs (Klemm *et al.*, 2011; Ström *et al.*, 2013). Its application is furthermore hampered by the fact that MFC cannot be redispersed after dehydration due to irreversible aggregation of the cellulose fibrils (hornification)(Déléris & Wallecan, 2017).

In recent years more efficient and less energy-consuming production methods for MFC have been developed, making MFC more affordable and allowing for its commercialisation as a functional food ingredient (Ström *et al.*, 2013). Several researchers established the potential of MFC to improve foam stability (Ström *et al.*, 2013) and to stabilise oil-in-water emulsions (e.g. Aaen *et al.*, 2019; Lu *et al.*, 2019; Nomena *et al.*, 2018; Ström *et al.*, 2013; Turbak *et al.*, 1983b, 1984; Winuprasith & Suphantharika, 2015). Apart from its emulsifying properties, its gel-like characteristics make MFC suitable as clean-label thickener in foods (Blok *et al.*, 2021). As a result MFC has been studied in various foods including soups, gravies, dips, puddings, toppings (Turbak *et al.*, 1982) and fruit-fillings in cookies (Kleinschmidt *et al.*, 1988). Several authors studied the effect of addition of MFC to mayonnaises. Choublab and Winuprasith (2018) found that it is possible to produce egg-free mayonnaise by using MFC as the sole emulsifier. The viscosifying effect of MFC in mayonnaises has been established by Heggset and colleagues (2020), who demonstrated that lower viscosity and moduli (*G′*, *G″*) evoked by fat reduction in mayonnaises can be regained by addition of 0.42 wt% MFC. Although these authors thoroughly characterised rheological properties of the mayonnaises, they did not explore the effect of MFC on sensory properties. Golchoobi *et al.* (2016) studied the effect of a range of mixtures of thickeners on rheological and hedonic properties of low-fat mayonnaises. Addition of 1 wt% MFC to low-fat mayonnaise resulted in hedonic evaluations similar to commercially available low-fat mayonnaise, which is typically thickened by addition of starch. This study focused on hedonic evaluations of mayonnaises by trained assessors and did not explore sensory properties of mayonnaises or the impact of MFC concentration on mayonnaise properties. To the best of our knowledge, no multidisciplinary studies have been performed so far that compared rheological, tribological and sensory characteristics of semi-solid mayonnaises thickened with MFC with other frequently used thickeners such as chemically modified corn starch, native waxy corn starch and xanthan gum.

The aim of this study was to determine the effect of type and concentration of thickener on rheological, tribological and sensory properties of low-fat mayonnaises. Furthermore, we sought to determine relationships between sensory, rheological and tribological properties of the mayonnaises. Low-fat mayonnaises were prepared with four types of biopolymers (MFC, modified starch, waxy corn starch, xanthan gum) varying in concentration. By comparing rheological, tribological and sensory properties of low-fat mayonnaises thickened by different biopolymers, we examine whether MFC can be used as a clean-label, low-calorie alternative thickener. As the effect of MFC on rheological and sensory properties in aqueous model foods has been established previously (Blok *et al.*, 2021) this study focuses on oil-in-water emulsions, in particular low-fat mayonnaises.

3.2. Materials & Methods

3.2.1. Mayonnaise preparation

Low-fat mayonnaises (20 wt% fat) were prepared with four thickening agents: microfibrillated cellulose (MFC), chemically modified corn starch (MS; E1442; Ingredion Incorporated, Westchester, IL, USA), native waxy corn starch (WCS; Novation® 2300, Ingredion Incorporated, Westchester, IL, USA) and xanthan gum (XG; Jungbunzlauer, Basel, Switzerland). Each thickening agent (MFC, MS, WCS, XG) was added to low-fat mayonnaises at three concentrations which were categorized as low, medium, and high, resulting in twelve low-fat mayonnaises in total. Table 3.1 summarises the composition of the low-fat mayonnaises. We aimed at obtaining comparable viscosities of the continuous phase of the four mayonnaises (before emulsification) at a certain thickener concentration (low, medium and high), *i.e.* viscosity of MFC-low was comparable to MS-low, WCS-low and XG-low. The choice of MS and WCS concentrations was based on knowledge and experience on using these ingredients in mayonnaises and reflects starch concentrations used in commercial low-fat mayonnaises. The concentrations of MFC and XG were based on previous work on these ingredients (Blok *et al.*, 2021).

The first step in preparation of the low-fat mayonnaises was to prepare the aqueous hydrocolloid solutions and dispersions. Modified and waxy corn starch were first mixed with water and cooked for 5 min at 85°C in a Thermomix® while continuously stirring (Thermomix® TM5, Vorwerk, Germany). The starch pastes were left to cool down to 50°C and the amount of water lost due to evaporation was added back to the paste. Xanthan gum was dissolved in water by mixing at room temperature for at least 60 min using an overhead stirrer. Microfibrillated cellulose dispersions were prepared by first suspending citrus fibre powder (HERBACEL® AQ® Plus, Herbafood Ingredients, Werder, Germany) in deionised water. pH of the samples was adjusted to pH 4 using 1M HCl (Sigma-Aldrich, Saint Louis, MO, USA). The suspensions were thoroughly mixed using a L5M-A Silverson laboratory mixer with a 1 mm screen hole (Silverson Machines Ltd., Chesham, United Kingdom) at 3,000 rpm for 10 min, followed by one passage through a high-pressure homogeniser (Microfluidizer M-110S, Microfluidics™, Newton, MA, USA) with a z-shape geometry (ø 87 µm) at a pressure of 1200 bar.

The hydrocolloid solutions and dispersions were subsequently mixed with the other ingredients of the aqueous phase and combined with egg yolk (Table 3.1). Sucrose (coarse medium, 0.315-1.25 mm), salt (salt evaporated non-iodized), sorbic acid (Nutrinova®) and calcium disodium ethylenediaminetetraacetic acid ($CaNa_2$-EDTA, Solvitar (E385) Food) were obtained from Brenntag Nederland B.V. (Dordrecht, the Netherlands). Acetic acid (vinegar spirit 12%) was obtained from Carl Kühne KG (GmbH & Co., Hamburg, Germany). Lemon flavour was added to the oil phase. The soybean oil phase was added slowly to the aqueous phase while stirring at 5200 rpm using a L5M-A Silverson laboratory mixer with 1 mm hole emulsor screen (Silverson Machines Ltd., Chesham, United Kingdom). Once all oil was added, the speed was increased to 7200 rpm for 1 min and the beaker with the emulsion was moved around to ensure complete homogenisation. The finished mayonnaises were transferred to 200 ml glass jars and stored at 4°C until further use. Two batches of 2600 g were prepared for each mayonnaise.

Table 3.1. *Composition of low-fat mayonnaises prepared with four thickening agents. Each thickening agent was applied at three concentrations (low, medium, high) resulting in twelve low-fat mayonnaises. Concentrations are given as wt%.*

	Microfibrillated cellulose (MFC)			**Modified starch (MS)**			**Waxy corn starch (WCS)**			**Xanthan gum (XG)**		
	low	medium	high	low	medium	high	low	medium	high	low	medium	high
Thickener	1.44	1.6	1.76	5.0	5.5	6.0	5.5	6.0	6.5	1.6	1.8	2.0
Water	67.9	67.8	67.6	64.4	63.9	63.4	63.9	63.4	62.9	67.8	67.6	67.4
Soybean oil	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0
Egg yolk	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sucrose	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
Acetic acid	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Salt (NaCl)	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Sorbic acid	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Lemon flavour	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
EDTA	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075	0.0075

3.2.2. Rheological characterisation

Rheological properties of mayonnaises were determined using a MCR 302 rheometer (Anton Paar, Graz, Austria) equipped with a parallel plate geometry (ø 50 mm) with a gap of 1 mm. Shear viscosity was determined as a function of logarithmically increasing shear rate from 0.1 s^{-1} to 500 s^{-1} in 50 steps (10 sec per data point). After loading, a waiting step of 5 min was applied to allow for structural relaxation of the sample before the start of the measurement. Measurements were performed in duplicate at 35°C. Consistency index *K* and flow index *n* were determined using the Ostwald-de Waele power law model: $\sigma = K \cdot \dot{\gamma}^n$, where σ = shear stress (Pa), $\dot{\gamma}$ = shear rate (s^{-1}), *K* = consistency index ($Pa{\cdot}s^n$) and *n* = flow index. This model was fitted to data in the entire shear rate range used. Data was also fitted to the Herschel-Bulkley model as this model is commonly used to describe the rheology of mayonnaises (e.g. Golchoobi *et al.*, 2016; Lee *et al.*, 2013; Ma & Barbosa-Cánovas, 1995; Su *et al.*, 2010). This model however failed to describe the flow behaviour of mayonnaises with XG. We therefore chose to use the Ostwald-de Waele model, in order to use one single model that can describe the flow behaviour of all mayonnaises used in this study. Strain sweeps were performed and *G'* and *G"* were measured as a function of logarithmically increasing shear strain (0.01-100%) at constant oscillation frequency (1 Hz). Samples were pre-sheared at 100 s^{-1} for 1 min, followed by 2 min rest to allow for structural relaxation. Yield stress was determined from the strain sweeps as the stress applied at the intersect of *G'* and *G"*. Measurements were performed in triplicate at 35°C.

3.2.3. Tribological characterisation

Tribological properties of the mayonnaises were determined using a MCR 302 rheometer (Anton Paar, Austria) equipped with a tribological cell (T-PTD-200). A ball-on-three-pins set-up was used, with a glass ball and polydimethylsiloxane (PDMS) pins. All measurements were performed in triplicate at 35°C and a normal force F_N of 1 N was applied. Each measurement consisted of three consecutive runs in which rotational sliding speeds were logarithmically increased from 0.0001-2200 rpm ($4{\cdot}10^{-5}$-10^3 mm/s). Each run was preceded by a 5 min resting period in which a normal force of 1 N was applied. Data from the second run was used for data analysis. Friction coefficients were defined as the ratio of the frictional force divided by the normal load. PDMS pins were replaced by new pins after each replicate of the entire set of samples to limit the effect of wear on the PDMS pins. New PDMS were run-in by one run with deionised water, followed by one run with one of the mayonnaises (MFC-low).

3.2.4. Confocal Laser Scanning Microscopy (CLSM)

Microstructures of low-fat mayonnaises were visualised using a Zeiss LSM 510-META confocal laser scanning microscope (Carl Zeiss AG, Oberkochen, Germany). The fat phase of all mayonnaises was stained with 0.0001% (w/v) Nile Red (Sigma-Aldrich, Saint-Louis, MO, USA). Different samples of mayonnaises thickened with MS and WCS were stained with 0.1% (w/v) Acridine Orange (Sigma-Aldrich, USA) in 0.1M phosphate buffer (pH 7) to visualise starch and proteinaceous materials. Calcofluor White (American Cyanamid, Wayne, NJ, USA; 0.002%) was used to stain cellulose in mayonnaises thickened with MFC. A drop of the stained mayonnaise was placed on an object slide and images were acquired using a Plan-Apochromat 63x/1.4 oil DIC objective for Calcofluor White and an EC Plan-Neofluar 40x/1.30 oil DIC objective for Nile Red and Acridine Orange. Excitation wavelengths were 543 nm for Nile Red, 488 nm for Acridine Orange and 405 nm for Calcofluor White.

3.2.5. Sensory evaluation

Participants

Participants between 18-50 y from Wageningen and surroundings were recruited online and through posters at the Wageningen University campus. Participants had a BMI between 18-30 kg/m^2, were non-smokers, proficient in reading English and were generally in good health with normal smell and taste functions. Participants had no allergies for any of the mayonnaise ingredients, were familiar with mayonnaise and consumed mayonnaise on a regular basis. Female participants were not pregnant or breastfeeding. A total of n = 80 participants (13 male, 67 female; mean age 25±5 y; mean BMI 22±2 kg/m^2) completed the study. Participants signed an informed consent form and completed a general questionnaire at the start of the first session. Participants received financial reimbursement upon completion of the study. The study did not meet the requirements to be reviewed by the Medical Research Ethical Committee of The Netherlands according to the "Medical Research Involving Human Subjects Act" of The Netherlands. The study was conducted in agreement with the ethics regulations laid out in the Declaration of Helsinki (2013).

Rate-All-That-Apply (RATA) method

Each participant evaluated all twelve mayonnaises in two test sessions of 30-45 min each. Six samples were evaluated in each test session. Samples were presented monadically in random order during the two test sessions. In the first test session participants tasted two example mayonnaises representing the range of mayonnaises to be evaluated (WCS-low and XG-high) in order to familiarise participants with the samples. Participants selected one of these mayonnaises to answer the example question to get acquainted with the sensory evaluation method. Mayonnaises were evaluated using the Rate-All-That-Apply (RATA) method. For each sample, participants indicated which sensory attributes were applicable to describe the perception of the sample, followed by indicating the intensity of the selected sensory attributes on a 9-point scale anchored from *low* to *high* intensity. Applicable attributes were selected from a list of 18 sensory attributes, which were divided over three categories: appearance, flavour and texture. Definitions of the sensory attributes were provided to the participants (Table 3.2). After evaluation of a sample, participants could leave any additional remarks in a separate comment box. Mayonnaises (15-20 g) were presented in 30 mL transparent plastic cups labelled with random 3-digit codes, which were taken from the fridge 30 min prior to the start of the test session. A spoon was used to taste the mayonnaises. Participants could expectorate mayonnaises after evaluation. Crackers and water were provided for palate cleansing after evaluation of each sample. Data was collected in Qualtrics (Qualtrics, USA).

3.2.6. Data analysis

Intensity scores from sensory evaluation were reported as mean values with standard error. An intensity score of 0 was assigned to sensory attributes that were not selected by the participants. Two-way repeated measures ANOVA was performed on each of the sensory attributes (fixed factors: thickener type, concentration, thickener type:concentration interaction; random factor: participant). Bonferroni post-hoc tests were performed to determine statistically significant differences between samples. Principal Component Analysis (PCA) was performed and a bi-plot with 95% confidence ellipses was created. Multiple Factor Analysis (MFA) was performed to determine relationships between sensory, rheological and tribological properties of the mayonnaises. Data was analysed using RStudio (version 4.0.2) using the packages lmerTest (Kuznetsova *et al.*, 2017), emmeans (Lenth, 2021a), factoextra (Kassambara & Mundt, 2020) and FactoMineR (Lê *et al.*, 2008). A significance level of $\alpha = 0.05$ was used.

Table 3.2. *Sensory attributes used to evaluate twelve low-fat mayonnaises using RATA together with definitions, and examples of products high in intensity of the respective attribute.*

Attribute	Definition	Examples of products
Appearance		
Glossiness	The product has a shiny appearance, light is reflected from the surface of the product.	Olives, icing, custard
Sliminess	The product is thick, slippery and cohesive.	Gelatin pudding, oysters, raw egg white
Smoothness	The texture of the product is smooth and homogeneous; absence of lumps and grains.	Custard, milk, water (smooth) Cottage cheese (not smooth)
Thickness	The degree to which the product flows/deforms.	Greek yoghurt (thick) Water (not thick)
Yellowness	The intensity of the yellow colour.	Cauliflower, milk (white) Mustard, vanilla custard (yellow)
Flavour		
Fatty flavour	The intensity of the taste of fat.	Butter, whipped cream, French fries
Lemon flavour	The degree to which the product tastes like lemon.	Lemons, lemon zest, lemon curd
Saltiness	The intensity of the salt taste.	Salt, cheese, meat
Sourness	The intensity of the sour taste; acidity.	Citrus fruits, vinegar, yoghurt
Sweetness	The intensity of the sweet taste.	Sugar, lemonade
Texture		
Creaminess	The degree to which the product provides a silky, rich, full mouthfeel.	Ice cream, whipped cream
Melting	The degree to which the product becomes thin and fluid and distributes itself in the mouth.	Ice cream, chocolate
Mouthcoating	The feeling that a layer of the product remains behind on the palate (after swallowing).	Butter, oil, chocolate
Pulpiness	The product has a pulpy, mushy structure; the texture of the product is fibre-like.	Apple sauce, orange juice with pulp
Sliminess	The product is thick, slippery and cohesive in the mouth.	Gelatin pudding, oysters, raw egg white
Smoothness	The texture of the product is smooth and homogeneous; absence of lumps and grains.	Custard, milk, water (smooth) Cottage cheese (not smooth)
Stickiness	The degree to which the product sticks to the palate and teeth.	Honey, marshmallow, toffee
Thickness	The amount of force needed to make the sample flow or deform in the mouth.	Greek yoghurt (thick) Water (not thick)

3.3. Results & Discussion

3.3.1. Flow properties

Flow curves of low-fat mayonnaises thickened with different concentrations of microfibrillated cellulose (MFC), modified starch (MS), waxy corn starch (WCS) and xanthan gum (XG) are shown in Figure 3.1. All mayonnaises displayed shear-thinning behaviour. As expected, shear viscosity and consistency index *K* of the mayonnaises increased with increasing concentration of thickener (Table 3.3), which is consistent with results from previous studies on mayonnaises thickened with various biopolymers (e.g. Bortnowska & Tokarczyk, 2009; Golchoobi *et al.*, 2016; Heggset *et al.*, 2020; Lee *et al.*, 2013; Ma & Barbosa-Cánovas, 1995; Mozafari *et al.*, 2017; Mun *et al.*, 2009; Su *et al.*, 2010). Differences in viscosity and consistency index between thickener concentrations (low, medium, high) were small, which reflects the moderate variation in thickener concentrations used in the mayonnaises (Table 3.1). A narrow range of thickener concentrations was used in this study to represent concentrations used in commercial low-fat mayonnaises. Mayonnaises thickened with MFC and WCS generally had lower shear viscosities than mayonnaises thickened with MS or XG. Although applied in higher concentrations (Table 3.1), addition of WCS resulted in lower shear viscosities than addition of MS, which can be attributed to the improved resistance to shear and acidity of cross-linked MS (Chen *et al.*, 2018). On the other hand, shear viscosities of mayonnaises thickened with MFC and XG were similar at shear rates >100 s^{-1} due to more pronounced shear-thinning behaviour of XG-thickened mayonnaises. This can be observed from the flow index *n* (Table 3.3), as a lower flow index *n* indicates stronger shear-thinning behaviour. Our results support findings of Mozafari *et al.* (2017), who concluded that addition of xanthan gum to low-fat mayonnaises resulted in lower flow indices.

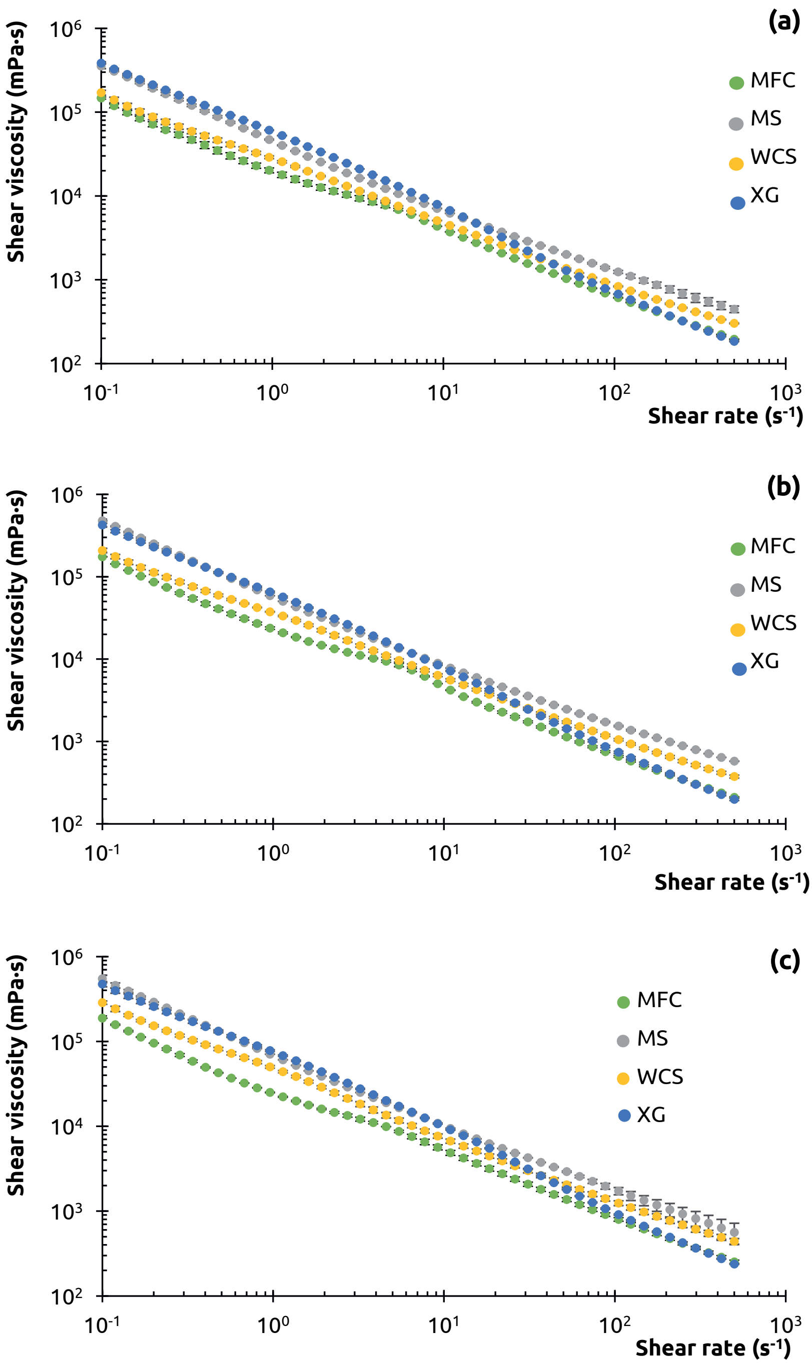

Figure 3.1. *Mean shear viscosity of low-fat mayonnaises with (a) low, (b) medium and (c) high concentrations of microfibrillated cellulose (MFC; green), modified starch (MS; grey), waxy corn starch (WCS; yellow) and xanthan gum (XG; blue)(2 replicates, error bars represent standard deviation).*

Table 3.3. *Rheological and tribological parameters of low-fat mayonnaises thickened with microfibrillated cellulose (MFC), modified starch (MS), waxy corn starch (WCS) and xanthan gum (XG) at low, medium and high concentration. Values are given as means (±SD). Consistency index (K) and flow index (n) were determined using the Ostwald-de Waele model. Yield stress σ_y was determined as stress applied at the intersect of G' and G" during a strain sweep at 1 Hz . $\mu_{BR(max)}$ represents the maximum friction coefficient in the boundary regime, $\mu_{HDR(start)}$ is the friction coefficient at the start of the hydrodynamic regime.*

	K (Pa·s^n)	n	σ_y (Pa)	$\mu_{BR(max)}$ (-)	$\mu_{HDR(start)}$ (-)
MFC-low	21.5 ± 1.4	0.24 ± 0.01	12.9 ± 1.4	0.191	0.123
MFC-medium	25.3 ± 0.2	0.22 ± 0.01	11.8 ± 1.1	0.192	0.131
MFC-high	27.6 ± 0.3	0.24 ± 0.01	15.3 ± 0.7	0.172	0.117
MS-low	47.2 ± 0.4	0.21 ± 0.01	44.1 ± 1.5	0.098	0.088
MS-medium	60.1 ± 0.4	0.21 ± 0.00	56.6 ± 0.7	0.102	0.086
MS-high	71.5 ± 0.5	0.19 ± 0.02	60.5 ± 0.4	0.106	0.089
WCS-low	27.2 ± 1.1	0.26 ± 0.01	6.8 ± 0.5	0.110	0.100
WCS-medium	34.7 ± 0.2	0.25 ± 0.01	6.5 ± 0.4	0.114	0.106
WCS-high	45.5 ± 2.0	0.23 ± 0.00	6.2 ± 0.5	0.116	0.093
XG-low	54.4 ± 0.5	0.08 ± 0.00	51.1 ± 0.1	0.124	0.079
XG-medium	58.7 ± 0.8	0.08 ± 0.01	60.6 ± 0.2	0.121	0.077
XG-high	69.7 ± 2.3	0.09 ± 0.01	69.3 ± 0.6	0.122	0.078

3.3.2. Viscoelastic properties

Storage modulus *G'* of all mayonnaises was larger than loss modulus *G"* up to strains of 5% (Figure 3.2), indicating that all mayonnaises exhibited solid-like behaviour in the linear viscoelastic region. The magnitude of *G'* and *G"* increased with increasing concentrations of biopolymers, which is in accordance with previous studies on mayonnaises thickened with MFC (Heggset *et al.*, 2020) or xanthan gum (Ma & Barbosa-Cánovas, 1995). An increase in thickener concentration results in the formation of a stronger network in the aqueous phase of the low-fat mayonnaises, which is reflected in larger *G'* and *G"*. Differences are observed when comparing the different biopolymers used to thicken the low-fat mayonnaises. Highest *G'* and *G"* were observed for mayonnaises thickened with MFC and lowest *G'* and *G"* for mayonnaises thickened with waxy corn starch. These values of *G'* and *G"* in MFC-thickened mayonnaises are in agreement with those found by Golchoobi *et al.* (2016) and Heggset *et al.* (2020), considering that these studies used lower concentrations of MFC (1 wt% and 0.25-0.42 wt%, respectively).

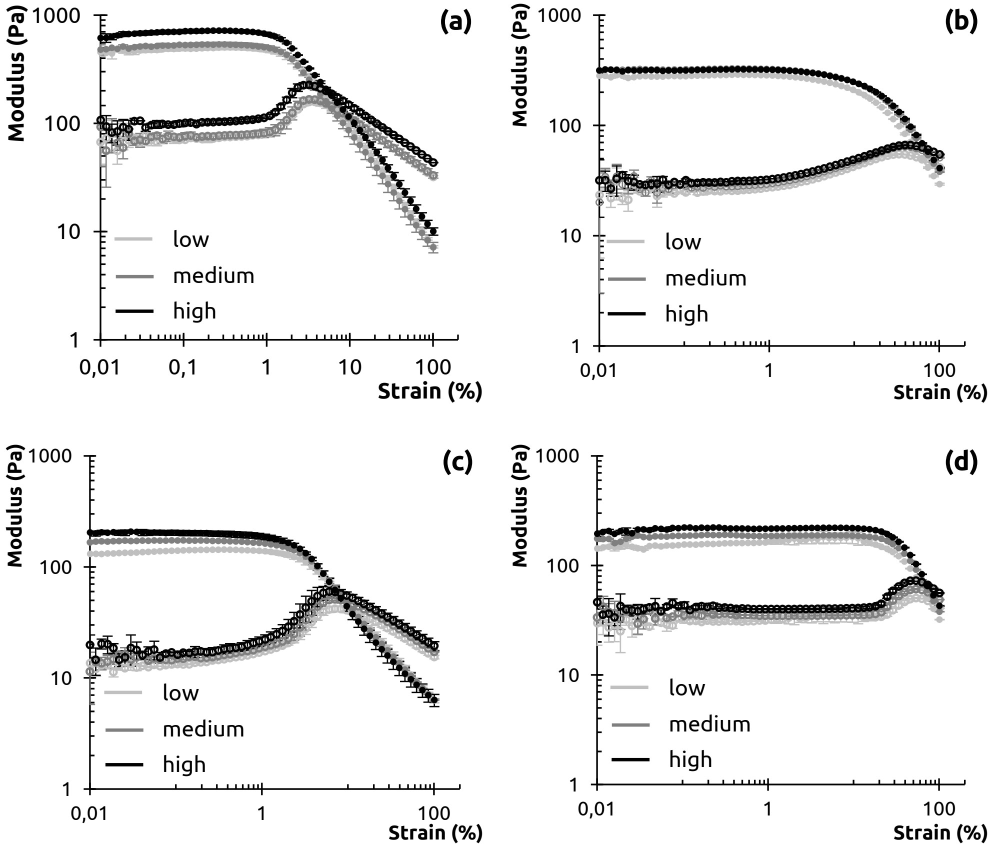

Figure 3.2. *Mean storage (G', filled symbols) and loss modulus (G", open symbols) of mayonnaises thickened with (a) microfibrillated cellulose (MFC), (b) modified starch, (c) waxy corn starch and (d) xanthan gum at low, medium and high concentration.*

Yield stress (σ_y) was determined from the crossover point between *G'* and *G"* (Table 3.3). Yield stress of low-fat mayonnaises generally increased with increasing concentration of thickener, which is in accordance with previous studies on fluids thickened with MFC (Agoda-Tandjawa *et al.*, 2010; Iotti *et al.*, 2011; Lowys *et al.*, 2001), starch (Evans & Haisman, 1980; Ross *et al.*, 2019), xanthan gum (Marcotte *et al.*, 2001; Ross *et al.*, 2019; Song *et al.*, 2006) and mayonnaises thickened with xanthan gum (Ma & Barbosa-Cánovas, 1995). Mayonnaises thickened with MS and XG exhibited the highest yield stresses, whereas yield stress of those thickened with MFC or WCS were at least 3 times smaller. The lower yield stress of low-fat mayonnaises with MFC or WCS cannot be attributed solely to a structural difference between MFC or WCS and the other biopolymers, because these mayonnaises also differ in shear viscosities (Figure 3.1).

3.3.3. Tribological properties

Tribological properties of the low-fat mayonnaises thickened with different biopolymers are shown in Figure 3.3. For all low-fat mayonnaises the boundary and mixed friction regimes are observed. The boundary regime occurs at low speeds or high loads, when the surfaces of the tribo-pair are in direct contact with each other and the lubricant (mayonnaise) is excluded from the gap (Stokes, 2012). Friction in the boundary regime therefore depends on the ability of (constituents of) the sample to form a lubricating boundary film, for example by surface adsorption. The friction curves demonstrate that friction in the boundary regime was mainly affected by the type of thickener rather than the concentration of thickener (Figure 3.3). Friction coefficients in the boundary regime ($\mu_{BR(max)}$; Table 3.3) were higher for mayonnaises thickened with MFC (0.17-0.19) compared to the other thickeners (0.10-0.12). We hypothesise that polymer adsorption occurred in mayonnaises thickened with MS, WCS and XG, whereas cellulose microfibrils were unable to form a boundary lubricating film on the tribological surfaces. The presence of starch lowers boundary friction, which has been attributed to the formation of an amylose film on the surface and/or a ball-bearing effect provided by intact starch granules (Morell *et al.*, 2017; Yakubov *et al.*, 2015; Zinoviadou *et al.*, 2008). Xanthan gum reduces friction by adsorption on PDMS and formation of a hydrated film (Stokes *et al.*, 2011). The large particle size (*i.e.* several micrometers in length) of MFC and their aggregates are expected to hinder its entrainment in the gap (Lavoine *et al.*, 2012; Lundahl *et al.*, 2018), which consequently inhibits formation of a lubricating film and results in high boundary friction.

In the mixed regime, friction is affected by boundary lubrication and the lubricant's bulk viscosity (Stokes, 2012). Although the differences are small, increased biopolymer concentration generally lowered friction coefficients for all biopolymers, which is consistent with earlier findings by Cassin *et al.* (2001), Malone *et al.* (2003a) and Garrec and Norton (2012). For mayonnaises thickened with MS or XG the hydrodynamic regime is observed at sliding speeds above 100 mm/s. In this regime the surfaces of the tribo-pair are fully separated by a layer of fluid and friction solely depends on bulk viscosity (Stokes, 2012). The onset of the mixed and hydrodynamic regimes shifts to lower sliding speeds for mayonnaises with highest shear viscosities, *i.e.* mayonnaises with MS or XG (Figure 3.1, Figure 3.3). This has previously been described (Chojnicka *et al.*, 2008) and is expected to result from a combination of the samples' shear viscosity and adsorption at the tribological surface (Bongaerts *et al.*, 2007; Stokes, 2012; Stokes *et al.*, 2011). Polymer adsorption can improve the wetting properties of the surfaces, thereby facilitating entrainment of the lubricant into the contact, which shifts the transitions to other regimes to lower velocities. Secondly, more viscous fluids will be entrained more easily than low viscous ones, enabling an earlier transition from the mixed to the hydrodynamic regime (Cassin *et al.*, 2001). The latter may also explain the discrepancy between our current results and previous results on aqueous MFC dispersions (Blok *et al.*, 2021). The higher viscosity of mayonnaise potentially promoted the entrainment of MFC between the tribopairs, while the continuous phase of simple aqueous MFC dispersions could not facilitate this. Moreover, shear-induced flocculation of MFC (Karppinen *et al.*, 2012) may have been hindered by the viscosity of mayonnaise, thereby limiting the formation of sheared gel particles that are expelled from the tribological gap.

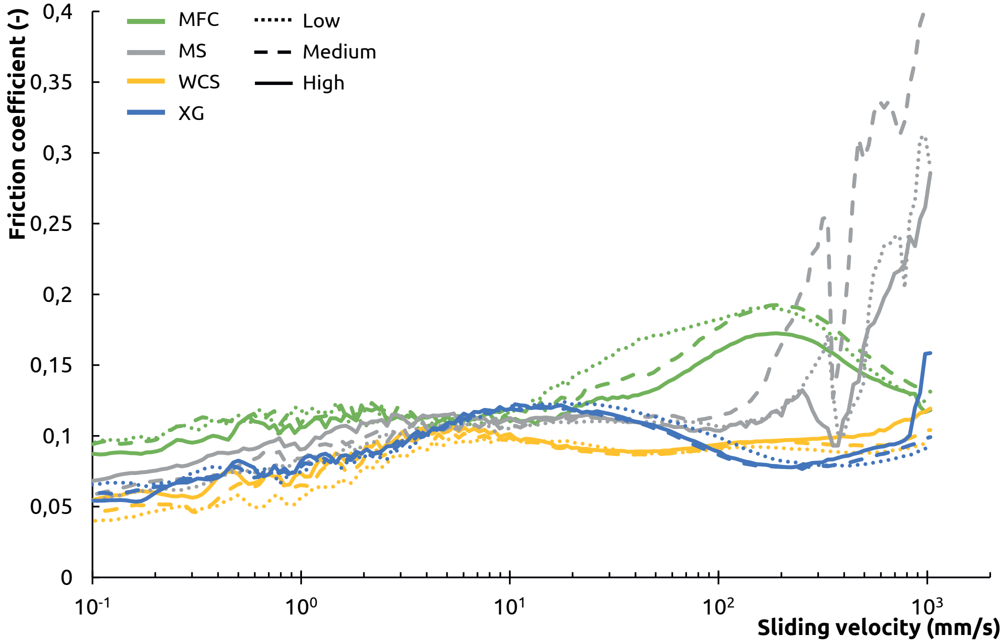

Figure 3.3. *Mean friction coefficients (triplicates) as a function of sliding speed of low-fat mayonnaises thickened with microfibrillated cellulose (MFC; green); modified starch (grey); waxy corn starch (yellow) or xanthan gum (blue). Dotted lines represent lowest concentration of thickener, dashed lines medium and solid lines highest concentration of thickener (See Table 3.1 for composition of the low-fat mayonnaises).*

3.3.4. Microstructure

Figure 3.4 shows CSLM images of low-fat mayonnaises containing the highest concentration of thickener (MFC-high, MS-high, WCS-high, XG-high). Oil droplets were generally smaller than 20 μm and were largest in mayonnaises thickened with MFC, followed by those thickened with XG. Addition of MFC or XG moreover resulted in polydisperse emulsions, whereas more monodisperse emulsions with smaller oil droplets (<5 μm, with the exception of a few large oil droplets) were obtained upon addition of MS or WCS to low-fat mayonnaises. As opposed to the uniform

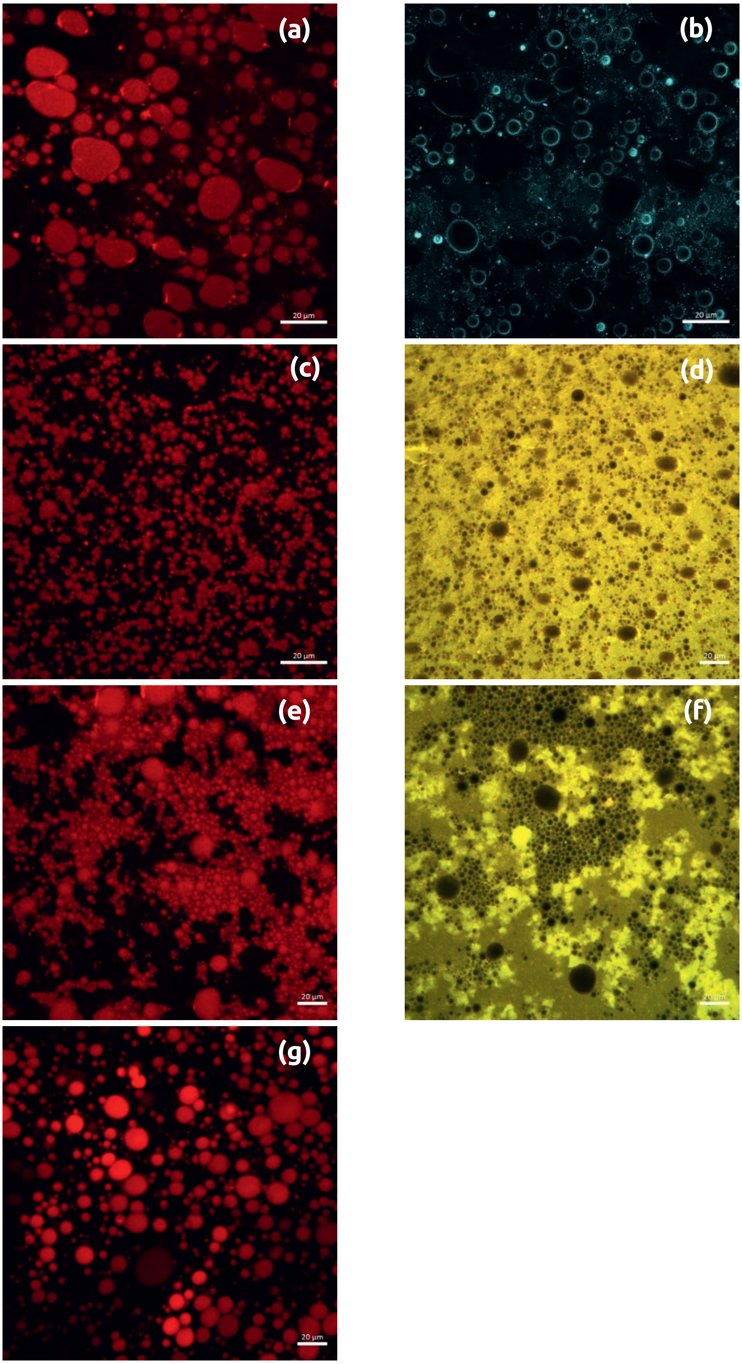

Figure 3.4. *Confocal laser scanning microscopy (CLSM) images of mayonnaises containing MFC (a,b), modified starch (c,d), waxy corn starch (e,f) and xanthan gum (g). The left column shows mayonnaises stained with Nile Red to visualize the fat phase, the right column shows the same mayonnaises stained with either Calcofluor White to visualise MFC (Figure 3.4b) or stained with Acridine Orange to visualise starch and protein (Figure 3.4d,f). Scale bars correspond to 20 µm.*

distribution of small oil droplets in MS-thickened mayonnaises, voids and clusters of aggregated oil droplets can be observed in mayonnaises thickened with WCS. The latter furthermore show uneven distribution of protein and starch throughout the emulsion, whereas a homogeneous network of starch and protein is present in mayonnaises with MS. Modification of starch is usually performed to improve its functional properties, including retrogradation and resistance to high temperature, high shear or low pH (Chen *et al.*, 2018). The relatively low pH of the mayonnaises (pH 3.6-3.9) could have affected the strength of the non-modified WCS network in the continuous phase of the mayonnaise, resulting in oil droplet aggregation (Figure 3.4e,f). This in turn can have an effect on the rheological properties of the mayonnaises, such as shear viscosity or yield stress (Figure 3.1, Table 3.3). MFC did not only form a microfibril network in the continuous phase of MFC-thickened mayonnaises, but was also present around the oil droplets (Figure 3.4b). There, MFC can act as an emulsifier on the oil-water interface, according to earlier studies in which the emulsifying properties of MFC have been established (Choublab & Winuprasith, 2018; Nomena *et al.*, 2018; Winuprasith & Suphantharika, 2013, 2015).

3.3.5. Sensory properties

Mean intensities of appearance, flavour and texture attributes of the low-fat mayonnaises are shown in Table 3.4. Type of thickener affected all sensory attributes, except for visual thickness (A-Thick) and sweetness (F-Sweet). The concentration of thickener significantly affected smooth and thick appearance, fatty and lemon flavour and smooth, sticky, thick, mouthcoating and melting texture. The Principal Component Analysis (PCA) bi-plot shows the position of the twelve mayonnaises in the sensory space (Figure 3.4). The first three principal components explain 40.6% of the total variation between the mayonnaises. Mayonnaises thickened with XG are separated from mayonnaises thickened with MFC, MS or WCS, which indicates that sensory perception of XG-thickened mayonnaises is different from the other mayonnaises. Mayonnaises thickened with MFC are located close to mayonnaises thickened with WCS and MS, which implies that sensory perception of mayonnaises thickened with MFC is similar to mayonnaise thickened with WCS and MS.

Table 3.4. *Mean intensities (± SE) of appearance, flavour and texture attributes obtained from RATA (n=80) of all mayonnaises. Samples in the same row containing the same letter are not significantly different from each other. Main effects (two-way ANOVA) of thickener type, concentration and their interaction with corresponding F- and p-values (n.s. = not significant; * p < 0.05; ** p < 0.01; *** p < 0.001) are reported.*

	Microfibrillated cellulose (MFC)			Modified starch (MS)			Waxy corn starch (WCS)			Xanthan gum (XG)		
	low	**medium**	**high**	**low**	**medium**	**high**	**low**	**medium**	**high**	**low**	**medium**	**high**
Appearance												
Glossy	4.3 ± 0.2^{d}	4.0 ± 0.2^{d}	3.9 ± 0.2^{d}	7.3 ± 0.2^{a}	7.1 ± 0.2^{a}	7.1 ± 0.2^{ab}	6.0 ± 0.2^{c}	5.8 ± 0.2^{c}	5.6 ± 0.2^{c}	5.9 ± 0.2^{c}	6.3 ± 0.2^{bc}	6.2 ± 0.2^{c}
Slimy	4.0 ± 0.3^{cd}	4.1 ± 0.3^{cd}	3.8 ± 0.2^{d}	4.6 ± 0.3^{bcd}	4.6 ± 0.3^{bcd}	4.5 ± 0.3^{cd}	4.9 ± 0.2^{abc}	4.7 ± 0.3^{bcd}	4.6 ± 0.3^{bcd}	5.4 ± 0.2^{ab}	5.4 ± 0.2^{ab}	5.7 ± 0.2^{a}
Smooth	6.9 ± 0.2^{abc}	6.0 ± 0.2^{de}	5.6 ± 0.2^{def}	7.5 ± 0.2^{a}	7.3 ± 0.2^{a}	7.1 ± 0.2^{ab}	6.4 ± 0.2^{bcd}	6.2 ± 0.2^{cd}	5.8 ± 0.2^{de}	4.8 ± 0.2^{fg}	5.3 ± 0.2^{efg}	4.5 ± 0.3^{g}
Thick	6.0 ± 0.2^{ab}	6.2 ± 0.2^{ab}	6.5 ± 0.2^{a}	5.8 ± 0.2^{ab}	6.3 ± 0.2^{ab}	6.4 ± 0.2^{a}	5.5 ± 0.2^{b}	6.0 ± 0.2^{ab}	6.5 ± 0.2^{a}	5.8 ± 0.2^{ab}	6.2 ± 0.2^{ab}	6.4 ± 0.2^{a}
Yellow	2.7 ± 0.2^{ab}	2.7 ± 0.2^{ab}	3.1 ± 0.2^{a}	2.0 ± 0.2^{c}	2.1 ± 0.2^{c}	2.1 ± 0.2^{c}	2.3 ± 0.2^{bc}	2.0 ± 0.1^{c}	1.9 ± 0.2^{c}	2.3 ± 0.2^{bc}	2.5 ± 0.2^{bc}	2.5 ± 0.2^{bc}
Flavour												
Fatty	5.0 ± 0.2^{abc}	4.8 ± 0.2^{bc}	5.2 ± 0.2^{abc}	5.4 ± 0.2^{abc}	5.7 ± 0.2^{a}	5.6 ± 0.2^{ab}	4.7 ± 0.2^{c}	5.0 ± 0.2^{abc}	5.4 ± 0.2^{abc}	4.7 ± 0.2^{c}	5.0 ± 0.2^{abc}	5.0 ± 0.2^{abc}
Lemon	5.9 ± 0.2^{a}	5.5 ± 0.2^{ab}	5.4 ± 0.2^{ab}	5.2 ± 0.2^{ab}	5.5 ± 0.2^{ab}	5.0 ± 0.2^{bc}	5.8 ± 0.2^{ab}	5.4 ± 0.2^{ab}	5.1 ± 0.2^{ab}	4.2 ± 0.2^{cd}	4.2 ± 0.2^{cd}	4.0 ± 0.3^{d}
Salty	3.7 ± 0.2^{a}	4.3 ± 0.3^{a}	4.0 ± 0.2^{a}	4.2 ± 0.2^{a}	3.9 ± 0.2^{a}	4.1 ± 0.2^{a}	4.3 ± 0.3^{a}	4.1 ± 0.2^{a}	4.2 ± 0.2^{a}	3.7 ± 0.3^{a}	3.5 ± 0.2^{a}	3.6 ± 0.3^{a}
Sour	4.9 ± 0.2^{a}	4.8 ± 0.2^{a}	4.4 ± 0.2^{abc}	4.4 ± 0.3^{abc}	4.7 ± 0.2^{a}	4.5 ± 0.3^{ab}	4.9 ± 0.2^{a}	4.6 ± 0.2^{a}	4.7 ± 0.2^{a}	3.6 ± 0.2^{c}	3.6 ± 0.2^{c}	3.7 ± 0.2^{bc}
Sweet	3.1 ± 0.2	3.1 ± 0.2	3.2 ± 0.2	3.2 ± 0.2	3.2 ± 0.2	3.2 ± 0.3	3.3 ± 0.2	3.1 ± 0.3	3.2 ± 0.2	3.0 ± 0.2	3.0 ± 0.2	2.9 ± 0.2
Texture												
Creamy	6.4 ± 0.2^{a}	6.4 ± 0.2^{a}	6.3 ± 0.2^{a}	6.4 ± 0.2^{a}	6.7 ± 0.2^{a}	6.8 ± 0.2^{a}	6.0 ± 0.2^{a}	6.0 ± 0.2^{a}	6.3 ± 0.2^{a}	4.4 ± 0.2^{b}	5.0 ± 0.2^{b}	4.7 ± 0.2^{b}
Melting	5.6 ± 0.2^{a}	5.4 ± 0.2^{ab}	5.3 ± 0.2^{ab}	5.4 ± 0.2^{ab}	4.7 ± 0.2^{bcd}	4.6 ± 0.2^{bcd}	5.7 ± 0.2^{a}	4.8 ± 0.2^{abc}	5.0 ± 0.2^{abc}	4.3 ± 0.3^{cd}	4.6 ± 0.3^{bcd}	3.9 ± 0.2^{d}
Mouthcoating	4.7 ± 0.2^{cd}	4.8 ± 0.2^{cd}	5.2 ± 0.3^{abcd}	5.1 ± 0.3^{abcd}	5.9 ± 0.2^{ab}	6.1 ± 0.2^{a}	4.6 ± 0.2^{d}	5.2 ± 0.3^{abcd}	5.6 ± 0.2^{abc}	4.6 ± 0.2^{d}	5.0 ± 0.3^{bcd}	4.9 ± 0.3^{cd}
Pulpy	0.7 ± 0.2^{d}	0.8 ± 0.2^{bcd}	0.8 ± 0.2^{cd}	0.6 ± 0.1^{d}	0.7 ± 0.1^{d}	0.8 ± 0.2^{cd}	0.9 ± 0.2^{bcd}	0.8 ± 0.2^{d}	0.7 ± 0.2^{d}	1.5 ± 0.3^{ab}	1.5 ± 0.2^{abc}	2.2 ± 0.3^{a}
Slimy	3.5 ± 0.3^{b}	3.5 ± 0.2^{b}	3.8 ± 0.2^{b}	3.8 ± 0.3^{b}	4.4 ± 0.3^{b}	4.1 ± 0.3^{b}	3.8 ± 0.3^{b}	3.9 ± 0.3^{b}	3.9 ± 0.3^{b}	5.6 ± 0.3^{a}	5.8 ± 0.3^{a}	5.9 ± 0.3^{a}
Smooth	7.2 ± 0.2^{a}	6.9 ± 0.2^{a}	6.9 ± 0.2^{a}	7.6 ± 0.1^{a}	7.3 ± 0.2^{a}	7.2 ± 0.2^{a}	7.2 ± 0.2^{a}	6.9 ± 0.2^{a}	6.9 ± 0.2^{a}	5.5 ± 0.3^{bc}	5.9 ± 0.2^{b}	4.8 ± 0.3^{c}
Sticky	2.9 ± 0.2^{d}	3.0 ± 0.3^{cd}	3.5 ± 0.3^{bcd}	3.7 ± 0.3^{bcd}	4.4 ± 0.3^{ab}	4.7 ± 0.3^{a}	3.3 ± 0.2^{cd}	3.3 ± 0.3^{cd}	3.8 ± 0.3^{abcd}	3.7 ± 0.3^{bcd}	3.9 ± 0.3^{abc}	3.9 ± 0.3^{abc}
Thick	4.4 ± 0.2^{e}	4.7 ± 0.2^{cde}	5.4 ± 0.2^{abc}	5.2 ± 0.2^{bcde}	6.2 ± 0.2^{a}	6.2 ± 0.2^{a}	4.5 ± 0.2^{de}	5.3 ± 0.2^{abcd}	5.6 ± 0.2^{ab}	4.8 ± 0.2^{bcde}	5.2 ± 0.2^{bcde}	5.7 ± 0.2^{ab}

Table 3.4 (continued). *Mean intensities (± SE) of appearance, flavour and texture attributes obtained from RATA (n=80) of all mayonnaises. Samples in the same row containing the same letter are not significantly different from each other. Main effects (two-way ANOVA) of thickener type, concentration and their interaction with corresponding F- and p-values (n.s. = not significant; * p < 0.05; ** p < 0.01; *** p < 0.001) are reported.*

	Two-way ANOVA effects					
	Thickener type		**Concentration**		**Interaction**	
	F(3,869)	*p*	*F*(2,869)	*p*	*F*(6,869)	*p*
Glossy	*F*=173.7	***	*F*=1.0	n.s.	*F*=1.3	n.s.
Slimy	*F*=34.9	***	*F*=0.2	n.s.	*F*=0.6	n.s.
Smooth	*F*=99.5	***	*F*=14.2	***	*F*=3.0	**
Thick	*F*=0.8	n.s.	*F*=15.9	***	*F*=0.6	n.s.
Yellow	*F*=26.9	***	*F*=0.9	n.s.	*F*=2.4	*
Flavour						
Fatty	*F*=9.1	***	*F*=3.7	*	*F*=0.8	n.s.
Lemon	*F*=36.4	***	*F*=4.6	*	*F*=1.0	n.s.
Salty	*F*=6.5	***	*F*=0.0	n.s.	*F*=1.4	n.s.
Sour	*F*=23.2	***	*F*=0.5	n.s.	*F*=1.0	n.s.
Sweet	*F*=1.4	n.s.	*F*=0.1	n.s.	*F*=0.1	n.s.
Texture						
Creamy	*F*=72.6	***	*F*=1.5	n.s.	*F*=1.0	n.s.
Melting	*F*=24.3	***	*F*=9.1	***	*F*=2.3	*
Mouthcoating	*F*=11.5	***	*F*=12.4	***	*F*=1.0	n.s.
Pulpy	*F*=33.3	***	*F*=2.1	n.s.	*F*=1.9	n.s.
Slimy	*F*=67.0	***	*F*=2.0	n.s.	*F*=0.6	n.s.
Smooth	*F*=86.3	***	*F*=7.2	***	*F*=2.6	*
Sticky	*F*=15.8	***	*F*=8.2	***	*F*=1.1	n.s.
Thick	*F*=16.8	***	*F*=30.1	***	*F*=1.1	n.s.

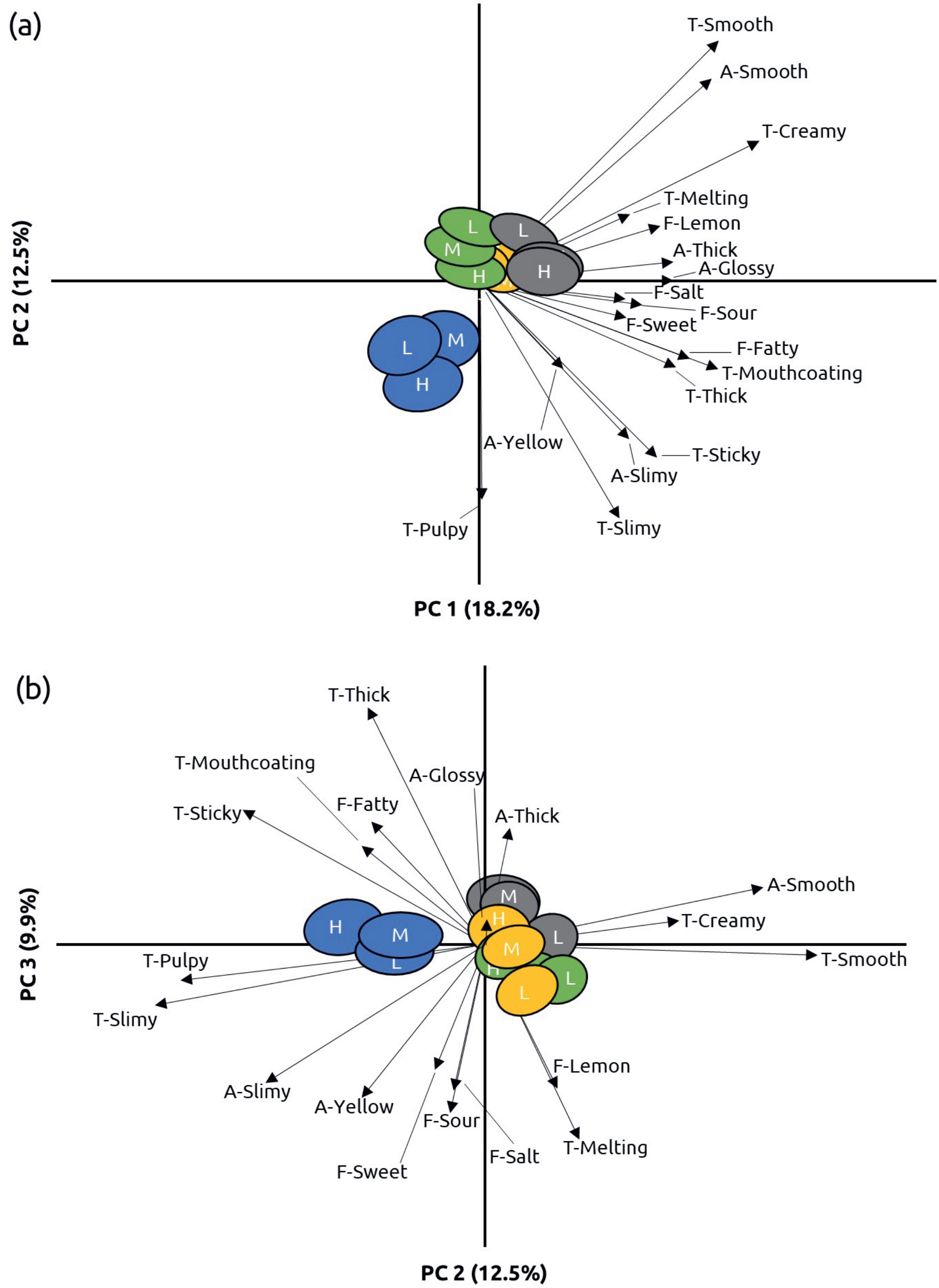

Figure 3.5. *Principal Component Analysis (PCA) bi-plots displaying loadings for the appearance, flavour and texture attributes and scores for the twelve low-fat mayonnaises thickened by microfibrillated cellulose (MFC; green), modified starch (grey), waxy corn starch (yellow) and xanthan gum (blue) with their respective 95% confidence ellipses. Letters in circles correspond to the concentration of thickener used: low (L), medium (M) and high (H) thickener concentration. Figure (a) displays principal components (PC) 1 and 2; (b) displays PC 2 and 3.*

Appearance

Mayonnaises thickened with MS were perceived as the glossiest, whereas mayonnaises thickened with MFC were the least glossy (Table 3.4). Although yellowness intensities of the mayonnaises were generally low, addition of MFC resulted in a slight increase of yellowness. The larger oil droplet size of MFC-thickened mayonnaises (Figure 3.4) might have caused this difference in appearance. Larger oil droplets lead to increased yellowness and reduced light scattering efficiency, resulting in decreased glossiness (Chantrapornchai *et al.*, 1998; Chantrapornchai *et al.*, 1999; Winuprasith & Suphantharika, 2015). Visual smoothness generally decreased for higher concentrations of thickener, except for mayonnaises thickened with XG. These mayonnaises had a less smooth but slimier appearance than the other mayonnaises. As anticipated, visual thickness increased with increasing thickener concentration, yet it was not affected by the type of thickener used. Participants did not observe differences in visual thickness between mayonnaises thickened with different biopolymers although flow properties and shear viscosities of the mayonnaises differed (Figure 3.1), suggesting that the rheological differences between mayonnaises differing in the type of thickener added were too small to cause changes in visual thickness.

Flavour

No perceptual differences between the twelve mayonnaises were found for sweet and salty taste, and no clear trend in terms of fatty flavour intensity was observed. As the same concentrations of salt, sugar and fat were used in all mayonnaises, this was expected. Mayonnaises thickened with XG had lower sourness and lemon flavour intensities compared to mayonnaises thickened with MFC, MS or WCS. We hypothesise that this is caused by the texture of mayonnaises thickened with XG, as multiple participants indicated that these mayonnaises had a cohesive gel-like texture that did not distribute well in the mouth upon oral processing. The cohesiveness of these mayonnaises might have resulted in a smaller surface area that is in contact with the tongue and saliva, and consequently might have reduced flavour release from the matrix. Our results confirm those of Pangborn and colleagues (1978) who observed that xanthan gum suppressed sourness and flavour intensity in thickened beverages. Furthermore, several participants mentioned the presence of an off-flavour for mayonnaises thickened with WCS, which is in accordance with previous findings (Lotong *et al.*, 2003; Matta *et al.*, 2006).

Texture

As anticipated, perceived thickness increased with increasing concentrations of thickener. Highest thickness intensities were found for mayonnaises thickened with MS, whereas lowest intensities were found for mayonnaises thickened with MFC. This was expected, since mayonnaises thickened with MFC also had the lowest shear viscosities (Figure 3.1). Similar to thickness, mouthcoating and stickiness intensities generally increased with increasing thickener concentration, which is in line with results of Ross *et al.* (2019). Perceived mouthcoating was slightly higher for mayonnaises thickened with MS compared to the other mayonnaises. Highest stickiness intensities were found for mayonnaises thickened with MS and XG, while MFC was the least sticky. This is in line with previous studies that reported high adhesiveness or stickiness in samples thickened with starch (Nguyen *et al.*, 2017; Ong *et al.*, 2018b) and xanthan gum (Blok *et al.*, 2021; Bortnowska & Tokarczyk, 2009). Mayonnaises thickened with MFC, MS or WCS were very similar in terms of creaminess, pulpiness, sliminess and smoothness (Table 3.4, Figure 3.5). Mayonnaises thickened with XG differentiated themselves from the other mayonnaises by high sliminess and low creaminess, melting and smoothness intensities. This confirms earlier work in which foods thickened with XG imparted high sliminess (Blok *et al.*, 2021; Gössinger *et al.*, 2018; Matta *et al.*, 2006), but low creaminess and smoothness (Nguyen *et al.*, 2017; Terpstra *et al.*, 2009). Although pulpiness intensities were generally low, pulpiness increased upon addition of XG. Pulpiness was included as a sensory texture attribute since MFC consists of insoluble fibres. Model foods thickened with MFC have previously been reported to be pulpier than those thickened with xanthan gum (Blok *et al.*, 2021). However, in the current study higher pulpiness was observed for mayonnaises with XG, which is also reflected in Figure 3.5. This is consistent with findings that XG induced heterogeneity in mayonnaises (Terpstra *et al.*, 2009), considering their definition of heterogeneity was similar to our definition of pulpiness. An alternative explanation could be that participants did not experience pulpiness in any of the mayonnaises and hence used this attribute to evaluate the aforementioned cohesive, gel-like texture of XG-thickened mayonnaises. This would confirm earlier findings, as products thickened with XG were found to exhibit cohesiveness (Ross *et al.*, 2019; Tobin *et al.*, 2020) and cohesiveness of mayonnaises increased with increasing concentrations of XG (Bortnowska & Tokarczyk, 2009).

3.3.6. Linking sensory to rheological and tribological properties of low-fat mayonnaises

In order to link sensory characteristics to rheological and tribological properties of the low-fat mayonnaises with different thickeners, Multiple Factor Analysis (MFA) was performed (Figure 3.6). Parameters located close to each other are positively correlated, whereas parameters opposing each other are negatively correlated. Since oral and visual thickness perception are located close to several shear viscosity parameters, these are positively correlated, as anticipated (e.g. Akhtar *et al.*, 2005; Cutler *et al.*, 1983). Yield stress and consistency index *K* are positively correlated with shear viscosity at low shear rates (η at 10 s^{-1}), which indicates that mayonnaises with higher viscosity at low shear rates also had higher yield stress and consistency index *K*. Flow index *n* is negatively correlated with sliminess, suggesting that stronger shear-thinning mayonnaises were perceived as slimier. This is in contrast with previous work in which sliminess was found to be associated with higher *n*-values (*i.e.* weaker shear-thinning behaviour) (Szczesniak & Farkas, 1962; Wood, 1974) or small deformation viscosity at 50 rad·s^{-1} (Richardson *et al.*, 1989). Many of the flavour and taste attributes (F-Lemon, F-Sour, F-Salty, F-Sweet) are located close to each other and opposite to sliminess, suggesting that samples with low sliminess generally had higher flavour intensities. This correlation might be driven by the XG-thickened mayonnaises as these were slimy and had lower flavour intensity. This correlation might therefore be specific for the current sample set and might not be generalisable to other foods. Sliminess and pulpiness are moreover placed opposite of smoothness and creaminess, which implies that mayonnaises with higher pulpiness and sliminess were perceived as less smooth and less creamy. The fact that creaminess is negatively correlated with pulpiness is consistent with findings of Terpstra *et al.* (2009), considering the definition they used to evaluate the homogeneity of mayonnaises is similar to the definition used for pulpiness in the current study.

A strong positive correlation between friction at the start of the hydrodynamic regime ($\mu_{HDR(start)}$) and melting texture was found. Since friction in the hydrodynamic regime predominantly depends on the shear viscosity of the material between the surfaces (Stokes, 2012), a higher friction coefficient at the start of the hydrodynamic regime is likely caused by a lower shear viscosity of the mayonnaise. Less viscous mayonnaises might have been perceived as more melting, as these rapidly become thin in the mouth and melting texture was defined in this study as 'the degree to which the product becomes thin and fluid and distributes itself in the mouth'. This corresponds with results from a study by de Wijk and colleagues, in which 'melting' was regarded as the semantical opposite of 'thick' (de Wijk *et al.*, 2003). Maximum friction coefficient in the boundary regime ($\mu_{BR(max)}$) is negatively correlated with a sticky texture, indicating that mayonnaises with higher friction in this regime were less sticky. While this is inconsistent with findings of Devezeaux de Lavergne *et al.* (2016), de Wijk and Prinz (2005) also concluded that increased friction generally resulted in decreased sensations of stickiness of custard desserts. In the boundary regime the surfaces of the tribo-pair are in contact and thus exclude the sample from the tribological gap. The friction in this regime is therefore largely determined by the properties of the surfaces, although these can be affected by adsorption of molecules to the surfaces (Stokes, 2012). Adsorption of biopolymers on (one of) the surfaces could reduce friction in the boundary regime by film formation on the one hand, and might induce stickiness on the other hand. It should be noted that saliva was not included in the experimental set-up and that friction properties were determined using a glass ball and PDMS pins, which does not perfectly resemble the oral environment.

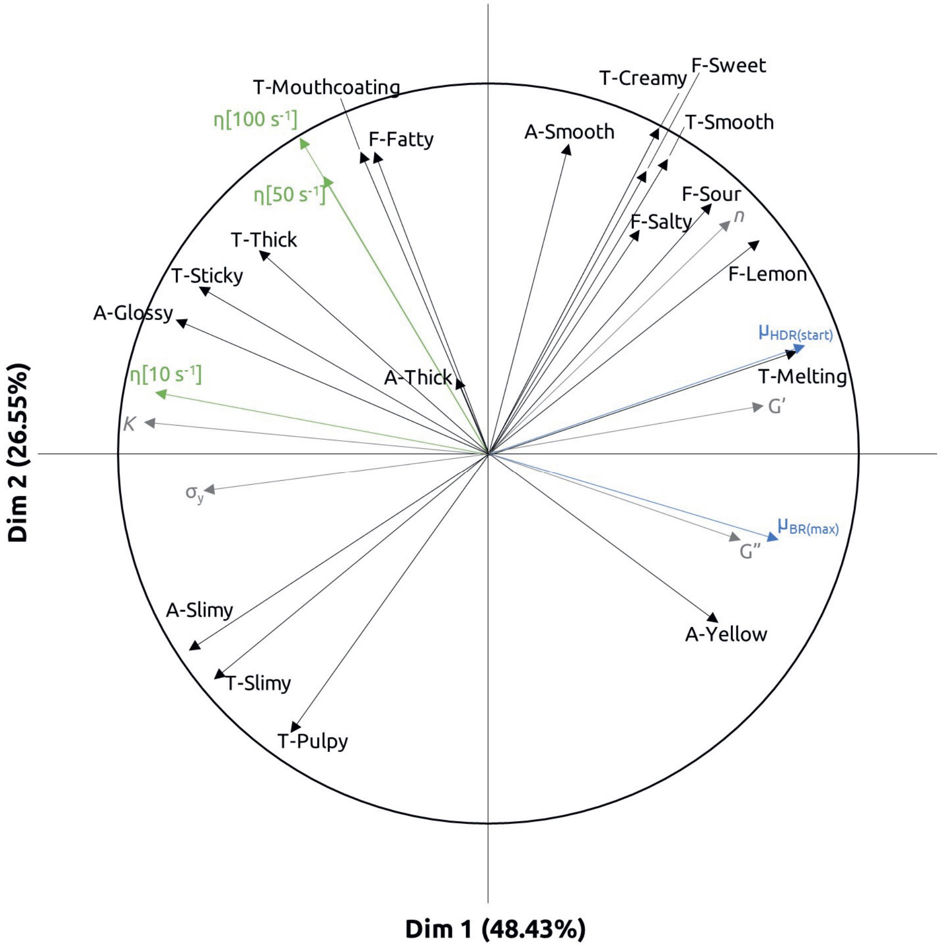

Figure 3.6. *Multiple Factor Analysis (MFA) plot including all sensory attributes (black), shear viscosity parameters (η at 10, 50 and 100 s^{-1}; green), rheological parameters (consistency index K, flow index n, storage modulus G', loss modulus G" and yield stress σ_y; grey) and friction parameters (maximum μ in the boundary regime ($\mu_{BR(max)}$) and μ at the start of the hydrodynamic regime ($\mu_{HDR(start)}$) ; blue).*

3.4. Conclusions

The aim of this study was to determine the effect of type and concentration of thickener on rheological, tribological and sensory properties of low-fat mayonnaises. Independent of thickener type, higher biopolymer concentrations generally led to increased shear viscosities, *G'* and *G"*, yield stress and enhanced lubrication (*i.e.* lower friction). Increasing the biopolymer concentration moreover enhanced several sensory texture attributes, including thickness, stickiness and mouthcoating. Rheological properties of low-fat mayonnaises depended on the type of biopolymer used. Addition of xanthan gum resulted in low-fat mayonnaises with strong shear-thinning behaviour and high yield stress compared to mayonnaises with modified starch, waxy corn starch or MFC, despite having comparable shear viscosities (>10 s^{-1}). Mayonnaises thickened with xanthan gum were furthermore sensorially different from the other mayonnaises due to higher sliminess and pulpiness, but low creaminess, smoothness and melting intensities. Sensory texture characteristics of mayonnaises with MFC closely resembled those of mayonnaises thickened with waxy corn starch and modified starch, despite differences in microstructure, rheological and tribological properties (*i.e.* lower shear viscosity and yield stress, higher *G'*, *G"* and friction, larger oil droplet size). We therefore conclude that MFC can be used as thickener in low-fat food mayonnaise as a substitute for conventionally used modified starch without greatly affecting sensory properties of the mayonnaise. The use of natural fibres such as MFC offers several benefits compared to the use of (modified) starch, including a low nutritional value, the potential of being used as a clean-label thickener and the possibility to use agricultural waste streams for its production.

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4

Spray-dried microfibrillated cellulose particles as texture modifier in liquid foods and their effect on rheological, tribological and sensory properties

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Abstract

Microfibrillated cellulose (MFC) has potential to be used as clean label texture modifier in foods due to its structural and mechanical properties. These properties deteriorate upon drying of MFC dispersions due to aggregation of the microfibrils. In this study dried MFC particles were prepared by spray-drying MFC dispersions in a surplus of maltodextrin to prevent hornification. The aim of this study was to evaluate the effect of MFC particle concentration and MFC:maltodextrin ratio of dried MFC powders on rheological, tribological and sensory texture properties of liquid foods. Scanning Electron Microscopy demonstrated that after spray-drying, MFC powders with polydisperse particle size distribution were obtained (1-30 µm). Upon suspension in water, maltodextrin dissolved in the aqueous continuous phase whereas spherical MFC networks retained their shape and co-existed in a mixture with individual fibrils. Spray-dried MFC powders were added to skimmed milk and tomato soup at different concentrations. With increasing concentration of dried MFC particles, shear viscosity, consistency index *K*, storage and loss modulus of skimmed milks and tomato soups increased whereas flow index *n* decreased. Addition of spray-dried MFC particles to milks and soups significantly ($p < 0.05$) increased sensory thickness and creaminess. Milks and soups with varying concentrations of MFC particles displayed similar tribological properties, which was presumably caused by exclusion of the MFC network from the tribological gap. Low MFC:maltodextrin ratio particles enhanced rheological and sensory texture properties of soups and milks more effectively than high MFC:maltodextrin ratio particles. We conclude that spray-dried microfibrillated cellulose particles can be used as thickener or fat replacer in liquid foods.

4.1. Introduction

Cellulose is the most abundant renewable biopolymer on earth, consisting of long chains of β-(1-4)-linked D-glucose assembled into microfibrils (Wüstenberg, 2014). It can be obtained from agricultural crops and waste streams such as fruit and vegetable pulp or peels, which offers environmental benefits (e.g. Koul *et al.*, 2021; Usmani *et al.*, 2021; Zain *et al.*, 2014). The water insolubility of unrefined cellulose however hampers its application in foods and has led to the development of several cellulose nanomaterials, including rod-like cellulose nanocrystals (CNC) and microfibrillated cellulose (MFC). In contrast to CNC, which undergoes hydrolysis to remove amorphous cellulose, microfibrillated cellulose consists of both crystalline and amorphous cellulose (Lavoine *et al.*, 2012). MFC is prepared by disintegration of cellulose fibres using mechanical treatment (usually high-pressure homogenisation, microfluidization or grinding) resulting in delamination of the cellulose fibres and the release of individual microfibrils. In polar liquids such as water, an entangled fibrous network is formed and a stable dispersion is obtained (Siró & Plackett, 2010). The mechanical properties of the fibrillated material are improved due to its enlarged surface area, which makes MFC dispersions suitable as texture modifiers in foods (e.g. Blok *et al.*, 2021; Gómez *et al.*, 2016; Ström *et al.*, 2013).

MFC has thus far mostly been applied and studied in dispersed form. Transport and storage of MFC dispersions is inefficient and expensive, which hampers the development of foods with MFC as texture modifier. One of the main challenges of applying MFC in food products has been the aggregation of microfibrils upon drying (Déléris & Wallecan, 2017). The enhanced surface area of MFC gives rise to the exposure of additional hydroxyl groups, which can form strong hydrogen bonds with hydroxyl groups of adjacent cellulose molecules. Upon drying of MFC dispersions, water removal causes the cellulose fibres to move closer to each other and aggregate, a phenomenon referred to as hornification. Hornification induces irreversible binding of the fibrils and increases the crystallinity of the material. The process essentially reverses the fibrillation procedure, resulting in inferior mechanical properties upon resuspension of dried MFC. Several studies investigated how to improve the drying of MFC. Different drying and preparation methods have been compared (e.g. Eyholzer *et al.*, 2010; Jiang & Hsieh, 2014; Peng *et al.*, 2013; Silva *et al.*, 2021; Yan *et al.*, 2016) and drying MFC in the presence of other components has been described (Butchosa & Zhou, 2014; Cantiani *et al.*, 2002; Lowys *et al.*, 2001; Missoum *et al.*, 2012). A promising method to prevent hornification was proposed recently by Velásquez-Cock *et al.* (2018), who mixed MFC dispersions with various amounts of maltodextrin prior to drying. The described method includes removal of maltodextrin by vacuum filtration and hot water rinsing following redispersion of the dried material yielding stable MFC dispersions. However, the processing steps required for the removal of maltodextrin might limit the applicability of these processes for incorporation of MFC in foods.

In the present study, MFC was prepared in a surplus of maltodextrin and subsequently spray-dried to obtain dried MFC particles. MFC and maltodextrin were spray-dried in two different ratios. The two types of spray-dried MFC were applied in two liquid foods: skimmed milk and instant tomato soup. The effect of concentration of spray-dried MFC particles and MFC:maltodextrin ratio on rheological, tribological and sensory texture properties of both liquid foods were determined in order to assess whether spray-dried MFC particles can be used as texture modifier in liquid foods.

4.2. Materials & Methods

4.2.1. Preparation of spray-dried MFC particles

Two types of spray-dried MFC powders were prepared differing in MFC:maltodextrin (MFC:MD) ratio: (i) 1:25 with 3.8 wt% MFC in the final powder (MFC:MD-Low) and (ii) 1:7.5 with 11.6 wt% MFC in the final powder (MFC:MD-High) (Table 4.1). Citrus fibre powder (HERBACEL® AQ® Plus, Herbafood Ingredients, Werder, Germany) containing ~65 wt% cellulose (Fechner *et al.*, 2013) was dispersed in demineralised water using a L5M-A Silverson laboratory mixer operated at 8,000 rpm for 15 min (Silverson Machines Ltd., Chesham, United Kingdom). A second mixing step of 15 min was performed after addition of maltodextrin (Glucidex® 29, Roquette, Lestrem, France), followed by addition of tricalcium phosphate (anhydrous ($Ca_3(PO_4)_2$; Brenntag, Essen, Germany) and mixing for 2 min. The mixture was subsequently homogenised using a lab scale high-pressure microfluidizer (Microfluidizer M-110S, Microfluidics™, Newton, MA, USA) equipped with a 200 μm Z-cell at 900 bar, to obtain microfibrillated cellulose. This was followed by a second passage through the same microfluidizer equipped with an 87 μm Z-cell at 1200 bar. The resulting slurry was spray-dried (Mini Spray Dryer B-290, Büchi Labortechnik AG, Flawil, Switzerland) using a 1.5 mm two-fluid nozzle having an atomizing pressure of 2 bars. The inlet (controlled) and outlet (measured) air temperatures were 165°C and 60-75°C, respectively. The flow rate was set at 0.3-0.5 L/h (=15-25% pump). Different set flow rates were used because the actual flow rate depends on the viscosity of the sample. Lower flow rates were used when the measured outlet temperature became lower than 65°C. A gas flow rate of 30 was used and the nozzle cleaner was set at 5. The composition and other details of the dispersions and the resulting spray-dried powders can be found in Table 4.1. The obtained spray-dried powders were stored in glass jars for up to three months until further use.

Table 4.1. *Composition of the two spray-dried MFC powders differing in MFC:maltodextrin (MFC:MD) ratio before and after spray-drying (in wt%). * Moisture content after spray-drying was not quantified.*

	Before spray-drying		**After spray-drying***	
	MFC:MD-Low (1:25)	**MFC:MD-High (1:7.5)**	**MFC:MD-Low (1:25)**	**MFC:MD-High (1:7.5)**
Citrus fibre	0.3	1	3.8	11.6
Maltodextrin	7.5	7.5	94.3	86.7
Calcium phosphate	0.15	0.15	1.9	1.7
Demineralised water	92.05	91.35	*	*

4.2.2. Sample preparation

Spray-dried MFC powders were dispersed at different concentrations in skimmed milk (0% fat milk, Albert Heijn, Zaandam, the Netherlands) and instant tomato soup (Knorr tomato drink bouillon (<0.5% fat), Unilever, London, United Kingdom). Table 4.2 presents the final composition of the liquid foods. The concentration of maltodextrin in all liquid samples was constant (7.5 wt%) to standardise potential effects of maltodextrin on flavour and mouthfeel. Differences in rheological, tribological and sensory properties between samples are therefore related to MFC. In order to compare the two types of spray-dried MFC powder (MFC:MD-Low vs MFC:MD-High), a concentration of MFC:MD-High was selected to match the final MFC concentration to that in M-L8 and S-L8 (Table 4.2). Commercial full-fat milk (3.6% fat; Albert Heijn, the Netherlands) was added as a reference to the skimmed milk sample set.

For the skimmed milks, spray-dried MFC powder and maltodextrin were gradually added to the skimmed milk under continuous stirring on a magnetic stirrer plate at room temperature. Samples were stirred for 45 min, until all ingredients were fully dissolved. Instant tomato soups were prepared according to the instructions on the product. 175 ml of boiling water was added to 8.1 g instant soup powder and stirred for 5 min at room temperature. The soup was subsequently cooled using an ice bath and sieved to remove pieces of herbs and undissolved ingredients. Spray-dried MFC and maltodextrin were gradually added to the soups while stirring at room temperature for 60 min until all ingredients were dissolved.

Table 4.2. *Composition of the liquid samples used for rheological, tribological and sensory analyses. Skimmed milks and instant tomato soups were used as liquid matrices. Full-fat milk was used as a commercial reference product for set of milks.*

	Sample	MFC:maltodextrin ratio Low (1:25) (wt%)	MFC:maltodextrin ratio High (1:7.5) (wt%)	Maltodextrin (MD) (wt%)	Matrix (wt%) (soup / milk)	Final composition of milks and soups MFC (wt%)	Final composition of milks and soups Maltodextrin (MD) (wt%)
Milk	M-0	-	-	7.5	92.5	-	7.5
	M-L2	2	-	5.6	92.4	0.075	7.5
	M-L4	4	-	3.7	92.3	0.15	7.5
	M-L8	8	-	-	92.0	0.3	7.5
	M-H2.6	-	2.6	5.3	92.1	0.3	7.5
	M-FF	-	-	-	100 (full-fat)	-	-
Soup	S-0	-	-	7.5	92.5	-	7.5
	S-L2	2	-	5.6	92.4	0.075	7.5
	S-L4	4	-	3.7	92.3	0.15	7.5
	S-L8	8	-	-	92.0	0.3	7.5
	S-H2.6	-	2.6	5.3	92.1	0.3	7.5

4.2.3. Scanning Electron Microscopy (SEM) and Cryo Scanning Electron Microscopy (cryoSEM)

Spray-dried MFC powders were characterised using Scanning Electron Microscopy (SEM). For this the powders were fixed on SEM stubs using carbon adhesive tabs (EMS Washington USA) and sputter coated with 12 nm Tungsten (Leica EM SCD 500, Vienna, Austria). SEM images were recorded using a FEI Magellan 400 at an acceleration voltage of 2.0 kV and 13 pA. Secondary electrons (SE) were detected using an Everhart-Thornley detector at a working distance of ~4.7 mm.

The behaviour of spray-dried MFC powder in aqueous matrices was characterised using cryoSEM on a dilute suspension of MFC:MD-Low in water. For the cryoSEM images a small droplet of the suspension was placed between aluminium (HPF) platelets (Wohlwend, Sennwald, Switzerland). The sample was then frozen by plunging in liquid ethane. The frozen samples were transferred to a cryo-preparation system (MED 020/VCT 100, Leica, Vienna, Austria) onto a sample stage at -92˚C in high vacuum. In this cryo-preparation system the samples were freeze-fractured and kept for 15 minutes at -92˚C for ice sublimation and etching of the samples. After coating with 12 nm of tungsten, the samples were transferred under vacuum to the field emission scanning microscope (Magellan 400, FEI, Eindhoven, the Netherlands) on the sample stage at -120°C. SEM images were recorded at an acceleration voltage of 2.0 kV and 13 pA.

4.2.4. Rheology: Flow and viscoelastic properties

Flow and viscoelastic properties of the samples were determined using an MCR 302 rheometer (Anton Paar, Graz, Austria) equipped with a concentric cylinder (ø = 17 mm). A double gap measuring system (ø = 26.7 mm) was used for M-0 and M-FF due to the low viscosity of these samples. To determine flow properties, shear viscosity was measured while increasing the shear rate from 0.1-1000 s^{-1} in 50 logarithmic steps. Flow curves were fitted to the Ostwald-de Waele power law model from which consistency index *K* and flow index *n* were determined: $\sigma = K \times \dot{\gamma}^n$, in which σ = shear stress (Pa), $\dot{\gamma}$ = shear rate (s^{-1}), *K* = consistency index ($Pa{\cdot}s^n$) and *n* = flow index. Viscoelastic properties were determined by increasing the strain from 0.1-100% under a constant oscillation frequency of 1 Hz. Values of G′ and G″ were recorded at 1% shear strain. All rheological measurements were performed in triplicate at 35°C to mimic the temperature in the oral cavity. Each measurement was preceded by a 5 min waiting step to allow for structural relaxation of the sample after loading.

4.2.5. Tribology: Friction properties

Tribological (friction) properties were determined using an MCR 302 rheometer (Anton Paar, Austria) equipped with a tribological set-up (T-PTD 200). A ball-on-three-pins set-up was used with a glass ball and three polydimethylsiloxane (PDMS) pins. 600 µl of the sample was loaded into the sample cup. Measurements were performed in triplicate at 35°C. Each measurement consisted of two runs in which the rotational speed was increased from 0.0001 to 2200 rpm (equivalent to 0.00004-1000 mm/s). Each run was preceded by a 5 min resting period to allow for structural relaxation of the sample. A normal force of 1 N was applied during runs and waiting steps. New pins were conditioned by performing one run (0.0001-2200 rpm) with 1 ml demineralised water, followed by a run with 1 ml of the respective sample without MFC particles (M-0 or S-0). Data from the second run was used for analysis, as the first run showed variation due to running-in effects and adjustments in the alignment of the pins. Maximum friction μ_{max} was determined as the maximum friction coefficient over the entire speed range. Since for soups the mixed regime was considered to start immediately after reaching the maximum in the boundary regime (μ_{max}), the sliding speed at which the mixed regime starts was extracted as the speed at which μ_{max} occurred. This parameter was determined manually for milks, as μ_{max} did not represent the start of the mixed regime for these samples. The slope in the mixed regime was determined as the slope between the start of the mixed regime until the friction measured at 1000 mm/s.

4.2.6. Sensory evaluation

Participants were recruited from Wageningen and surroundings. Individuals were excluded from participation in the study when they were allergic or intolerant to milk or when they had diabetes. Women were excluded from participation when they were pregnant or breastfeeding. Participants (*n* = 72; 18 male, 53 female, 1 other) were between 18-37 years old (mean 24 ± 3 y), had a BMI between 17.9-28.7 kg/m^2 (mean 21.9 ± 2.6 kg/m^2) and were non-smokers. Participants from 17 different countries were recruited, and most were students of Wageningen University. Participants received financial reimbursement upon completion of the study. The study did not meet the requirements to be reviewed by the Medical Research Ethical Committee of The Netherlands according to the "Medical Research Involving Human Subjects Act" of The Netherlands. The study was conducted in agreement with the ethics regulations laid out in the Declaration of Helsinki (2013). All participants gave written informed consent.

The consumer panel evaluated milks and soups in two sessions of 45 min on different days. The order in which participants attended the two test sessions was randomised. Samples were evaluated using the rank-rating method, in which all samples were presented simultaneously and evaluated for one attribute at a time. Samples were evaluated for thickness and creaminess, see Table 4.3 for attribute definitions and examples of products low and high in intensity of the respective attribute. The order in which participants evaluated the two attributes was randomised, new samples (with different 3-digit codes) were provided when participants moved to evaluation of the second sensory attribute. Samples within a sample set (5 samples for soup, 6 samples for milk) were presented simultaneously in random order, in transparent 30 ml cups labelled with random 3-digit codes. Participants were instructed to focus on the mouthfeel of the samples, disregarding any potential differences in appearance or flavour. Participants evaluated the samples by tasting the samples and placing the 3-digit codes corresponding to the samples on a 100 mm unstructured line scale representing the intensity of the attribute. In this way all samples were placed on the same line scale, which was anchored *'weak'* on the left and '*strong'* on the right end. A comment box was provided in case participants had any additional comments about the sample set. Participants were allowed to expectorate the sample after evaluation, and were asked to rinse their mouth with water between different samples. Between evaluation of the first and second attribute, participants cleansed their palate with crackers and water. Each test session was preceded by a familiarisation task to become acquainted with the test methodology and the attributes to be evaluated. Data was collected in English in EyeQuestion® (Logic8, the Netherlands).

Table 4.3. *Sensory attribute definitions and examples of products with high and low intensity of the attributes.*

Sensory attribute	Definition	Examples
Thickness	The amount of force needed to make the sample flow in the mouth (the easier the sample flows in the mouth, the thinner the sample)	Low: water High: Greek yoghurt
Creaminess	The degree to which the sample provides a silky, smooth, velvety, rich, full mouthfeel (which is often associated with the presence of fat)	Low: water High: (un)whipped cream

4.2.7. Data analysis

Results from instrumental analyses were reported as mean values with standard deviation. One-way ANOVA and Tukey post-hoc analyses were performed to determine significant differences in rheological and tribological properties between samples prepared from the same matrix (milk or soup). Results from sensory evaluation of thickness and creaminess intensity were reported as mean values with standard error. Thickness and creaminess intensities (scale 0-100) were analysed using Repeated Measures ANOVA, with sample as fixed factor and participant as random factor. Bonferroni post-hoc analyses were performed to determine statistically significant differences between the samples. Pearson correlations between sensory thickness and creaminess were calculated for milks and soups separately. Pearson correlations between sensory and instrumental data were computed using the means of the pooled datasets (n = 11). Data analysis was performed using RStudio (version 4.0.2) using the packages lmerTest (Kuznetsova *et al.*, 2017), emmeans (Lenth, 2021b) and Hmisc (Harrell & Dupont, 2018). A significance level of $\alpha = 0.05$ was used.

4.3. Results & Discussion

4.3.1. Morphology of spray-dried MFC powders differing in composition before and after resuspension

Scanning Electron Microscopy images of the two types of spray-dried MFC powders revealed that both powders have a polydisperse particle size distribution (Figure 4.1a-b). Both powders contain smaller and larger particles, varying in size between ~1-30 μm. In the current study, a surplus of maltodextrin was added to MFC dispersions to improve the redispersibility of spray-dried MFC. The presence of polymers such as maltodextrin on the cellulose microfibrils prevents the formation of agglomerates during drying (hornification) (Lowys *et al.*, 2001; Velásquez-Cock *et al.*, 2018).

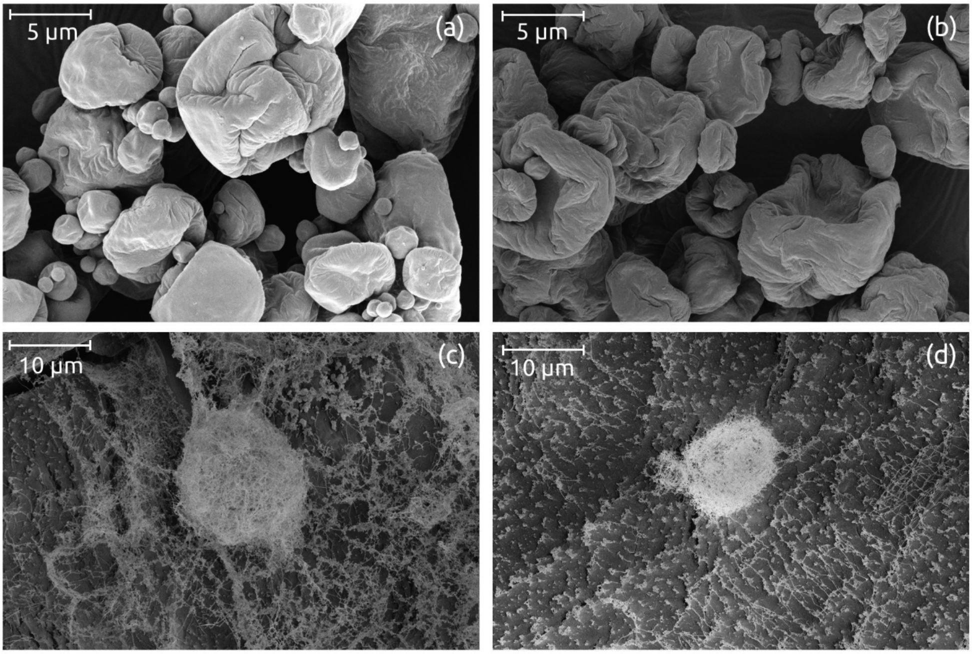

Figure 4.1. *Scanning Electron Microscopy (SEM) images of (a) MFC:MD-Low and (b) MFC:MD-High spray-dried MFC particles at 10,000x magnification. Panel (c) and (d) show cryoSEM images at 5,000x magnification of MFC:MD-Low after suspension in water.*

Figure 4.1c and 4.1d show the morphology of spray-dried MFC powders that have been resuspended in water. The powder particles swell and adopt a more spherical shape upon suspension in water. Three-dimensional spherical networks of entangled MFC are observed with particle sizes of approximately 10 µm. These MFC particles stay intact after suspension in water and are surrounded by individual microfibrils and small networks thereof. Two possible explanations for the presence of individual microfibrils and aggregates thereof are: (i) not all MFC contributes to the formation of spherical particles, or (ii) some of the spherical MFC networks are not sufficiently strong to maintain their shape upon suspension and therefore collapse or disintegrate. It is expected that maltodextrin is present in the voids within the MFC network and that suspension of the powders results in dissolution of maltodextrin in the continuous aqueous phase (Figure 4.2a-b). The spherical particles formed by the MFC network can potentially modify the texture of the liquid matrix in which the particles are suspended. We speculate that the lower maltodextrin content in MFC:MD-High enhances entanglement between the microfibrils during the spray-drying process. A larger number of contact points due to increased entanglement might strengthen the network, and these particles therefore might be denser and more rigid compared to MFC:MD-Low particles. We hypothesise that at constant MFC content within a food, fewer (*i.e.* lower volume fraction) but denser particles might be formed at high MFC:MD ratio compared to a lower ratio (Figure 4.2c-d).

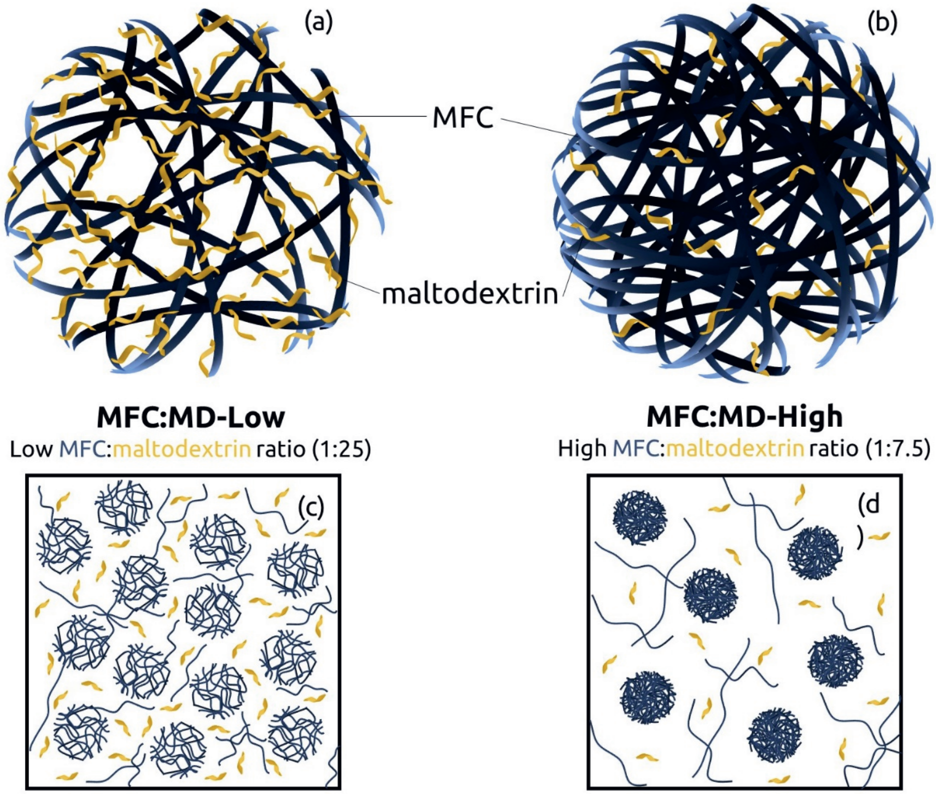

Figure 4.2. *Schematic overview of the proposed differences in morphology between spray-dried powders with (a,c) low MFC:maltodextrin ratio (1:25, MFC:MD-Low) and (b,d) high MFC:maltodextrin ratio (1:7.5, MFC:MD-High). The MFC network is displayed in dark blue, maltodextrin in yellow. Figure a and b represent spray-dried MFC powder before suspension in water, higher MFC:maltodextrin ratio results in particles with more entanglements between cellulose microfibrils. Figure c and d represent MFC particles after suspension in water, and spherical MFC networks are surrounded by individual microfibrils and aggregates thereof. It is hypothesised that at constant MFC content, fewer and denser particles might be formed at higher MFC:maltodextrin ratio.*

4.3.2. Rheological properties

Viscosity

All samples (soups and milks) displayed shear-thinning behaviour with the exception of skimmed milk and soup without MFC particles (M-0, S-0; Figure 4.3) which displayed near-Newtonian behaviour. An increase in the concentration of MFC:MD-Low resulted in an increase in shear viscosity and consistency index *K*, and a lower flow index *n* (Figure 4.3, Table 4.4). Although the final MFC (0.3 wt%) and maltodextrin content (7.5 wt%) of samples with 2.6 wt% MFC:MD-High were identical to those with 8% MFC:MD-Low (M-L8 and S-L8), flow curves of M-H2.6 and S-H2.6 overlapped with flow curves of M-L4 and S-L4. This suggests that MFC:MD-High powder is less effective at enhancing viscosity of liquid foods, and is therefore a less effective thickener than MFC:MD-Low. We speculate that this difference in rheological properties between high and low MFC:MD ratio powders might be caused by density differences of the respective MFC networks of these powders and consequently the number of spherical MFC particles that can be formed with the same amount of MFC (Figure 4.2). Spray-dried MFC powder with high MFC:maltodextrin ratio contains relatively more MFC and therefore a lower proportion of maltodextrin. As maltodextrin impairs agglomeration of MFC, more junction zones might be formed between cellulose microfibrils at higher MFC:maltodextrin ratio and a stronger and denser MFC network might be formed. At constant MFC content fewer of such denser particles might be acquired compared to lower MFC:maltodextrin ratios, resulting in lower viscosity.

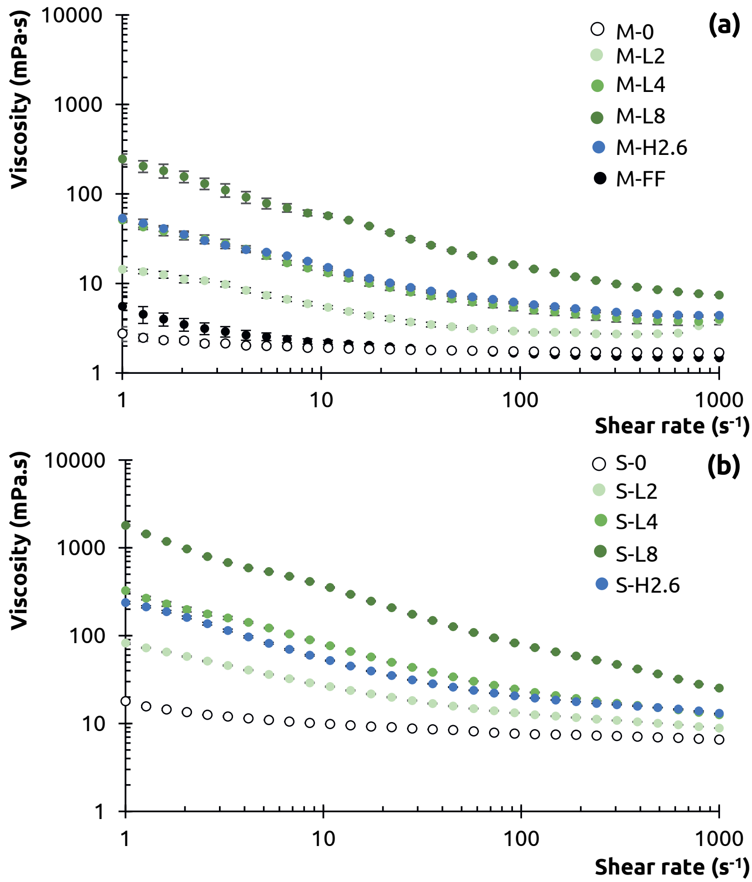

Figure 4.3. *Flow curves of (a) milks and (b) soups to which two types of spray-dried MFC were added at various concentrations. Sample codes and composition are summarised in Table 4.2. Means are presented with error bars representing standard deviations (triplicates).*

Viscoelastic properties

Storage and loss moduli (*G′* and *G″*) of skimmed milks and tomato soups increased with increasing concentration of spray-dried MFC particles (Table 4.4). At spray-dried MFC powder concentrations below 4% (M-0, M-L2, S-0, S-L2), corresponding to MFC concentrations below 0.15 wt%, milks and soups displayed liquid-like behaviour with loss modulus *G″* being larger than storage modulus *G′*. At higher concentrations gel-like behaviour was observed, as storage modulus *G′* was larger than loss modulus *G″* in the linear viscoelastic regime. Storage and loss moduli were higher for soups than for milks, and it should be emphasised that M-L4 and M-H2.6 exhibited only weak viscoelastic behaviour. A clear increase in storage moduli *G′* and *G″* was observed upon increasing the concentration of MFC:MD-Low. This was expected, as a higher volume fraction of spray-dried MFC particles results in increased flow resistance and enhanced viscoelastic properties. Storage and loss moduli were strongly positively correlated with shear viscosity parameters (η at 10, 50 and 100 s^{-1}, consistency index *K*; Table 4.5). These parameters were in turn negatively correlated with flow index *n*, which means that samples with higher viscosity, consistency index *K* and dynamic moduli (*G′*, *G″*) were more shear-thinning (*i.e.* exhibited less Newtonian behaviour).

Table 4.4. *Rheological properties of milks and soups to which two types of spray-dried MFC were added at various concentrations (mean ± standard deviation). Flow index n and consistency index K were determined by fitting the Ostwald-de Waele power law model to the experimental data. For each liquid matrix (milk/soup), samples sharing the same letter within a column are not significantly different from each other (p>0.05). Sample codes and composition are summarised in Table 4.2.*

	Sample	Flow index *n*	Consistency index *K* (Pa·s^n)	Storage modulus *G′* (Pa) (at 1% strain)	Loss modulus *G″* (Pa) (at 1% strain)
Milk	M-0	0.95 ± 0.01^{a}	0.002 ± 0.000^{c}	0.01 ± 0.01^{c}	0.04 ± 0.03^{b}
	M-L2	0.76 ± 0.01^{c}	0.011 ± 0.001^{bc}	0.10 ± 0.02^{c}	0.06 ± 0.05^{b}
	M-L4	0.60 ± 0.01^{d}	0.039 ± 0.003^{b}	0.41 ± 0.10^{bc}	0.14 ± 0.07^{b}
	M-L8	0.48 ± 0.03^{e}	0.202 ± 0.027^{a}	3.77 ± 0.12^{a}	0.57 ± 0.02^{a}
	M-H2.6	0.63 ± 0.02^{d}	0.041 ± 0.002^{b}	0.62 ± 0.18^{b}	0.16 ± 0.05^{b}
	M-FF	0.84 ± 0.03^{b}	0.004 ± 0.001^{c}	0.18 ± 0.30^{c}	0.10 ± 0.14^{b}
Soup	S-0	0.88 ± 0.00^{a}	0.014 ± 0.000^{e}	0.02 ± 0.03^{d}	0.11 ± 0.02^{c}
	S-L2	0.68 ± 0.00^{b}	0.064 ± 0.001^{d}	0.27 ± 0.12^{d}	0.23 ± 0.02^{c}
	S-L4	0.52 ± 0.01^{d}	0.259 ± 0.010^{b}	1.90 ± 0.10^{b}	0.63 ± 0.04^{b}
	S-L8	0.39 ± 0.00^{e}	1.475 ± 0.007^{a}	21.67 ± 0.58^{a}	3.40 ± 0.12^{a}
	S-H2.6	0.57 ± 0.01^{c}	0.173 ± 0.006^{c}	1.03 ± 0.06^{c}	0.48 ± 0.02^{b}

Table 4.5. *Pearson correlations between sensory and instrumental parameters. Data from milks and soups were pooled (n = 11). Asterisks indicate statistically significant differences: (*) $p < 0.05$; (**) $p < 0.01$; (***) $p < 0.001$, n.s. = not significant.*

	Thick-ness	Creami-ness	η_{10s}^{-1}	η_{50s}^{-1}	η_{100s}^{-1}	Flow index *n*	Consis-tency index *K*	Storage modulus *G'*	Loss modulus *G''*	μ_{max}	Start of mixed regime	Slope mixed regime
Thickness	-	0.974^{***}	0.710^{*}	0.711^{*}	0.708^{*}	-0.913^{***}	0.701^{*}	0.707^{*}	0.707^{*}	n.s.	n.s.	n.s.
Creaminess		-	0.736^{**}	0.732^{*}	0.725^{*}	-0.901^{***}	0.729^{*}	0.730^{*}	0.732^{*}	n.s.	n.s.	n.s.
η_{10s}^{-1}			-	0.997^{***}	0.992^{***}	-0.669^{*}	0.999^{***}	0.988^{***}	0.999^{***}	n.s.	n.s.	n.s.
η_{50s}^{-1}				-	0.999^{***}	-0.691^{*}	0.993^{***}	0.975^{***}	0.994^{***}	n.s.	n.s.	n.s.
η_{100s}^{-1}					-	-0.701^{*}	0.986^{***}	0.964^{***}	0.987^{***}	n.s.	n.s.	n.s.
Flow index *n*						-	-0.649^{*}	-0.621^{*}	-0.656^{*}	n.s.	n.s.	n.s.
Consistency index *K*							-	0.993^{***}	1.000^{***}	n.s.	n.s.	n.s.
Storage modulus *G'*								-	0.993^{***}	n.s.	n.s.	n.s.
Loss modulus *G''*									-	n.s.	n.s.	n.s.
μ_{max}										-	n.s.	n.s.
Start of mixed regime											-	0.854^{***}
Slope mixed regime												-

4.3.3. Tribological properties

Tribological properties of milks and soups are displayed in Figure 4.4 and Table 4.6. Samples to which varying concentrations of spray-dried MFC were added displayed comparable friction properties, as their friction curves largely overlap. The fact that no large differences in friction coefficients were observed between milk and soups differing in concentration of added spray-dried MFC and those without MFC suggests that the spherical MFC particles were not entrained between the PDMS pins and the glass ball. We hypothesise that the high shear rates arising in the tribological set-up disrupt the spherical network formed by MFC. Individual microfibrils and aggregates thereof are released into the continuous phase, where these can form volume-spanning networks similar to those never-dried MFC dispersions. Previously, such MFC networks have been postulated to be excluded from the contact zone due to their large hydrodynamic volume (Blok *et al.*, 2021). Exclusion of the particles implies that tribological properties of the continuous phase of the samples were determined instead, which consists of skimmed milk or soup with varying amounts of maltodextrin. Friction properties in milk samples may therefore be determined by the proteins present in the milk (Chojnicka-Paszun *et al.*, 2012), while friction of soups is possibly dominated by the presence of other biopolymers (*e.g.* starch and guar gum) or the small amount of fat (plate-out effect, Upadhyay *et al.* (2020)). Friction properties of full-fat milk on the other hand are significantly lower than those of the other milk samples over the entire speed range. This most probably results from the higher fat content of full-fat milk, as fat can form a lubricating film on the tribological surfaces. Although the increase in friction coefficient from 10 mm/s onwards appears to be the start of the hydrodynamic regime, this is refuted by the decline in friction coefficient starting around 400 mm/s. It is hypothesised that the lubricating fat film is disrupted at high sliding speeds, allowing the entrainment of proteins which in turn provide poorer lubrication (Chojnicka-Paszun *et al.*, 2012). The slope of the mixed friction regime shows a strong positive correlation with the sliding speed at which the mixed regime starts (r = 0.854, p < 0.001; Table 4.5). Samples for which the mixed regime started at higher sliding speeds had a less steep negative slope in the mixed regime.

Table 4.6. *Tribological properties of milks and soups to which two types of spray-dried MFC were added at different concentrations. μ_{max} represents the maximum friction coefficient measured over the entire speed range. For soups, the start of the mixed regime was defined as the speed at which μ_{max} occurred, while this parameter was determined manually for milks. The slope in the mixed regime was defined as the slope between the start of the mixed regime and the friction at 1000 mm/s. Values represent mean values ± standard deviation of triplicate measurements. For each matrix (milk/soup), samples sharing the same letter within a column are not significantly different from each other (p>0.05). Sample codes and composition are summarised in Table 4.2.*

	Sample	Maximum friction μ_{max}	Sliding speed representing start of mixed regime (mm/s)	Slope in mixed regime ($\cdot 10^{-4}$ s/mm)
Milk	M-0	0.46 ± 0.03^{ab}	12.6 ± 0.0^{b}	-2.67 ± 0.06^{b}
	M-L2	0.51 ± 0.03^{a}	13.4 ± 1.9^{b}	-2.75 ± 0.37^{b}
	M-L4	0.48 ± 0.03^{a}	19.4 ± 0.6^{b}	-2.51 ± 0.28^{b}
	M-L8	0.47 ± 0.03^{ab}	23.4 ± 0.0^{b}	-2.39 ± 0.22^{b}
	M-H2.6	0.46 ± 0.03^{ab}	18.3 ± 1.2^{b}	-2.63 ± 0.25^{b}
	M-FF	0.39 ± 0.04^{b}	469.2 ± 26.5^{a}	-1.12 ± 0.15^{a}
Soup	S-0	0.35 ± 0.01^{b}	37.0 ± 4.2^{a}	-3.14 ± 0.19^{a}
	S-L2	0.36 ± 0.01^{b}	27.6 ± 5.4^{a}	-3.25 ± 0.15^{a}
	S-L4	0.36 ± 0.02^{b}	21.1 ± 6.1^{a}	-3.05 ± 0.26^{a}
	S-L8	0.41 ± 0.01^{a}	2.0 ± 0.2^{b}	-3.42 ± 0.10^{a}
	S-H2.6	0.35 ± 0.01^{b}	20.7 ± 3.6^{a}	-2.91 ± 0.08^{a}

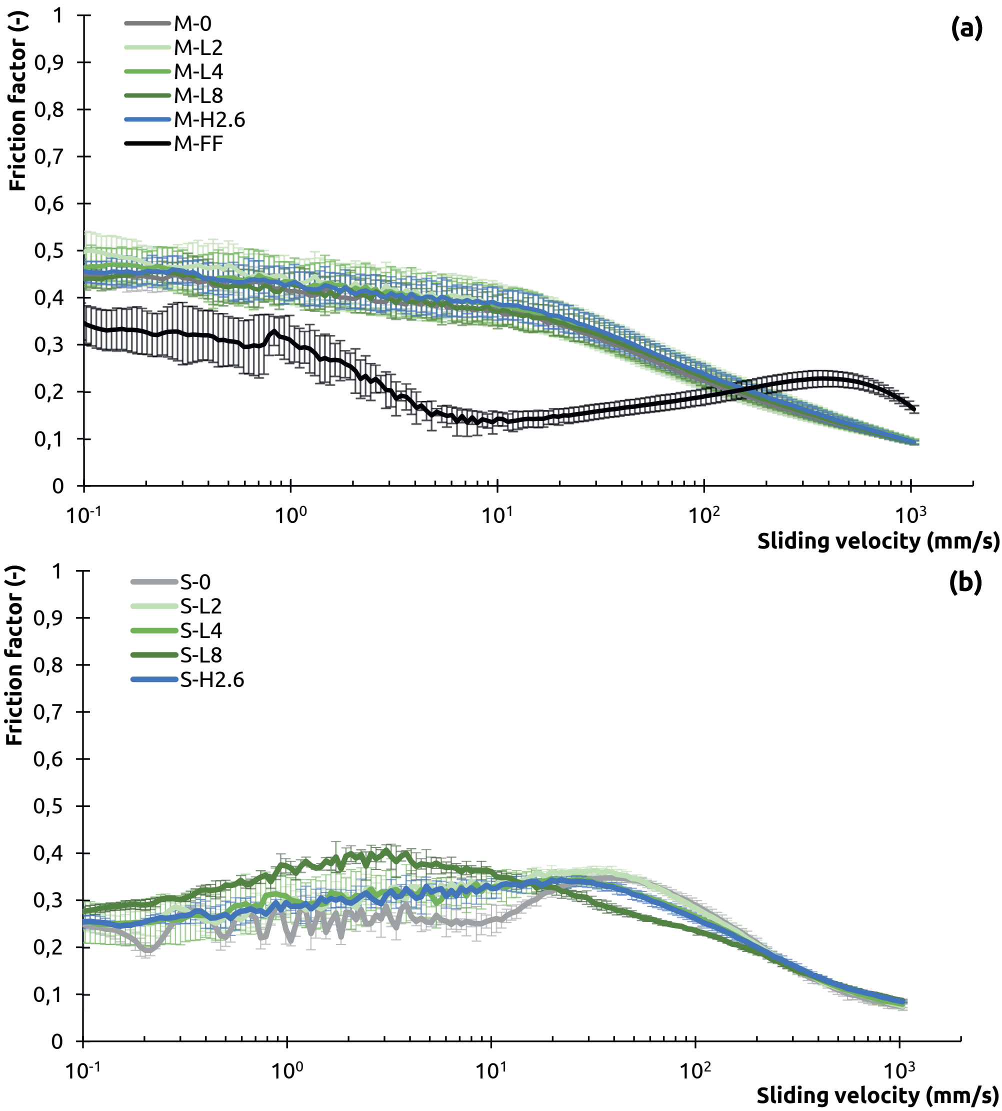

Figure 4.4. *Tribological properties of (a) milks and (b) soups to which two types of spray-dried MFC were added at different concentrations. Means are displayed with error bars representing standard deviations (triplicates). Sample codes and composition are summarised in Table 4.2.*

<ins>4.3.4. Sensory properties</ins>

A linear increase in thickness intensity was observed upon increasing the concentration of MFC:MD-Low in milks and soups (Figure 4.5, 4.6). The only exceptions were M-L2 and S-L2, which were not significantly thicker than milk or soup without MFC particles (M-0 and S-0). Moreover, thickness of commercial full-fat milk (M-FF) was not significantly different from skimmed milk without MFC particles to which 7.5 wt% maltodextrin was added (M-0). Similar to thickness, creaminess intensity of milks and soups increased linearly with increasing concentration of MFC:MD-Low (Figure 4.5, 4.6). While the presence of 2% spray-dried MFC:MD-Low did not affect thickness of soups and milks, creaminess of soup was significantly enhanced by addition of 2% MFC:MD-Low (corresponding to 0.075 wt% MFC) compared to soups without added spray-dried MFC particles. This effect on creaminess appears to be matrix-dependent, as it was not observed for milk. Milk presumably possesses a certain degree of intrinsic creaminess, which was not surpassed by addition of a small amount of MFC particles. No significant differences in creaminess were observed between M-0, M-L2 and M-FF, which was not expected. Full-fat milk contains more fat than the milk samples prepared from skimmed milk, and full-fat milk was therefore expected to have a creamier mouthfeel. It should be recalled that M-0 contained 7.5 wt% maltodextrin and had similar viscosity to M-FF (Figure 4.3a), which might explain why M-FF was not perceived as thicker nor creamier than M-0. Milks and soups to which the highest concentration of MFC particles were added (M-L8 and S-L8) were significantly thicker and creamier than all other samples. Remarkably, M-H2.6 and S-H2.6 displayed thickness and creaminess intensities comparable to those of M-L4 and S-L4, while MFC and maltodextrin content of these samples were matched to those of M-L8 and S-L8. The analogy between these samples is in line with results on the samples' rheological properties (Figure 4.3, Table 4.4). The similarity between M-H2.6, S-H2.6, M-H4 and S-H4 is hypothesised to be caused by differences in the (number of) particles formed upon spray-drying, as illustrated in Figure 4.2.

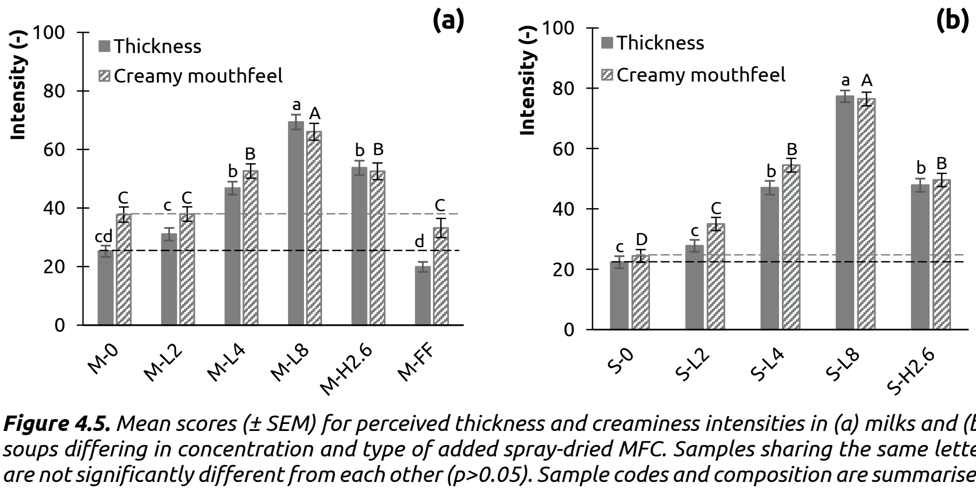

Figure 4.5. *Mean scores (± SEM) for perceived thickness and creaminess intensities in (a) milks and (b) soups differing in concentration and type of added spray-dried MFC. Samples sharing the same letter are not significantly different from each other (p>0.05). Sample codes and composition are summarised in Table 4.2.*

The increase in thickness intensity upon increasing the concentration of MFC:MD-Low is fairly comparable for milk and soup, while the slopes for creaminess show modest differences between the two liquid matrices (Figure 4.6). For soup a stronger increase in creaminess was achieved upon addition of MFC particles compared to milk. Initial creaminess for soup without MFC particles (S-0) started at lower creaminess intensity but ended at an intensity higher than that of milk at high MFC particle concentration (S-L8). This implies that the skimmed milk used in the current study was generally perceived as creamier than the tomato soup before addition of MFC particles, but soups became creamier more rapidly once MFC particles were added. Addition of MFC particles thus results in a more efficient enhancement of creaminess in soups compared to milks, which is reflected in a slope that was 76% larger for soup compared to milk (6.7 vs 3.8; Figure 4.6b). This suggests that an interaction effect exists between creaminess and the matrix in which the particles are suspended. As thickness and creaminess intensities increase concurrently (Figure 4.5 and 4.6) the results moreover suggest that perceived creaminess is largely determined by the perceived thickness of the samples, which is in accordance with literature (e.g. Blok *et al.*, 2020; Janhøj *et al.*, 2008; Kokini & Cussler, 1983). The significant positive correlations found between sensory thickness and creaminess intensities substantiate this postulation: $r = 0.53$ ($p < 0.001$) for milks, $r = 0.64$ ($p < 0.001$) for soups. Pooling the two datasets reveals an exceptionally strong correlation between thickness and creaminess intensities ($r = 0.974$, $p < 0.001$; Table 4.6), which is a result of these correlations being based on the mean values ($n = 11$) instead of the raw sensory data ($n = 792$). The results demonstrate that spray-dried MFC particles enhance sensory thickness and creaminess of liquid foods and can thus be used to modify texture of such foods, for instance as thickener or creamer.

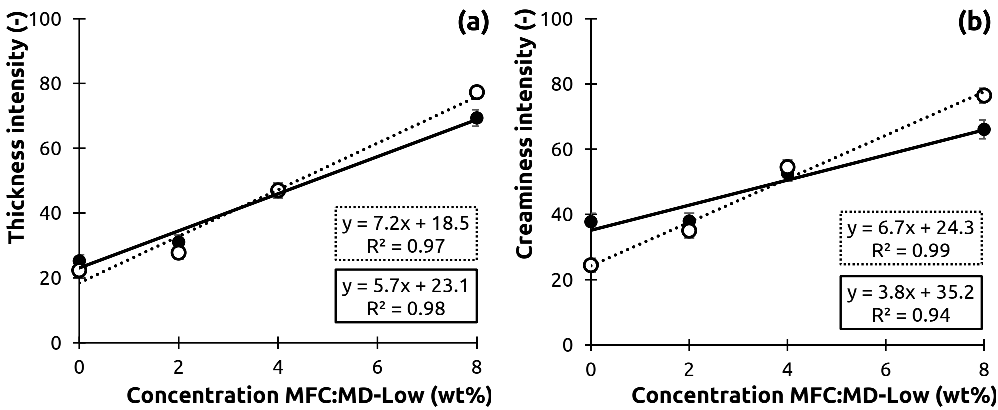

Figure 4.6. *Intensity of (a) thickness and (b) creaminess as a function of concentration of MFC:MD-Low particles (mean ± SEM) added to milk (filled symbols) and soup (open symbols). Linear trendlines are plotted to guide the eye.*

Strong positive correlations were found between the sensory texture attributes and the majority of rheological parameters (r = 0.701-0.974; Table 4.6), which demonstrates that rheological properties were important drivers of sensory perception of thickness and creaminess. On the other hand no statistically significant correlations were observed between the sensory texture attributes and tribological properties of the samples. This can be attributed to the fact that friction properties could not be determined as intended due to exclusion of MFC particles from the tribological gap. It can, however, not be excluded that friction in the mouth may be different from that in the tribological set-up used in this study. In the oral cavity, MFC particles might be present between the tongue and the palate where these could contribute to creaminess through lubrication.

4.4. Conclusions

The present work examined the effect of concentration and MFC:maltodextrin ratio of spray-dried MFC powders on rheological, tribological and sensory texture properties of liquid foods. Spray-dried MFC powders were prepared in a surplus of maltodextrin at two ratios and applied at different concentrations in skimmed milk and instant tomato soup. An increase in the concentration of spray-dried MFC resulted in higher viscosity, consistency index K, storage and loss modulus (G', G'') and a lower flow index n. At concentrations above 4 wt% (corresponding to 0.075 wt% MFC), weak gel-like behaviour was observed in milks and soups. Increasing the concentration of spray-dried MFC particles substantially increased perceived thickness and creaminess of milks and soups. Creaminess was positively correlated with perceived thickness, and both sensory attributes were strongly correlated with rheological parameters.

SEM images demonstrated that both powders yielded a polydisperse particle size distribution. Three-dimensional spherical MFC networks were formed that did not disintegrate upon suspension in water, while maltodextrin dissolved in the continuous phase. High MFC:maltodextrin ratio particles were less effective at enhancing rheological and sensory texture properties of liquid foods compared to particles with low MFC:maltodextrin ratio, probably due to the lower volume fraction of particles that can be obtained with the same amount of MFC. No clear lubrication effect of the dried MFC particles could be established, since the particles were presumably not entrained between the tribological surfaces.

Spray-drying of MFC in the presence of maltodextrin prevents hornification and improves transportation and storage efficiency compared to MFC dispersions, which may provide financial benefits compared to liquid MFC dispersions. Based on the results presented, we conclude that spray-dried MFC particles can be used in liquid foods as thickener or even as a fat replacer due to their effect on rheological properties, sensory thickness and creaminess. One of the advantages of this material is its easy incorporation in foods by food manufacturers compared to the use of liquid MFC dispersions. The material furthermore shows potential as a thickener or creamer for direct use by consumers. In case spray-dried MFC particles are used to reduce the energy density of foods, for example in low-fat foods, replacing maltodextrin by less calorie dense materials should be considered.

Acknowledgements

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5

Contributions of viscosity and friction properties to oral and haptic texture perception of iced coffees

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Abstract

Creaminess is affected by bulk properties (*i.e.* viscosity) and surfaces properties (*i.e.* friction). This study aimed (i) to assess contributions of viscosity and friction properties to creaminess, thickness and slipperiness perception; and (ii) to compare oral and haptic thickness and slipperiness perception of iced coffees. Three iced coffees differing in viscosity and friction properties were prepared: low viscosity – high friction (LV-HF); low viscosity – low friction (LV-LF) and high viscosity – low friction (HV-LF) iced coffee. Viscosity of iced coffees was adjusted by addition of maltodextrin, and viscosity of HV-LF was 2.5 times higher than that of LV-HF and LV-LF (10 vs 4 mPa·s at 100 s^{-1}). Friction coefficients of LV-LF were reduced by addition of polyethylene glycol (PEG, M_w 6,000), and were up to 25% lower than those of LV-HF. Forty-seven untrained panellists (18-27 y) performed two-alternative forced choice (2-AFC) and rank-rating tests to compare creaminess by oral assessment, and thickness and slipperiness by oral and haptic assessment. Results from 2-AFC and rank-rating congruently showed that HV-LF was creamier, thicker and more slippery than LV-HF and LV-LF, both orally and haptically. LV-LF was orally perceived as less creamy and less thick, but haptically as more slippery than LV-HF. Creaminess was stronger correlated to thickness than to slipperiness. Oral and haptic evaluation of thickness were congruent, whereas differences between oral and haptic slipperiness evaluation were product-dependent. We conclude that increasing viscosity enhances creaminess, whereas increasing lubrication is not necessarily sufficient to increase creaminess in iced coffees.

5.1. Introduction

Creaminess is a desired mouthfeel property in foods (Ares *et al.*, 2010; Richardson-Harman *et al.*, 2000) and can generally be enhanced by increasing fat content (Akhtar *et al.*, 2006; Kilcast & Clegg, 2002; Sala *et al.*, 2007). However, nowadays the food industry aims to reduce fat content of foods while maintaining desired mouthfeel properties. More specifically, retaining creaminess in low-fat foods poses a challenge. Several ingredients have been suggested to mimic fat in terms of mouthfeel perception, such as modified starches (Chen *et al.*, 2020), inulin (Guggisberg *et al.*, 2009; Meyer *et al.*, 2011), microbubbles (Rovers *et al.*, 2016) and microparticulated whey protein (Liu, Stieger, *et al.*, 2016; Liu, Tian, *et al.*, 2016). Nonetheless, no substitute has yet been found that can provide creaminess similar to that of fat (Upadhyay *et al.*, 2020). A better understanding of the mechanisms underlying creaminess perception and the physical-chemical food properties contributing to creaminess may help to develop fat-reduced foods with desirable sensory properties.

Increasing the complexity of tongue movements during oral processing enhances creaminess perception of semi-solids (de Wijk *et al.*, 2003). The fact that a combination of tongue movements is required for optimal creaminess perception suggests that creaminess of foods is affected by more than one single food property. During oral processing, a shift from the rheological domain to the tribological domain occurs (Chen & Stokes, 2012; Stokes *et al.*, 2013). Bulk rheological properties such as viscosity dominate the beginning of oral processing, and are related to thickness perception (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; Cutler *et al.*, 1983; Richardson *et al.*, 1989). Tribological properties are important during subsequent stages of oral processing, when the film between tongue and palate becomes thinner. Tribology is the study of the wear, friction and lubrication of interacting surfaces in relative motion (Stokes *et al.*, 2013). As foods are subjected to friction due to squeezing and pressing between tongue and palate, tribology provides a tool to elucidate sensory perception of mouthfeel attributes by linking friction to sensory properties. Tribological properties have, for instance, been linked to slipperiness perception of guar gum solutions (Malone *et al.*, 2003a) and particle dispersions (Chojnicka-Paszun *et al.*, 2014). In summary, both rheological and tribological food properties contribute to creaminess (Kokini, 1987; Kokini & Cussler, 1983; Upadhyay *et al.*, 2020).

Increasing the viscosity of a liquid food can enhance its creaminess, as more viscous foods are generally perceived to be creamier (Bom Frøst & Janhøj, 2007). One of the earliest works on creaminess perception was performed by Wood (1974), who concluded that soups require a viscosity of at least 50 mPa·s in order to be perceived as creamy. Another study on model soups confirmed that creaminess increased when the viscosity of soups increased (Daget & Joerg, 1991). In two studies on o/w emulsions, Akhtar and colleagues (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005) observed that increased viscosity by addition of hydrocolloids resulted in enhanced creaminess. Similarly, van Aken *et al.* (2011) increased o/w emulsion viscosity by addition of gum arabic, which resulted in higher creaminess ratings according to a trained panel. Furthermore, Janhøj *et al.* (2008) found that creaminess of acidified milk drinks increased with increasing viscosity.

In addition to viscosity, friction properties are increasingly recognised to contribute to creaminess of liquid foods (de Wijk *et al.*, 2006; Dickinson, 2018; Kokini & Cussler, 1983; Upadhyay *et al.*, 2020). The majority of studies that investigated the effect of friction on texture perception modified friction properties by altering either fat or protein content. For instance, reduced friction due to increased fat or protein content resulted in higher creaminess in milks (Chojnicka-Paszun *et al.*, 2012; Meyer *et al.*, 2011), custards (de Wijk & Prinz, 2005) and yoghurts (Sonne *et al.*, 2014). Moreover, a negative correlation between friction and perceived smoothness was found in o/w emulsions with varying oil content (Upadhyay & Chen, 2019). Laguna *et al.* (2017) demonstrated that discrimination between fat-free and full-fat dairy products was mainly based on differences in tribological properties. By varying the fat or protein content, these studies did not only alter friction properties, but rheological properties of the foods as well. Increasing the fat or protein content reduced friction, but at the same time increased viscosity, two parameters that both have a positive effect on creaminess. Hence, strictly speaking, from these studies one cannot assess the individual, relative contributions of viscosity and friction properties to perception of texture attributes such as creaminess.

Texture can be evaluated by different approaches, such as visually, haptically or orally. Shama and Sherman (1973) compared oral and non-oral methods for viscosity evaluation and concluded that stimuli used for such evaluations depend on the evaluation method employed. Correspondingly, liquids were perceived as thicker when evaluated by stirring with a spatula, compared to stirring with the index finger (BergmannTiest *et al.*, 2012). Christensen and Casper (1987) found that solutions thickened by sodium alginate were perceived to be thicker when assessed orally compared to visual or haptic evaluation. In a study on o/w emulsions, oral and haptic smoothness scores were generally similar, but emulsions were better discriminated by oral smoothness (Upadhyay & Chen, 2019). Differences between oral and haptic texture perception might be caused by differences in the mechanoreceptors involved in sensing texture. Mechanoreceptors in the human skin and oral cavity comprise slowly adapting (SA) and rapidly adapting (RA) receptors (Engelen & de

Wijk, 2012; Trulsson & Essick, 1997). The majority of mechanoreceptors on the tongue and finger tips consists of RA receptors (Johansson & Vallbo, 1979; Trulsson & Essick, 1997), whereas SA receptors are more predominant in other parts of the human body (Edin & Abbs, 1991; Johansson *et al.*, 1988; Nordin & Hagbarth, 1989). Although SA and RA receptors have been found in similar proportions and function similarly in the oral cavity and the finger tips, mechanoreceptors on the tongue have been found to be more sensitive to force (Rath & Essick, 1990; Trulsson & Essick, 1997).

While it is well known that rheological and tribological properties influence texture perception of foods, less is known about the relative contributions of rheological and tribological properties to texture perception of attributes such as creaminess. Therefore, the objective of this study was to better understand the effects of viscosity and friction properties on oral and haptic texture perception by varying viscosity of iced coffees with minimal changes in friction properties and *vice versa*. Viscosities of iced coffees were varied by adding maltodextrin, whereas friction properties were adjusted by adding polyethylene glycol (PEG), a food grade polymer (E1521) that can be used as a lubricant in foods without considerably changing viscosity (Matlock *et al.*, 1999). Especially high molecular weight PEG (M_w > 1,000,000 g/mol) can reduce friction in aqueous solutions considerably without strongly affecting viscosity. Legislation limits the use of PEG in foods to low molecular weight PEG (M_w < 10,000 g/mol). The goal of this study was (i) to assess contributions of viscosity and friction properties to creaminess, thickness and slipperiness perception of iced coffees; and (ii) to compare oral and haptic thickness and slipperiness perception of these beverages.

5.2. Materials & Methods

5.2.1. Product preparation

Three iced coffees differing in viscosity and friction properties were prepared: low viscosity – high friction (LV-HF); low viscosity – low friction (LV-LF); and high viscosity – low friction (HV-LF) iced coffee (Table 5.2). Products were prepared from commercially available ready-to-drink milk-based iced coffee (20% coffee; 0.9% fat (w/v); Koffiecino, Holland Foodz, the Netherlands). Viscosity of HV-LF was adjusted by adding 30 wt% maltodextrin (Fantomalt, Nutricia, Danone, France), and friction properties of LV-LF were adjusted by adding 7 wt% polyethylene glycol (PEG (E-1521); M_w 6,000, Merck, Germany). No PEG was added to HV-LF, as the increase in viscosity due to maltodextrin addition was accompanied by a decrease in friction properties. Since addition of PEG to LV-LF resulted in a small increase in viscosity, 16.8 wt% maltodextrin was added to LV-HF to obtain matching shear viscosities. Iced coffees were mixed with the ingredients for 30 min at room temperature to ensure dissolution of maltodextrin and PEG. Products were freshly prepared each day and stored in the refrigerator at 4°C until 1h before use.

5.2.2. Characterisation of viscosity and friction properties

Flow curves of the iced coffees were determined using an MCR 302 rheometer equipped with a double gap concentric cylinder geometry (DG26.7/Ti, Anton Paar, Austria). Flow curves were measured while increasing the shear rate from 0.1 to 1000 s^{-1} in 50 logarithmic steps in 10 min. Measurements were performed in triplicate at 35°C.

Friction properties of the three iced coffees were characterised using a ball-on-three-pins set-up in an MCR 302 rheometer (Anton Paar, Austria). A glass ball was used to represent the human palate, whereas cylindrical shaped polydimethylsiloxane (PDMS) pins were used to mimic the hardness and surface of the human tongue. One mL of sample was transferred to the sample holder. Each test consisted of three consecutive runs of 10 min in which sliding speeds were increased from 0.0001 – 2200 rpm (equivalent to $4 \cdot 10^{-5}$ – 10^{3} $mm \cdot s^{-1}$) using a logarithmic ramp, while a normal force of 1 N was applied. Friction coefficients were obtained as the ratio of the frictional force divided by the normal load. Runs were separated by 5 min breaks in which a normal force of 1 N was applied. Data from the second run was used for analysis. Tests were conducted in triplicate at 35°C to simulate the oral environment. Prior to measuring iced coffees, PDMS surfaces were run-in by (i) one run with 1 mL demineralised water and (ii) one run with 1 mL plain iced coffee.

For measures of friction properties of the iced coffees in the presence of saliva, unstimulated saliva was collected over 15 minutes from one healthy volunteer after a fasting period of 60 min based on the protocol of Silletti *et al.* (2007). After rinsing the mouth with water, saliva was collected with closed lips and expectorated in a plastic tube. Saliva was centrifuged at 956 *g* for 10 min to remove cellular debris and was subsequently kept on ice. Saliva was mixed with iced coffee (ratio 1:1) just before addition to the sample holder. The same experimental conditions were used as for the tribological measurements of the iced coffees without saliva. Data from the second run was used for analysis. Tests were conducted in triplicate at 35°C to simulate the oral environment. Prior to measuring the saliva-iced coffee mixtures, PDMS surfaces were run-in by (i) one run with 1 mL demineralised water and (ii) three runs with 1 mL saliva.

5.2.3. Participants

Dutch-speaking participants were recruited from the surroundings of Wageningen using flyers and social media. Individuals were eligible for participation in the study when they were between 18-35 years and had a BMI between 18.5-25 kg/m^2. Individuals were excluded from participation when they had problems with general or oral health, when they did not have normal smell and taste function or when they had mastication or swallowing disorders. Individuals with food allergies or intolerances for milk products and pregnant or breastfeeding women were excluded from participation. A total of 50 subjects agreed on participation in the study, of which *n* = 47 (8 male, 39 female) completed all test sessions. Mean age was 21.5 (±2.2) years and mean BMI was 21.7 (±1.7) kg/m^2. Most participants were frequent coffee drinkers, as 47% consumed coffee daily and 31% consumed coffee multiple times a week. Participants signed an informed consent and completed a general questionnaire in the first test session. Participants received financial reimbursement after completion of the three test sessions.

5.2.4. Sensory evaluation

Participants (*n* = 47) completed three test sessions on separate days. In each test session, two sensory methods were used: the 2-alternative forced choice method (2-AFC) followed by rank-rating, both of which are described in more detail below. A test session lasted approximately 30 min. During each test session only one of the following sensory texture attributes was evaluated: thickness, slipperiness or creaminess. The order in which participants assessed each of the three attributes was randomised. In test sessions in which participants evaluated creaminess, the 2-AFC method and rank-rating were performed orally by tasting the iced coffees. In test sessions in which thickness and slipperiness of the iced coffees were evaluated, the two sensory methods were first performed orally by tasting, followed by performing the same two sensory methods by haptic evaluation in hand. Haptic assessment was performed by rubbing the iced coffee between the fingers. Creaminess was not evaluated haptically, as we assumed that a meaningful evaluation of creaminess by hand is not possible.

To become acquainted with the attributes to be evaluated, participants received two reference products that represented products low and high in the respective attribute. Subsequently, an example question was presented to familiarise participants with the sensory method and the products used in the study. For the example question, two of the actual products (LV-HF, LV-LF or HV-LF) were presented to the participant. Definitions of the attributes and instructions were provided to the participants (Table 5.1).

Table 5.1. *Definitions and instructions of attributes (translated into English from Dutch) and reference products.*

		Definition / instruction	Reference products	
			Low intensity	High intensity
Oral	Creaminess	"The degree to which you experience a silky, rich, full feeling in your mouth"	Skim milk	Full-fat milk
	Thickness	"How easily does the product flow in your mouth?"	Plain iced coffee	Iced coffee + 50% maltodextrin
	Slipperiness	"How easily does your tongue move over your palate when you consume the product?"	Water	Olive oil
Haptic	Thickness	"Evaluate the thickness of the product by rubbing a small amount between thumb and index finger"	Plain iced coffee	Iced coffee + 50% maltodextrin
	Slipperiness	"Evaluate the slipperiness of the product by rubbing a small amount between thumb and index finger"	Water	Aqueous PEG solution (M_w $4{\cdot}10^6$, 1.0 g/L)

Participants were seated in individual sensory booths with standard white light and were asked to refrain from drinking coffee two hours prior to the test session. Participants wore nose clips during the entire session, as this facilitated focusing on texture attributes only and limited the perception of off-flavours caused by addition of PEG to LV-LF. Iced coffees (15-20 mL) were presented in 30 mL transparent plastic cups labelled with random 3-digit numbers. Participants were asked to expectorate the product after evaluation, and to rinse their mouth after tasting each set of iced coffees. Crackers and water were provided for palate cleansing between the product pairs. For sessions in which attributes were assessed haptically, participants used tap water and tissues to clean their hands. Data was collected in Dutch using EyeQuestion® (Logic8, the Netherlands).

2-Alternative forced choice method (2-AFC)

In each test session, the three iced coffees were first evaluated by using the 2 alternative forced choice method (2-AFC). Each of the iced coffees was compare with both other products using three 2-AFC tests. All comparisons were performe in duplicate within one session. The order of pairs and of products within pairs wa randomised among participants. For each pair of iced coffees, participants wer asked to determine which of the two products was more intense in the attribute o interest (*i.e.* 'Which of the two products is creamier?'). Participants were allowed t re-taste products if desired and were forced to guess if no difference could b detected.

Rank-rating

In each test session, evaluation using the 2-AFC method was followed by a rank rating test. The rank-rating test was performed to obtain an estimate of th perceived differences between the three iced coffees. In the rank-rating test th three iced coffees were presented simultaneously. Participants placed the product on an unstructured 100 mm line scale that represented the attribute of interest anchored from "not at all' on the left to "extremely" on the right. The order o products within the set was randomised among participants, and participants wer asked to evaluate the products from left to right. Participants were allowed to re taste the products if desired.

<u>5.2.5. Data analysis</u>

For the 2-AFC method, percentages of products chosen to be more intense tha their counterpart were determined, and the Bradley-Terry model was applied to th multiple paired comparisons data (Turner & Firth, 2012). Results from rank-ratin were reported as mean values with standard error. A repeated measures ANOV with participants as random factor was performed on rank-rating scores of eac attribute to determine significant differences between the three iced coffees. Two way repeated measures ANOVAs were performed to test for significant difference between haptic and oral rank-rating scores of thickness and slipperiness. Pearso correlation coefficients were determined between the sensory attributes usin pooled rank-rating scores of the three iced coffees. Data was analysed using RStudi (version 3.5.2) and a significance level of $\alpha = 0.05$ was used.

5.3. Results

5.3.1. Viscosity and friction properties

Mean shear viscosities (± SD) of the three iced coffees at 35 °C are shown in Figure 5.1. Flow curves of LV-HF and LV-LF overlap, whereas the viscosity of HV-LF is approximately 2.5 times higher within this range of shear rates (Table 5.2). Mean friction coefficients (± SD) of the three iced coffees as a function of sliding speed are displayed in Figure 5.2 in absence (a) and presence (b) of saliva. In both conditions (with and without saliva), highest friction coefficients were observed for LV-HF for nearly the entire sliding speed range. Addition of PEG (LV-LF) led to a reduction in friction coefficient relative to LF-HF in presence and absence of saliva, without affecting viscosity (Figure 5.1). Addition of 30 wt% maltodextrin to iced coffee resulted in an increase in viscosity (Figure 5.1) and a decrease in friction coefficient (Figure 5.2, Table 5.2). In the presence of saliva, boundary friction was generally lower, and the mixed regime shifted towards higher sliding speeds (Figure 5.2b). Moreover, differences in friction between the three products became smaller in the presence of saliva.

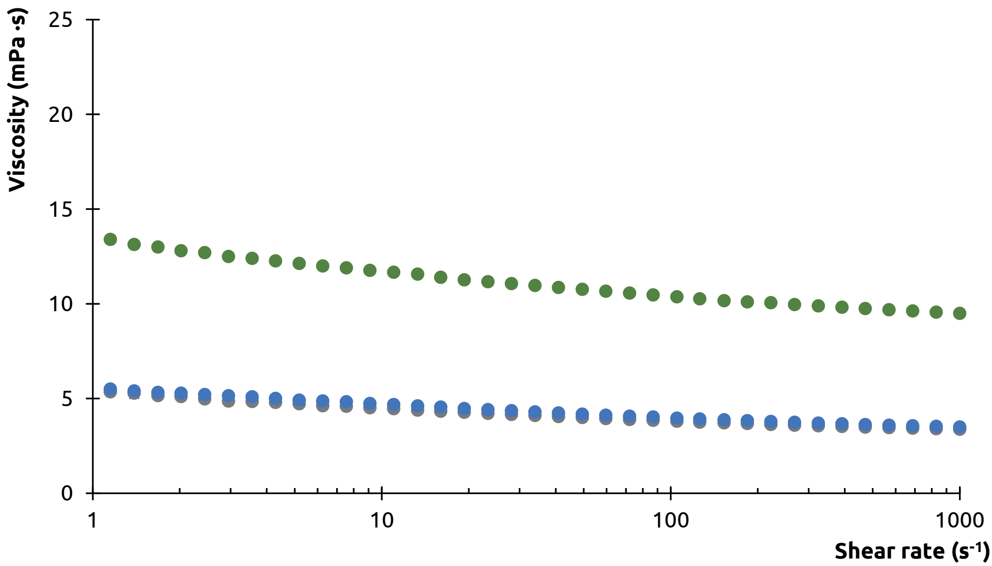

Figure 5.1. *Flow curves of the three iced coffees (35 °C). LV-HF = grey; LV-LF = blue; HV-LF = green. LV-HF denotes low viscosity – high friction, LV-LF low viscosity – low friction and HV-LF high viscosity – low friction iced coffee. Error bars represent standard deviation of triplicate measures.*

Table 5.2. *Overview of composition (wt% maltodextrin (MD) and wt% PEG), viscosity (mean η (± SD) at 1, 10 and 100 s^{-1} at 35°C) and friction properties (mean friction coefficient μ (± SD) in absence of saliva at 35°C) of the three iced coffees. Friction coefficients at 1 mm·s^{-1} (boundary regime), 10 mm·s^{-1} and 100 mm·s^{-1} (mixed regime), and exponent b (slope of curve in the mixed regime between 100-1000 mm·s^{-1}) are displayed. LV-HF denotes low viscosity – high friction, LV-LF low viscosity – low friction and HV-LF high viscosity – low friction iced coffee.*

	wt% MD	wt% PEG	Shear viscosity *η* (mPa·s)			Friction coefficient μ			
			1 s^{-1}	10 s^{-1}	100 s^{-1}	1 mm·s^{-1}	10 mm·s^{-1}	100 mm·s^{-1}	Exponent *b*
LV-HF	16.8	-	5.4 ± 0.1	4.6 ± 0.1	3.9 ± 0.0	0.63 ± 0.05	0.39 ± 0.02	0.39 ± 0.03	-0.494
LV-LF	-	7.0	5.6 ± 0.2	4.7 ± 0.1	4.0 ± 0.0	0.56 ± 0.05	0.32 ± 0.04	0.30 ± 0.03	-0.457
HV-LF	30.0	-	13.3 ± 0.1	11.8 ± 0.1	10.5 ± 0.1	0.55 ± 0.04	0.31 ± 0.02	0.18 ± 0.02	-0.699

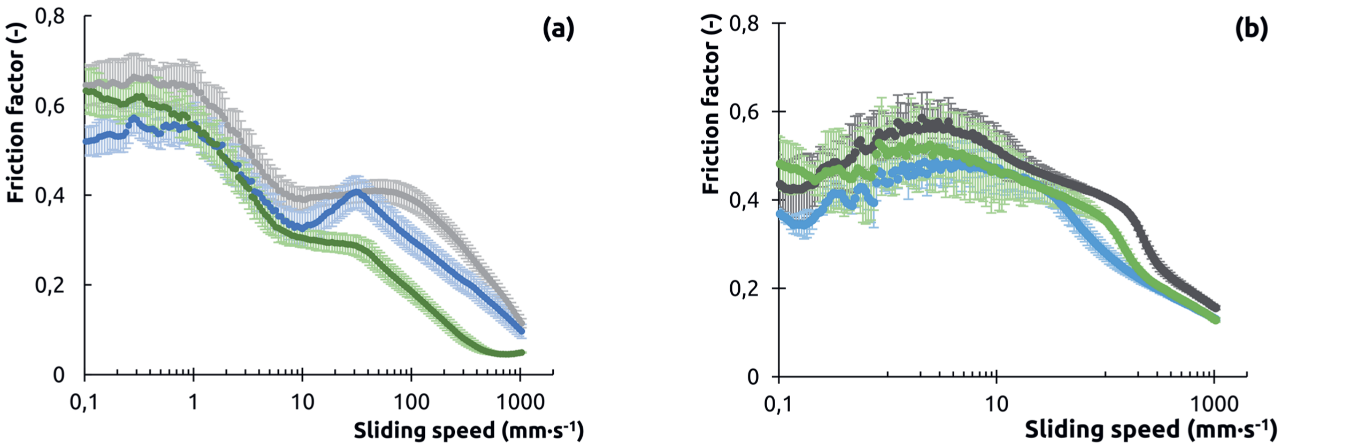

Figure 5.2. *Friction properties as a function of sliding speed of the three iced coffees in absence (a) and presence (b) of saliva. LV-HF = grey; LV-LF = blue; HV-LF = green. LV-HF denotes low viscosity – high friction, LV-LF low viscosity – low friction and HV-LF high viscosity – low friction iced coffee. Error bars represent the standard deviation of triplicate (a) or duplicate (b) measures.*

5.3.2. Paired comparisons

The frequency of selection of iced coffees being perceived as more intense than their counterpart was obtained from 2-AFC tests (Figure 5.3). HV-LF was consistently chosen as more intense for all attributes when compared with LV-HF and LV-LF iced coffees. This implies that HV-LF iced coffee was, both orally and haptically, perceived to be thicker, more slippery and creamier than the LV-HF and the LV-LF iced coffees. LV-HF was orally perceived to be thicker and creamier than LV-LF. Concerning haptic evaluation of slipperiness, LV-LF was found to be more slippery than LV-HF. There were no significant differences between LV-HF and LV-LF iced coffees regarding haptic thickness and oral slipperiness.

When comparing oral evaluation of thickness and slipperiness with those from haptic evaluation, results are consistent for paired comparisons that included HV-LF iced coffee. In oral and haptic assessment of both thickness and slipperiness, HV-LF was perceived as thicker and more slippery than LV-HF and LV-LF. However, oral and haptic thickness were not evaluated similarly for paired comparisons between LV-HF and LV-LF. No significant difference in thickness was found between the two iced coffees during haptic assessment, while LV-HF was perceived to be thicker when assessed orally. Regarding slipperiness perception, no significant difference was found between LV-HF and LV-LF during oral assessment, whereas LV-LF was found to be more slippery when assessed haptically.

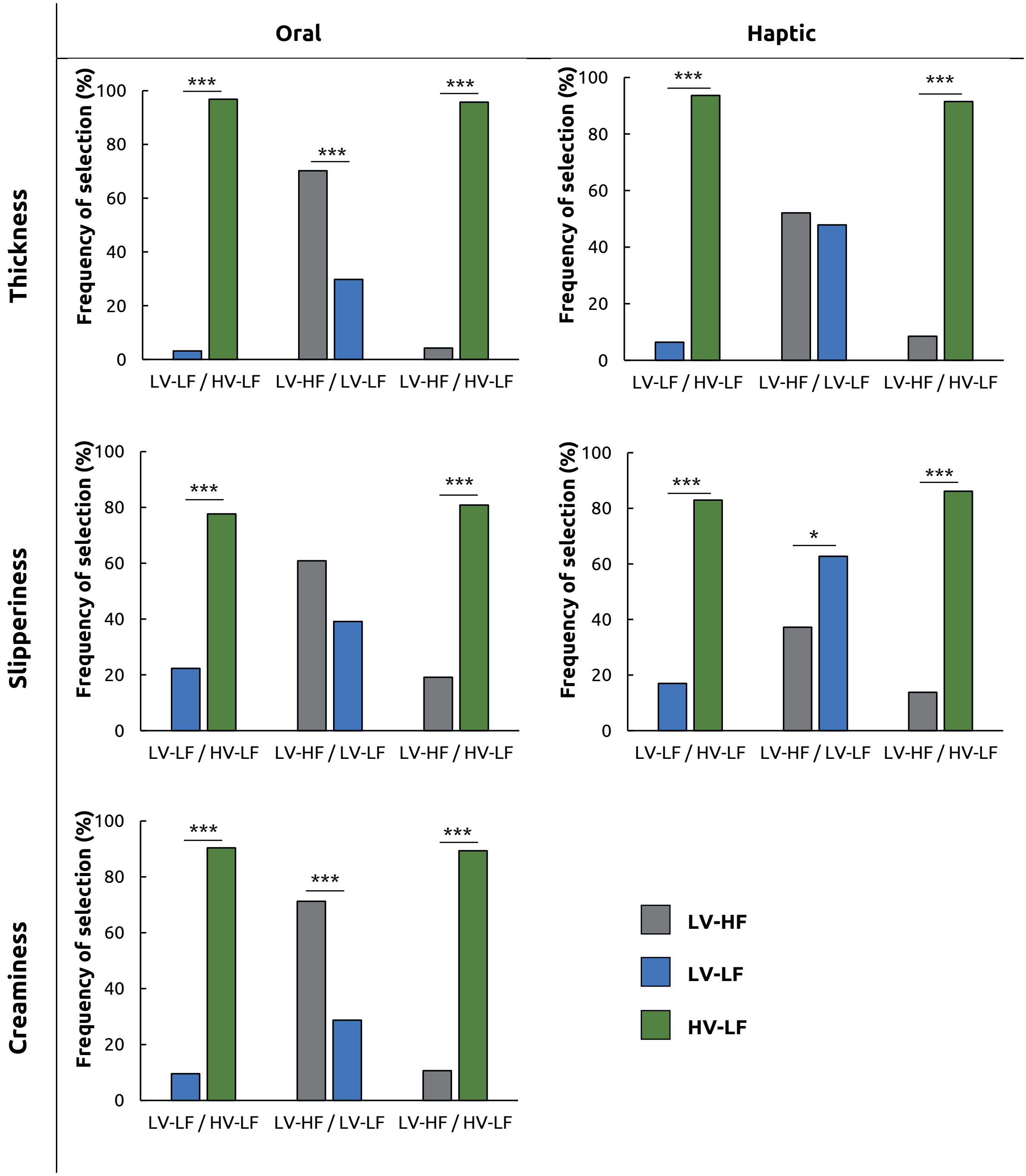

Figure 5.3. *2-Alternative Forced Choice comparisons (2-AFC): frequency of selection of iced coffees chosen to be more intense in oral and haptic thickness, oral and haptic slipperiness, and oral creaminess (n = 47, n = 46 for oral slipperiness comparisons of LV-HF and LV-LF; all in duplicate). LV-HF denotes low viscosity – high friction, LV-LF low viscosity – low friction and HV-LF high viscosity – low friction iced coffee. Asterisks indicate statistically significant differences: (*) p < 0.05; (***) p < 0.001.*

5.3.3. Perceived intensities (rank-rating)

Mean perceived intensities of the attributes obtained from rank-rating are shown in Figure 5.4. Results from rank-rating are generally in agreement with the results obtained from paired comparisons. HV-LF was perceived to be higher in intensity compared to LV-HF and LV-LF for all texture attributes. No significant differences were found between LV-HF and LV-LF in terms of haptic thickness and oral slipperiness. LV-LF was orally perceived as less creamy and less thick, but haptically more slippery than LV-HF.

Thickness perception was not affected by evaluation method (oral vs haptic; p = 0.46), but a main effect of product was found (LV-HF, LV-LF, HV-LF; $p < 0.001$). For slipperiness no main effect of evaluation method was found (p = 0.39), but a main effect of product ($p < 0.001$) and an interaction between product and evaluation method were found ($p < 0.001$). This indicates that thickness was evaluated similarly during oral and haptic assessment, whereas oral and haptic slipperiness ratings were less congruent.

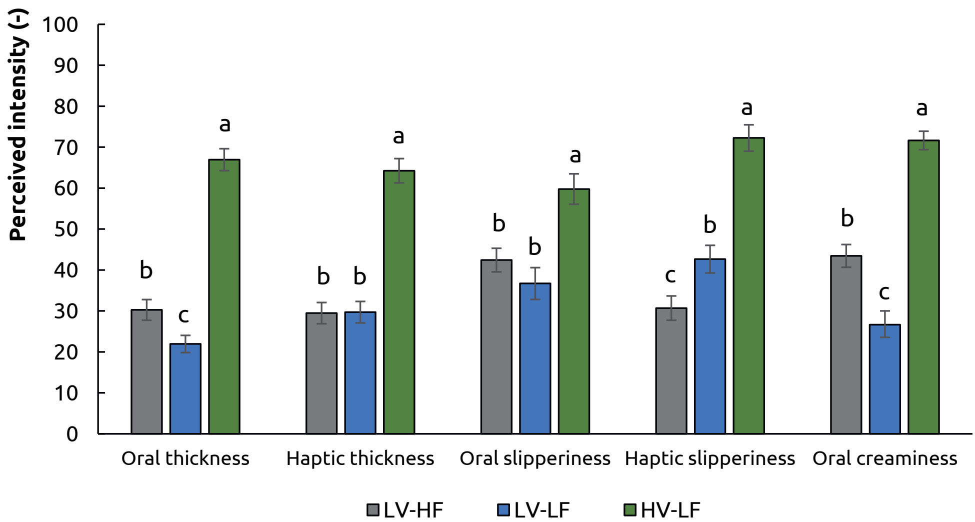

Figure 5.4. *Mean perceived intensities (± SEM) of oral and haptic thickness, oral and haptic slipperiness and oral creaminess of LV-HF, LV-LF and HV-LF iced coffees (n = 47, except for oral creaminess (n = 46)). LV-HF denotes low viscosity – high friction, LV-LF low viscosity – low friction and HV-LF high viscosity – low friction iced coffee. Within each attribute, statistically significant differences are present between the iced coffees (p < 0.001). Products containing the same letter are not significantly different from each other.*

5.3.4. Correlations between creaminess, slipperiness and thickness

Pearson correlation coefficients between rank-rating scores of the different texture attributes were calculated. Creaminess was positively correlated with oral thickness (r = 0.64, p < 0.001) and haptic thickness (r = 0.49, p < 0.001). Moderate positive correlations were found between creaminess and oral slipperiness (r = 0.35, p < 0.001) and haptic slipperiness (r = 0.45, p < 0.001). Rank-rating scores obtained from oral assessment of thickness correlated positively with those of haptic assessment (r = 0.59, p < 0.001). Slightly weaker positive correlations were found between oral and haptic slipperiness (r = 0.44, p < 0.001).

5.4. Discussion

This study aimed (i) to assess contributions of viscosity and friction properties to creaminess, thickness and slipperiness perception of iced coffees; and (ii) to compare oral and haptic thickness and slipperiness perception of these beverages. Results from paired comparisons and rank-rating were in good agreement and show that high-viscosity iced coffee (HV-LF) was more intense in all attributes (creaminess, oral and haptic thickness, oral and haptic slipperiness) compared to low-viscosity iced coffees. When comparing the two low-viscosity products, low-friction iced coffee (LV-LF) was orally perceived as less creamy and less thick, and haptically more slippery than high-friction iced coffee (LV-HF). Creaminess was stronger correlated to thickness than to slipperiness.

5.4.1. Effect of viscosity on thickness, slipperiness and creaminess perception

Results from paired comparisons and rank-rating uniformly demonstrate that increasing the viscosity of iced coffees enhanced oral and haptic perception of thickness and slipperiness, and oral perception of creaminess. These results reflect those of other studies, that found large effects of viscosity on texture perception of o/w emulsions (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; van Aken *et al.*, 2011). On the other hand, viscosity may not be the most important driver of texture perception in semi-solid foods, such as sour cream (Jervis *et al.*, 2014), cream cheese (Janhøj *et al.*, 2009) and other dairy products (Bom Frøst & Janhøj, 2007).

Participants were able to distinguish oral thickness of iced coffees with viscosities differing by a factor of 2.5. This was expected, as Camacho *et al.* (2015) reported a Just Noticeable Difference (JND) for oral thickness of 3.1 mPa·s in thin liquids (η = 10 mPa·s) and the viscosity difference in the current study was twice this reported JND value (Table 5.2). Moreover, participants were able to discriminate low- and high-viscosity iced coffees based on haptic thickness. This is in line with results from Zhong *et al.* (2018), who reported that a 1.83 and 2.05 fold increase in viscosity could be detected by haptic assessment of thickened water and thickened milk, respectively.

Liquid foods are generally perceived as creamier when viscosity increases (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; Bom Frøst & Janhøj, 2007; Daget & Joerg, 1991; Janhøj *et al.*, 2008; van Aken *et al.*, 2011; Wood, 1974), which explains why high-viscosity iced coffee was perceived as creamier than the low-viscosity products. As slipperiness has been reported to be related to friction in guar gum solutions (Malone *et al.*, 2003a) and particle dispersions (Chojnicka-Paszun *et al.*, 2014), a positive effect of viscosity on slipperiness was not expected in this study. In line with our results, others have demonstrated that more viscous liquids (*i.e.* gum-thickened water (Ong *et al.*, 2018a), apple juice, orange juice and soymilk (Kim *et al.*, 2017)) were perceived to be more slippery in mouth. An explanation may be provided by Kokini and colleagues (Kokini, 1987; Kokini *et al.*, 1977), who postulated that slipperiness is induced by the total force applied on the tongue. According to their model, slipperiness is proportional to both frictional and viscous forces, which might provide an explanation for our observation that higher viscosity resulted in increased oral and haptic slipperiness. This rationale is supported by the fact that more viscous fluids display reduced friction properties (de Vicente *et al.*, 2005, 2006), which are in turn related to slipperiness perception (Chojnicka-Paszun *et al.*, 2014; de Wijk & Prinz, 2005; Malone *et al.*, 2003a).

5.4.2. Effect of friction properties on thickness, slipperiness and creaminess perception

Results from paired comparisons and rank-rating of low-viscosity iced coffees show that reduced friction by addition of PEG (LV-LF vs LV-HF) leads to decreased oral thickness and creaminess, but increased haptic slipperiness. Nevertheless, sensory differences due to addition of PEG were generally smaller than those elicited by increasing the viscosity of iced coffee. The fact that sensory differences between low-viscosity products were relatively small suggests that the difference in friction properties only led to subtle effects in texture perception. It should be noted that the range of friction properties covered in this study (maximum difference in friction coefficients between iced coffees was a factor 1.3) was smaller than that of viscosity (maximum difference in viscosity between iced coffees was a factor 2.5), which can partially explain the limited effect of friction properties on sensory characteristics of the iced coffees (see section 4.5 Limitations).

Reduction of friction by addition of maltodextrin resulted in increased perceived oral and haptic thickness, whereas addition of PEG did not affect perceived haptic thickness of low-viscosity iced coffees (Figure 5.3 and 5.4). This was in line with our expectations, as thickness perception is associated with viscosity (Akhtar *et al.*, 2006;

Akhtar *et al.*, 2005; Cutler *et al.*, 1983; Richardson *et al.*, 1989) and the two low-viscosity iced coffees were iso-viscous. In contrast, perceived oral thickness decreased when friction was reduced by adding PEG. In a study on vanilla custard desserts (de Wijk & Prinz, 2005), no correlation was found between friction properties and perceived thickness evaluated by a trained panel. A dumping effect may have occurred as participants in the current study were instructed to focus on one attribute, while other attributes were present that distinguished the products.

As expected, low-friction iced coffees were haptically perceived as more slippery than high-friction iced coffee. Reduction of friction by addition of maltodextrin also enhanced oral slipperiness of the high-viscosity product, whereas addition of PEG did not affect oral slipperiness of low-viscosity iced coffee. As negative relationships between friction and oral slipperiness were previously established in guar gum solutions (Malone *et al.*, 2003a), particle dispersions (Chojnicka-Paszun *et al.*, 2014) and vanilla desserts (de Wijk & Prinz, 2005), a similar effect was expected in this study. The fact that participants only perceived a difference in slipperiness during haptic evaluation of low-viscosity products, suggests a possible effect of the oral environment on slipperiness perception of iced coffees. Iced coffees are warmed by the oral surfaces and diluted by saliva, which may consequently affect oral texture perception of the beverages. Saliva affects lubrication (Tabak, 1995) and the presence of saliva in the oral environment might therefore affect texture perception of the iced coffees (Engelen *et al.*, 2003). Moreover, food-saliva interactions might have occurred that affect texture perception (Dresselhuis *et al.*, 2008; Rossetti *et al.*, 2008; Selway & Stokes, 2013). These factors may clarify why differences in friction properties between the three iced coffees become smaller upon addition of saliva (Figure 5.2b), as has previously been reported by Joyner *et al.* (2014) as well. It should be noted that the iced coffees were mixed with saliva at a 1:1 ratio, *i.e.* the products were diluted, which may partially explain the smaller differences in friction properties upon addition of saliva. A mixing ratio of 1:1 was chosen to test whether saliva had any effect on friction properties of iced coffees. This mixing ratio overestimates the amount of saliva mixed with iced coffee, since under realistic drinking conditions the amount of saliva that mixes with iced coffee during oral processing is likely to be considerably smaller due to the short residence time of iced coffee in the oral cavity.

Reduced friction was hypothesised to enhance creaminess of iced coffees, as this has been described in several reviews (de Wijk *et al.*, 2006; Dickinson, 2018; Upadhyay *et al.*, 2020). While this was observed upon addition of maltodextrin in high-viscosity iced coffee, this was not the case when PEG was added to low-viscosity iced coffee. When comparing the low-viscosity products, low-friction iced coffee was perceived as less creamy than high-friction iced coffee (Figure 5.3 and 5.4). In a comparable study on milks, no effect on creaminess was observed by a trained panel after reduction of friction by addition of inulin (Meyer *et al.*, 2011). It is known that perceived thickness is important for creaminess of foods (Bom Frøst & Janhøj, 2007; Kokini & Cussler, 1983). The fact that participants perceived low-viscosity iced coffee with low friction as less thick than the high-friction product (Figure 5.3 and 5.4) might therefore have caused a reduction in perceived creaminess.

5.4.3. Comparison of effect of viscosity and friction properties on creaminess perception

Identifying the roles of viscosity and friction properties could provide fundamental answers to understanding texture perception, and more specifically creaminess, of iced coffees. It was hypothesised that higher viscosity and lower friction would enhance creaminess of iced coffees (Kokini, 1987; Kokini & Cussler, 1983). Results from paired comparisons and rank-rating confirm that higher viscosity resulted in enhanced creaminess perception. Reduction of friction by addition of PEG, on the other hand, did not have the expected positive impact on creaminess. This finding is contrary to those from previous studies on milk (Chojnicka-Paszun *et al.*, 2012), custard desserts (de Wijk & Prinz, 2005), yoghurts (Sonne *et al.*, 2014) and cream cheese (Janhøj *et al.*, 2009) in which perception of creaminess was associated with lower friction coefficients. The reduction in friction properties achieved in this study was not necessarily sufficient to increase creaminess of iced coffees, whereas simultaneously increasing viscosity and decreasing friction enhanced creaminess.

Since viscosity is related to thickness perception (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; Cutler *et al.*, 1983; Richardson *et al.*, 1989) and friction properties are related to slipperiness perception (Chojnicka-Paszun *et al.*, 2014; Malone *et al.*, 2003a), it was hypothesised that perceived thickness and slipperiness are related to creaminess perception (Richardson-Harman *et al.*, 2000). In the current study, correlation coefficients between creaminess and oral thickness scores were higher (r = 0.64) compared to creaminess and oral slipperiness scores (r = 0.35). This finding is in agreement with results from others who suggested that viscosity was the major contributor to creaminess in o/w emulsions (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005) and acidified milk drinks (Janhøj *et al.*, 2008), and that adjustment of friction properties did not have a large effect on creaminess of milks (Meyer *et al.*, 2011).

Our results can be compared with early work of Kokini *et al.* (1977), who modelled creaminess perception in liquid foods. They determined that the creaminess of liquid foods could be predicted by perceived thickness, smoothness and slipperiness, using the following formula (R^2 = 0.88):

$$\log(\boldsymbol{Creamy}) = \mathbf{0.539} \cdot \log(\boldsymbol{Thick}) + \mathbf{0.728} \cdot \log(\boldsymbol{Smooth}) + \mathbf{0.220} \cdot \log(\boldsymbol{Slippery})$$

In line with our findings, the authors concluded that thickness contributes more to creaminess perception than slipperiness. Nevertheless, they also established a large contribution of perceived smoothness to the prediction of perceived creaminess. Kokini and colleagues postulated that smoothness is related to friction forces on the tongue. Our results suggest that small differences in friction properties are not sufficient to modify creaminess of iced coffees. Friction properties only become dominant during later stages of oral processing (Chen & Stokes, 2012; Stokes *et al.*, 2013), which may explain why a smaller contribution of friction properties to creaminess was found in the current study. Viscosity might be more important for creaminess perception of iced coffees, as oral texture perception is initially governed by viscous properties of foods. During this early stage of oral processing, viscosity may ensure that the iced coffee is retained in the mouth long enough for creaminess to be perceived (Wood, 1974). However, the oral processing time of iced coffee is relatively short and might therefore be too short for friction properties to be perceived.

5.4.4. Comparison of oral and haptic assessment of texture attributes

The second aim of the study was to compare oral evaluations of thickness and slipperiness with haptic evaluations. Although iced coffee is usually not touched by the hands during consumption, we included haptic evaluation to obtain a complete image on how texture perception might be influenced by evaluation methods. Participants were able to orally and haptically discriminate low viscous iced coffees with viscosities differing by a factor of 2.5 based on both thickness and slipperiness. They were moreover able to discriminate haptic slipperiness of low viscous iced coffees with friction coefficients differing by 25%, whereas these iced coffees could not be discriminated based on oral slipperiness. Despite the fact that mechanoreceptors on the tongue have been reported to be more sensitive to force than those in the finger tips (Rath & Essick, 1990; Trulsson & Essick, 1997), we found comparable results for different evaluation methods of thickness (*i.e.* oral vs haptic). In contrast, others found that liquids were perceived as more viscous when evaluated orally compared to non-oral methods (Christensen & Casper, 1987) or that oral viscosity discrimination was slightly better than haptic discrimination of syrup solutions (Aktar *et al.*, 2015). While in the current study rank-rating scores for oral thickness were congruent with those of haptic thickness, an interaction effect between evaluation method and product was found for slipperiness scores. Accordingly, the correlation between oral and haptic rank-rating scores for thickness (r = 0.59) is higher than the correlation between oral and haptic slipperiness scores (r = 0.44). All in all, our results imply that oral and haptic thickness are evaluated similarly, whereas haptic slipperiness is assessed differently from oral slipperiness.

5.4.5. Limitations

This study aimed to assess the individual contributions of viscosity and friction properties to texture perception of iced coffees. The effect of viscosity was assessed by comparing iced coffees with different viscosities, but similar friction properties. However, as viscosity and friction are physically related concepts, preparing liquids that vary in viscosity but not in friction poses a challenge. In practice it was difficult to obtain similar friction coefficients over the entire sliding speed range, as low friction was obtained in two ways: (a) by adding PEG to retain low viscosity; and (b) by adding maltodextrin, hence this decrease in friction was accompanied by an increase in viscosity. As friction in the mixed regime depends on surface properties as well as bulk viscosity of the liquid (de Vicente *et al.*, 2005, 2006; Stokes *et al.*, 2013), it was not possible to formulate a low-viscosity product with similar friction properties in the mixed regime as the high-viscosity iced coffee. Low-viscosity iced coffee (LV-LF) had reduced friction coefficients in the boundary regime (<1 $mm{\cdot}s^{-1}$), whereas high-viscosity iced coffee (HV-LF) exhibited reduced friction coefficients in the mixed regime (10-1000 $mm{\cdot}s^{-1}$). The fact that viscosity affects friction also explains why friction coefficients of the two low-viscosity products are comparable in the mixed regime, as viscosity is an important determinant for friction in this regime. Differences between the products regarding friction therefore depend on the sliding speed that is discussed, which makes it more difficult to compare the low-friction iced coffees.

Our results suggest that viscosity might contribute more to creaminess of iced coffees than friction properties. Although this is in accordance with previous findings on o/w emulsions (Akhtar *et al.*, 2006; Akhtar *et al.*, 2005), milks (Meyer *et al.*, 2011) and acidified milk drinks (Janhøj *et al.*, 2008), the difference in friction properties between high- and low-friction iced coffees in this study might have been too small to be reflected in perception of texture attributes. Viscosity of the iced coffees was varied by a factor of 2.5, whereas friction was varied by a factor of maximum 1.3 due to addition of PEG. If we assume a linear or semi-log relationship between the physical-chemical food properties and perceived creaminess, it is reasonable that a larger contribution of viscosity is found, as viscosity of the iced coffees was varied to a higher degree. Larger effects on perceived creaminess and slipperiness might arise when friction properties are modified to a larger degree. PEG with higher M_w is capable of reducing friction properties to a larger extent without considerably affecting viscosity. However, due to legal restrictions regarding the use of high M_w PEG in foods (Code of Federal Regulations, 1984; Joint FAO/WHO Expert Committee on Food Additives (JECFA)/Food and Agriculture Organization of the United Nations (FAO), 1980), such studies would be limited to haptic evaluations only.

Care should be taken when comparing results from the current study to other studies linking tribological properties to sensory attributes. The comprehensive review by Sarkar and Krop (2019) highlights that such tribology-sensory relationships are only valid for the specific food and experimental set-up used, as friction properties depend on the interplay of the food with the surfaces of the tribo-pair. Due to this and the fact that only iced coffees were used, results from the current study cannot be generalised to liquid foods. Further research using a standardised tribological set-up and methodology is needed to establish clear relationships between friction and perception of specific texture attributes.

Due to the addition of maltodextrin or PEG to the iced coffees, the final fat content of the three products was not identical (0.63-0.84 % (w/v)). Chojnicka-Paszun *et al.* (2012) found that friction properties and perceived creaminess of milks are independent of fat content at fat contents below 1%. Furthermore, Akhtar *et al.* (2005) showed that fat content had no effect on perception of thickness and creaminess of low-viscosity emulsions (8 mPa·s). Therefore, we argue that the small difference in fat content did not considerably influence the friction properties or perceived creaminess of the iced coffees.

As only small differences between the iced coffees were expected, a 2-AFC test was performed. Such discrimination tests do not require trained panellists and are generally better at detecting small differences between products compared to intensity ratings (Lawless & Heymann, 2010). However, as we were also interested in the magnitude of the differences between the products, the discrimination test was followed by a rank-rating procedure. The fact that results from both sensory methods led to the same conclusions indicates that participants were consistent in evaluating the iced coffees. While dumping effects may be eliminated by using a trained panel, results from an untrained panel are more representative of perception by consumers.

5.5. Conclusions

To the best of our knowledge, this is the first study that aimed to assess the relative contributions of viscosity and friction properties to texture perception by varying viscosity of iced coffees with minimal changes in friction properties and *vice versa*. This study has identified that increasing the viscosity enhances creaminess, thickness and slipperiness of iced coffees. On the other hand, reduced friction in the boundary regime resulted in lower oral thickness and creaminess scores, but increased perception of haptic slipperiness of iced coffees. Creaminess was stronger correlated to thickness than to slipperiness. The oral processing time of iced coffees was possibly too short for textural differences related to friction properties to be perceived, as texture perception is initially governed by viscous properties of food, thereby limiting the time for friction properties to be perceived. Therefore, in development of low-fat milk-based beverages with creamy texture, product developers might want to focus more on viscosity than friction properties. Future research should focus on developing a range of foods (*i.e.* liquid to semi-solid) that differ considerably in friction properties without strongly affecting viscosity to elucidate the individual contributions of friction and viscosity to creaminess perception in foods.

Acknowledgements

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6

General Discussion

6. General Discussion

This thesis aimed to '*unravel the unravelled*' by assessing the suitability of microfibrillated cellulose (MFC) as a food thickener. The effect of MFC on sensory, rheological and tribological properties of liquid and semi-solid foods was compared to other common thickeners. MFC was incorporated in several liquid and semi-solid (model) foods at various concentrations and multiple sensory (Rate-All-That-Apply, rank-rating) and instrumental methods (shear rheology, extensional rheology, tribology, microscopy) were employed for characterisation. Correlations between sensory and instrumental physico-chemical properties were determined in order to elucidate the mechanisms underlying perception. A schematic overview of the framework and the main outcomes of this thesis are presented in Figure 6.1.

In **Chapter 2**, liquid and semi-solid model foods containing different concentrations of MFC were compared to iso-viscous xanthan gum solutions. Shear viscosity and yield stress increased linearly with increasing xanthan gum concentration, whereas a power law relationship was observed between rheological properties and MFC concentration. At similar shear viscosity, MFC dispersions were less slimy, sticky and mouthcoating compared to xanthan gum solutions. The absence of these undesirable sensory properties was confirmed in **Chapter 3**, in which low-fat mayonnaises thickened with MFC were compared to mayonnaises thickened with various commercially used thickeners. Sensory mouthfeel properties of mayonnaises thickened with MFC closely resembled those of mayonnaises thickened with conventionally used chemically modified starch and native starch, whereas the presence of xanthan gum elicited undesirable sensory properties such as sliminess and pulpiness. Sliminess was correlated with shear-thinning behaviour of the mayonnaises. Although shear viscosity and yield stress of mayonnaises thickened with MFC were lower compared to the other thickeners, using MFC as a thickening agent induced highest storage modulus, loss modulus and boundary friction. As the use of MFC in dispersed form has disadvantages in terms of storage and transport efficiency, the impact of addition of spray-dried MFC particles to liquid foods on rheological, tribological and sensory properties was explored in **Chapter 4**. Particles with lower MFC:maltodextrin ratio were more efficient than particles with higher MFC:maltodextrin ratio at increasing the viscosity of milks and soups. As a consequence, perceived thickness and creaminess were enhanced more effectively upon addition of low-ratio particles. It is expected that higher MFC:maltodextrin ratios generated fewer but denser particles and were therefore less effective at enhancing viscosity, thickness and creaminess. Sensory thickness and creaminess were positively correlated and strongly correlated with rheological properties. The results demonstrate that spray-drying is a technology that allows drying of MFC while preserving its functionality upon redispersion, and that the acquired spray-dried MFC particles can be used as thickener in liquid foods. **Chapter 5** explored the impact of rheological and tribological properties of liquid foods on creaminess perception. The results suggest that creaminess of liquid foods (iced coffee) may be affected more by viscosity than by friction, and that reduction of friction does not necessarily enhance creaminess.

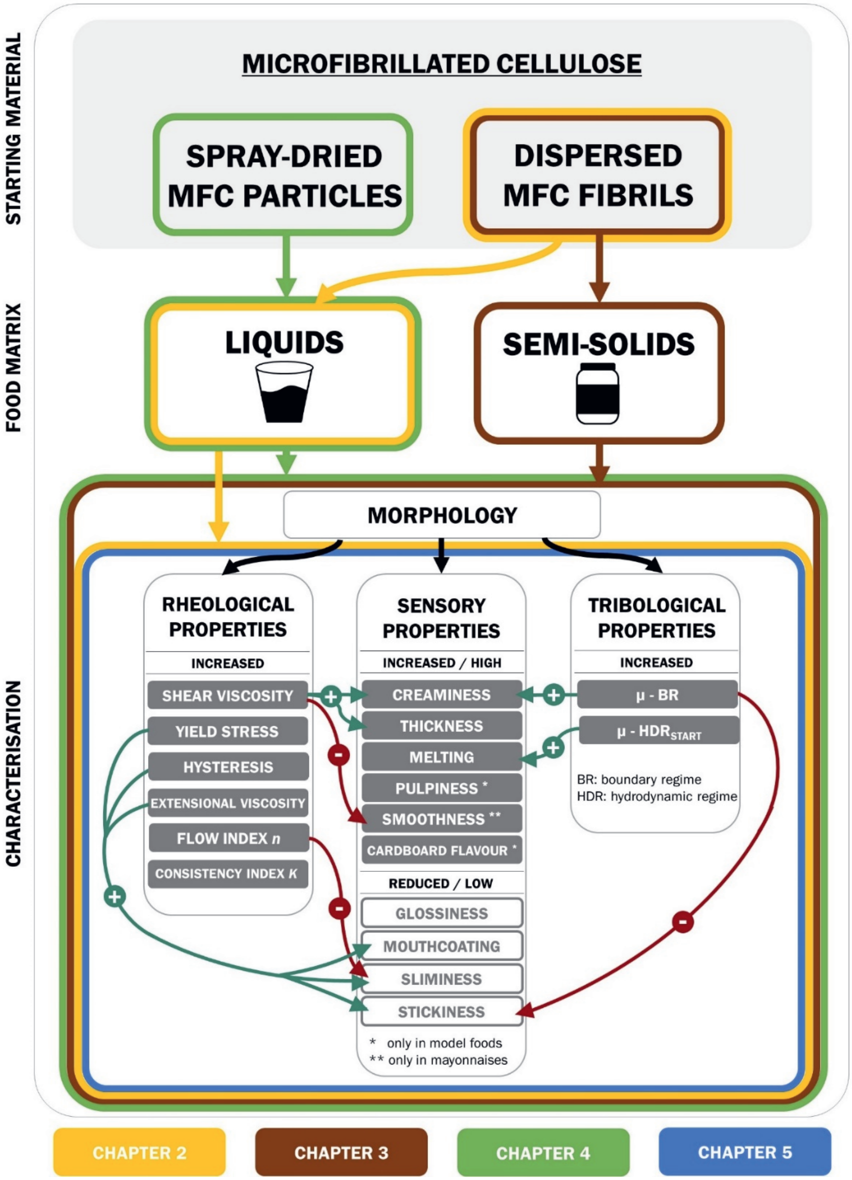

Figure 6.1. *Schematic overview of the microfibrillated materials, products and properties studied in this thesis. The overview shows the rheological, tribological and sensory properties determined for (model) foods containing microfibrillated cellulose and correlations found between these parameters. Green arrows represent positive correlations, red arrows represent negative correlations.*

6.1. Discussion of main results

6.1.1. Effect of MFC on rheological, tribological and sensory properties of liquid and semi-solid foods

Microfibrillated cellulose is traditionally incorporated in foods either by addition of previously prepared aqueous MFC dispersions, or by single stage preparation in which the cellulosic material is microfibrillated *in situ*. Although both methods are promising (e.g. Turbak *et al.* (1982), Kleinschmidt *et al.* (1988), Velásquez-Cock *et al.* (2019)) several attempts have been made to dry MFC for subsequent use as texture modifier in foods. Dehydration facilitates transport and storage of MFC by improving the material's stability and shelf life. This thesis studied the effect of two types of MFC prepared from the same starting material (citrus fibre): dispersed microfibrillated cellulose fibrils prepared using the conventional high-shear treatment developed in the 1980s (**Chapter 2 and 3**) and spray-dried microfibrillated cellulose particles (**Chapter 4**) (Figure 6.2). The following section compares the two types of MFC and discusses their effects on rheological, tribological and sensory properties of liquid and semi-solid foods.

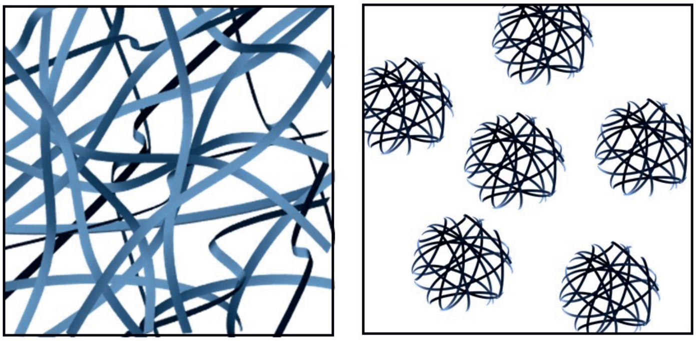

Figure 6.2. *Schematic representation of the microstructure of the entangled MFC network formed by dispersed microfibrillated cellulose fibrils (**Chapters 2, 3**) and spray-dried MFC particles (**Chapter 4**).*

Effect of MFC on rheological properties of liquid and semi-solid foods

Rheological properties such as viscosity, storage and loss modulus increased upon addition of MFC to foods in dispersed form (aqueous model foods, **Chapter 2**; low-fat mayonnaises, **Chapter 3**) or as spray-dried MFC-maltodextrin particles (milks and soups, **Chapter 4**). High shear homogenisation releases cellulose microfibril aggregates and individual microfibrils from the citrus fibre suspension, which have a large aspect ratio and can interconnect through the formation of junction zones. At sufficiently high MFC concentrations a disordered entangled network is formed displaying high viscosity or even gel-like characteristics. Lowys *et al.* (2001) determined the critical overlap concentration C^* for the formation of entangled MFC networks to be C^* = 3 g/l (0.3 wt%), which is similar to observations from this thesis in which stable dispersions were formed at concentrations of 0.2 wt% MFC. Higher MFC concentrations facilitate the formation of stronger entangled networks, which is reflected in higher viscosity η, storage modulus G' and yield stress σ_y (Figure 6.3-6.5, Iotti *et al.*, 2011; Pääkkö *et al.*, 2007). The MFC network can be spray-dried to facilitate storage of the powdered material (**Chapter 4**).

The relationship between polymer concentration and storage modulus G' shows a power law dependence ($G' \propto \phi^n$) in which ϕ represents the polymer (MFC) concentration and n the scaling factor (Guenet, 2000; Jones & Marques, 1990). In this thesis scaling factors of 1.8 and 2.6 were found for dispersed MFC fibrils added to low-fat mayonnaises and aqueous model foods, respectively (Figure 6.3). In literature, scaling factors n between 2-5 have been reported for aqueous MFC dispersions (Lundahl *et al.*, 2018). The wide range of scaling factors is caused by differences in the microstructure of the microfibrillated material. Morphological properties of MFC such as microfibril length and fibrillation degree depend on the origin of the cellulosic material, possible pre-treatments and the microfibrillation process (*i.e.* type of equipment used, the amount of shear applied, number of passes) and largely affect the power law relationship between MFC concentration and storage modulus G'. Slightly stronger power law relationships between G' and concentration of spray-dried MFC particles are observed, with scaling factors n of 2.6 and 3.2 in skimmed milks and soups, respectively. Interestingly, the power law exponent of spray-dried MFC particles in milk is identical to that of aqueous MFC dispersions, and these in turn match the exponent found by Agoda-Tandjawa *et al.* (2010). This suggests that an increase in MFC concentration results in the same proportional increase in storage modulus G' for dehydrated MFC particles compared to dispersed MFC fibrils. Strongest effects of MFC concentration on G' were found for spray-dried MFC particles in soups, which can be attributed to the presence of other biopolymers in the soup (*i.e.* starch, guar gum) that affect the strength of the network.

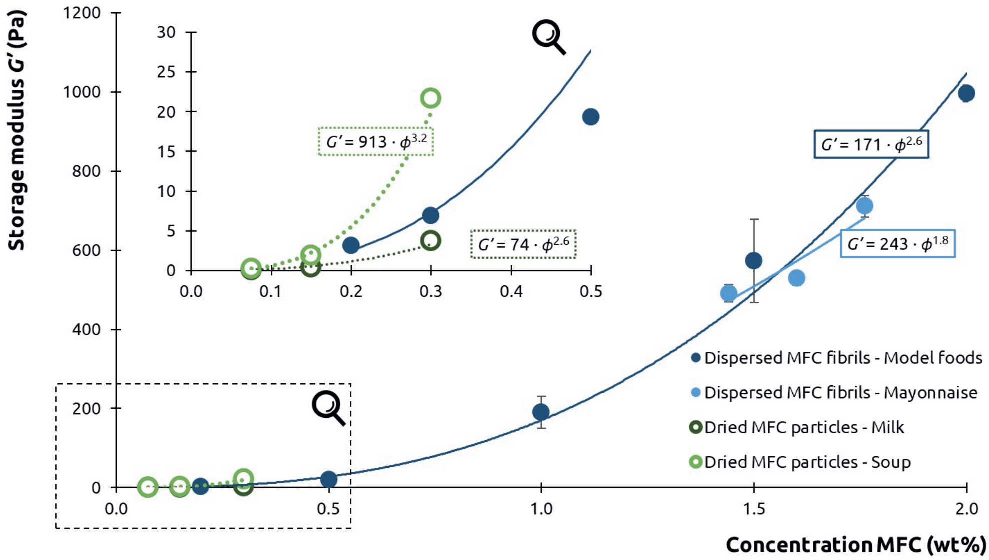

Figure 6.3. *Relationship between MFC concentration (wt%) and storage modulus G′. A comparison is made between dispersed MFC fibrils (in aqueous model foods (dark blue) and low-fat mayonnaises (light blue)) and spray-dried MFC particles (low MFC:maltodextrin ratio; in milk (dark green) and soup (light green)). The inset zooms in at lower MFC concentrations and displays in more detail the effect of spray-dried MFC particles on storage modulus G′. Please note that the horizontal axis displays the concentration of MFC in the final sample, rather than the concentration of MFC-maltodextrin particles added. Lines represent power law fits through the experimental data points.*

Power law relationships between MFC concentration and shear viscosity η at 50 s^{-1} (Figure 6.4) and between MFC concentration and yield stress σ_y (Figure 6.5) were observed (**Chapter 2 and 3**). Higher scaling factors *n*, and thus stronger power law relationships, were found for aqueous MFC dispersions compared to low-fat mayonnaises thickened with MFC. In addition to enhancing viscosity, MFC can act as a surfactant in emulsified foods such as mayonnaises (e.g. Choublab & Winuprasith, 2018). Microfibrils located at the oil-water interface are unable to simultaneously function as an emulsifier and viscosifier. When part of the added MFC adsorbs at the oil-water interface the amount of MFC available for the formation of a viscosifying entangled network is reduced, resulting in weaker networks and consequently smaller scaling factors *n*. Rather than a power law relation, an exponential dependence was observed between shear viscosity η (at 50 s^{-1}) and MFC concentration of spray-dried MFC-maltodextrin particles (Figure 6.4). This is expected to be due to differences in the mechanisms by which the respective materials enhance viscosity. An entangled space-spanning network is formed by dispersed MFC fibrils, whereas the presence of MFC particles increases viscosity by drag reduction rather than by the formation of a network (Figure 6.2). Considering that the materials were not applied in the same (food) matrix, it is difficult to make direct comparisons between the two types of MFC. The fairest comparison was assumed to be between aqueous model foods of dispersed MFC and skimmed milk

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with MFC particles, as both matrices predominantly consist of water (> 90%). Figure 6.4 demonstrates that at around 0.2-0.3 wt%, the effect of MFC concentration on shear viscosity η is relatively similar for dispersed MFC fibrils and MFC from spray-dried particles. This suggests that the effect of MFC on shear viscosity is independent of the form in which it is applied at this concentration. Milks and soups containing spray-dried MFC particles primarily exhibited viscous behaviour (*i.e.* loss modulus *G"* > storage modulus *G'*). These samples did not exhibit a yield stress, hence no comparison between spray-dried MFC particles and dispersed MFC fibrils could be made for yield stress in Figure 6.5. It should be noted that these outcomes on the dependency of rheological properties on MFC concentration cannot simply be generalised to other food matrices, as the results are based on a limited set of data. Especially for spray-dried MFC particles, trendlines are based on 3-4 datapoints and additional experiments should be performed to validate the models.

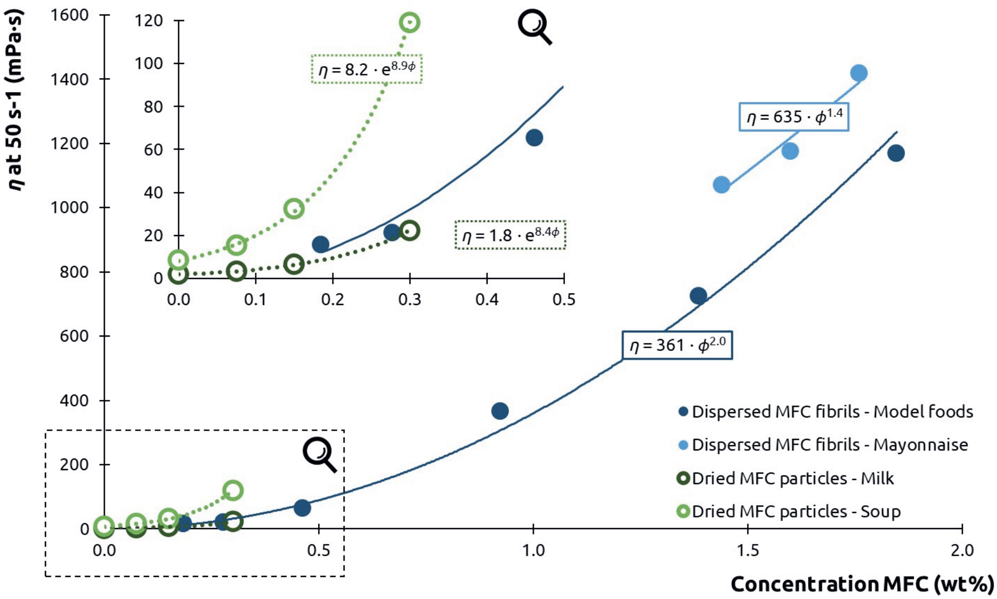

Figure 6.4. *Relationship between MFC concentration (wt%) and shear viscosity η (at 50 s^{-1}). A comparison is made between dispersed MFC fibrils (in aqueous model foods (dark blue) and low-fat mayonnaises (light blue)) and spray-dried MFC particles (low MFC:maltodextrin ratio; in milk (dark green) and soup (light green)). The inset zooms in at lower MFC concentrations and displays in more detail the effect of spray-dried MFC particles on shear viscosity η. Please note that the horizontal axis displays the concentration of MFC in the final sample, rather than the concentration of MFC-maltodextrin particles added. Lines represent power law or exponential fits through the experimental data points.*

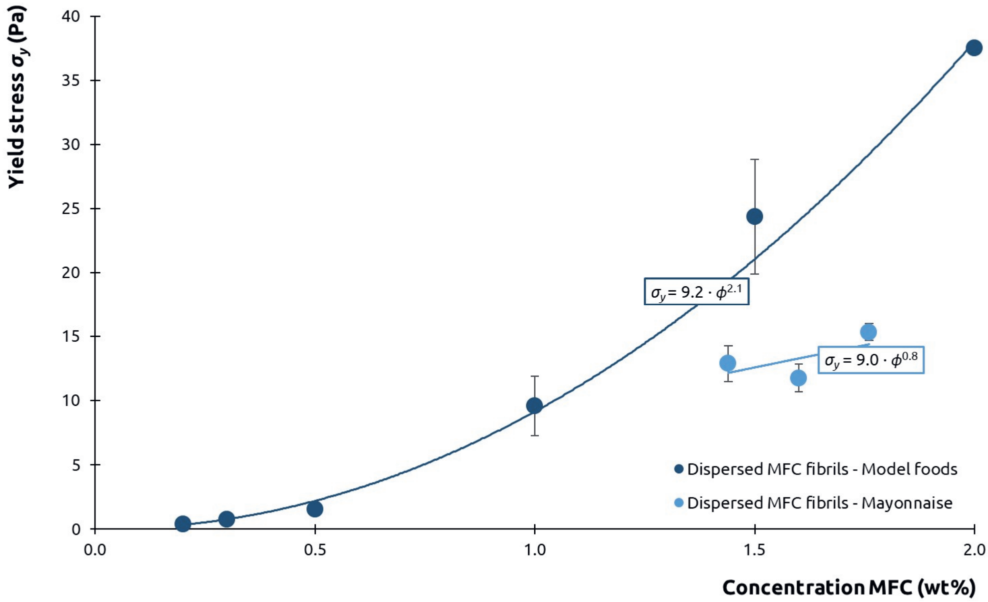

Figure 6.5. *Relationship between MFC concentration (wt%) and yield stress σ_y of dispersed MFC fibrils in aqueous model foods (dark blue) and low-fat mayonnaises (light blue). Lines represent power law fits through the experimental data points.*

Effect of MFC on tribological properties of liquid and semi-solid foods

Although varying the concentration of dispersed MFC fibrils caused minor differences in tribological properties of low-fat mayonnaises (**Chapter 3**), highly similar friction curves were obtained for (model) foods containing various concentrations of dispersed MFC fibrils and spray-dried MFC particles (**Chapter 2, 4**). This was unexpected as polymer concentration is well-known to affect friction in the boundary and mixed regime (Garrec & Norton, 2012; Stokes, 2012). Indeed, friction coefficients of xanthan gum solutions decreased with increasing xanthan gum concentration (**Chapter 2**). The increase in viscosity with increasing xanthan gum concentration contributes to the decline in friction coefficient. The fact that varying the MFC concentration hardly affected friction of liquid foods suggests that the microfibrillated material was excluded from the narrow gap between the glass ball and PDMS pins during the tribological measurement (Figure 6.6). Although cellulose microfibrils have lengths up to several micrometres, highly entangled three-dimensional networks with large hydrodynamic volumes are formed above the critical overlap concentration *C**. It is speculated that the hydrodynamic volume occupied by the MFC network is too large for MFC to be entrained between the tribological surfaces. Presumably the tribological properties of the continuous phase were determined instead, which explains the strong similarities between friction curves of samples with varying MFC contents. Removal of microfibrillated material from MFC dispersions by centrifugation yielded similar friction curves to uncentrifuged MFC dispersions (data not shown), which demonstrates that MFC was excluded from the tribological gap. Small differences in friction curves were observed between low-fat mayonnaises with varying MFC content (**Chapter 3**), suggesting that MFC fibrils embedded in mayonnaises were entrained between the tribological surfaces. These samples might be better lubricants for the hydrophobic PDMS pins due to their higher fat content (20%) compared to fat-free aqueous model foods, which might have facilitated entrainment between the surfaces (Dresselhuis *et al.*, 2007). Another possible explanation is that the higher viscosity of semi-solid mayonnaises facilitated entrainment of the MFC fibrils in the tribological gap (Cassin *et al.*, 2001). The fluid pressure that is built up at the contact zone increases with increasing lubricant viscosity, and in turn promotes entrainment of the lubricant (Garrec & Norton, 2012).

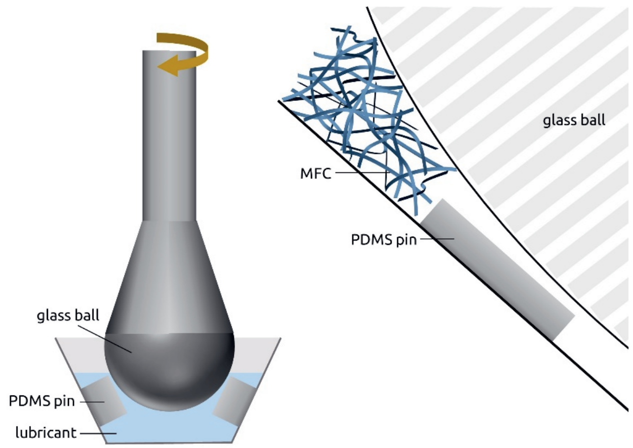

Figure 6.6. *Schematic overview of tribological set-up used throughout this thesis (ball-on-three-pins set-up) and the speculated mechanism behind the exclusion of MFC fibrils from the tribological gap.*

Similar to dispersed MFC fibrils, varying the concentration of spray-dried MFC particles did not affect friction of milks and soups (**Chapter 4**). The particles are around 10 μm in size, but their spherical network is hypothesised to be disrupted by the high shear employed in the tribological contact zone (>1000 s^{-1}; de Vicente *et al.* (2006)). The released rigid (aggregates of) microfibrils are unlikely to coil, which makes them subject to volume exclusion effects and promotes the formation of junction zones between them (Lundahl *et al.*, 2018). An entangled MFC network can be formed, which implies that the dispersion of spherical MFC particles is converted into a dispersion similar to the ones studied in **Chapters 2 and 3**. The volume-spanning network formed by the disintegrated MFC particles was presumably too large to enter the tribological gap. The anticipated lubricating effect of dispersed MFC fibrils and spray-dried MFC particles could thus not be properly determined using the current ball-on-three-pins set-up (Figure 6.6). This does not imply that MFC cannot act as a lubricant in liquid and semi-solid foods, since friction properties do not only result from the sample but also depend on the tribometer and tribological set-up used. The tribometer used throughout this thesis (ball-on-three-pins, Anton Paar) can exclusively perform rotational movements and therefore does not simulate the movements occurring in the mouth (Rudge *et al.*, 2019). Another deviation from oral conditions is the absence of saliva in the tribological experiments. Other ways of assessing the lubricating properties of MFC should therefore be explored, for instance using lower normal forces, larger surface areas or other types of tribometers. A more detailed discussion on methodological considerations on the tribological set-up used can be found in section 6.2.3.

Effect of MFC on sensory properties of liquid and semi-solid foods

The effect of MFC on rheological and potentially lubrication properties of foods is reflected in the way these foods are sensorially perceived. An increase in shear viscosity, yield stress and storage modulus (G') is typically expressed in higher thickness intensities, both in terms of appearance (**Chapter 2, 3**) and mouthfeel of liquid and semi-solid (model) foods (**Chapter 2-4**). This relation has been described extensively in literature (e.g. Cutler *et al.*, 1983; Deblais *et al.*, 2021; Kokini, 1987; Richardson *et al.*, 1989). Sensory perception of creamy, slimy, sticky and mouthcoating texture increased with higher MFC concentrations, which is presumably caused by the increase in viscosity (**Chapter 2-4**). These findings substantiate prior correlations between viscosity and creaminess (as reviewed in for instance Bom Frøst & Janhøj, 2007; de Wijk *et al.*, 2006; Dickinson, 2018; Upadhyay *et al.*, 2020), sliminess (e.g. Brandenstein *et al.*, 2015; Lyly *et al.*, 2003), stickiness (e.g. He *et al.*, 2016; Morris *et al.*, 1984; Ross *et al.*, 2019) and mouthcoating texture (e.g. He *et al.*, 2016; Ross *et al.*, 2019; Wagoner *et al.*, 2020). It was anticipated that perception of several sensory attributes is also associated with friction properties, such as creaminess (Kokini, 1987) and stickiness (Devezeaux de Lavergne *et al.*, 2016). However as described earlier, friction properties of foods containing MFC could not be determined properly and the contribution of tribological properties to sensory perception could therefore not be assessed. The presence of MFC furthermore affected the appearance of foods by lowering transparency, colour and glossiness intensities (**Chapter 2, 3**). The water insoluble microfibrils and microfibril aggregates scatter light and consequently reduce transparency of the foods in which they are dispersed (Hutchings, 1994). The presence of dispersed fibrils furthermore impairs reflection of light from the food surface, thereby reducing glossiness. Similar effects were observed upon suspension of spray-dried MFC particles (especially in soups), although these effects of MFC particles on appearance were not quantified using rank-rating (**Chapter 4**). It was furthermore speculated that the insoluble fibrils and fibril aggregates explain the reduced smoothness and increased pulpiness of MFC-containing (model) foods (**Chapter 2, 3**).

Flavour intensities of the studied (model) foods were suppressed at higher MFC concentrations (*i.e.* sweetness and strawberry flavour in **Chapter 2**, sourness and lemon flavour in **Chapter 3**). In literature several mechanisms have been proposed to be responsible for this effect of thickener concentration on perceived flavour intensity, including a physical or chemical interaction between flavour molecules and the thickening polymer (Pangborn & Szczesniak, 1974; Pangborn *et al.*, 1973), or a decrease in the diffusion rate of flavour molecules due to the food matrix' higher viscosity (Baines & Morris, 1987; Kokini *et al.*, 1982). Several studies demonstrated that the release of flavour molecules in the breath is not reduced at higher thickener concentrations (Cook *et al.*, 2003; Hollowood *et al.*, 2002; Linforth & Taylor, 2000). This refutes the hypothesis that thickening agents bind flavour molecules and supports the latter hypothesis that higher viscosity impedes the transport of taste and aroma molecules to receptors in the nose and mouth. Alternatively, an interaction at the neurological or perceptual level has been speculated to be responsible for the decreased flavour intensity at higher thickener concentrations. The presence of the tactile sensory stimulus of viscosity may moderate the perception of taste and flavour intensity through cross-modal interactions (Bult *et al.*, 2007; Cook *et al.*, 2003; Visschers *et al.*, 2006). The presence of MFC induced a cardboard-like or paper-like off-flavour in liquid and semi-solid model foods (**Chapter 2**, **Chapter 4** (data not shown)). This off-flavour is likely to be an inherent quality of the fibrous material, since slightly bitter and astringent off-flavours have been reported (Manninen *et al.*, 2021), including in orange fibre-enriched yoghurts (Kieserling *et al.*, 2019) and in bread fortified with citrus fibre (Spina *et al.*, 2019). The fact that only weak off-flavours were reported suggests that these can prospectively be masked by the food matrix itself or by addition of flavourings. This is consistent with previous findings in which incorporation of MFC did not significantly affect the flavour profile of ice cream (Yano *et al.*, 2016), mayonnaise (Golchoobi *et al.*, 2016) and hamburgers (Ström *et al.*, 2013). Likewise, no cardboard-like off-flavour was reported in low-fat lemon-flavoured mayonnaises thickened with MFC (**Chapter 3**). Although the off-flavour induced by MFC appears to be masked by the food matrix, food manufacturers are recommended to evaluate the effect of MFC and processing on potential off-flavour formation in foods.

6.1.2. Comparison of MFC with other commonly used thickeners

The results in **Chapter 2, 3 and 4** suggest that MFC, either as dispersed fibrils or as spray-dried particles, can be used as natural low-calorie, biodegradable, clean label thickener in liquid and semi-solid foods. This raises the question how MFC compares to other thickeners that are conventionally used in commercial food products, such as xanthan gum, (modified) starch and other nanocelluloses.

Comparison between MFC and xanthan gum

The use of MFC as a thickener was compared to xanthan gum in liquid and semi-solid foods in **Chapter 2 and 3**, respectively. Xanthan gum is a water soluble, pseudoplastic polymer consisting of linear glucose molecule chains with trisaccharide side chains (Sworn, 2021). Its backbone is relatively comparable to cellulose, as both consist of β-(1→4)-linked D-glucose units. Xanthan gum is produced and secreted by the bacterium *Xanthomonas campestris* through fermentation and can therefore be considered a natural thickener. Consumer perception of 'naturalness' on the other hand is not necessarily similar to its scientific interpretation, and the fact that xanthan gum is often listed with its E-number (E-415) on the product composition label may affect how consumers perceive its 'naturalness' (Evans *et al.*, 2010). Xanthan gum is used as thickener in many commercial products including dairy products, dressings, sauces, gravies, frozen foods, dry mixes and even in dysphagia management (Sworn, 2021). Results from this thesis suggest that MFC is capable of thickening liquid and semi-solid foods similar to xanthan gum at lower polymer concentrations as the relation between MFC concentration and shear viscosity conforms to a power law function (Figure 6.7), which is in accordance with results reported by (Lowys *et al.*, 2001). At similar shear viscosity, however, xanthan gum solutions exhibited larger yield stress compared to MFC dispersions. Less stress or force is required to make MFC dispersions flow which may be beneficial for food manufacturers, as this may prevent clogging and other problems related to transportation through pipelines in the food production process. The presence of a yield stress on the other hand improves spreadability and coating properties (*i.e.* glazing) and prevents particle settling in dressings or draining of sauces from bottles. The different rheological properties of MFC compared to xanthan gum are reflected in different sensory (texture) properties. One of the main outcomes of **Chapter 2 and 3** is that xanthan gum is significantly slimier and stickier than MFC, which is in line with earlier findings (Farpour *et al.*, 2021; Gössinger *et al.*, 2018; Gujral *et al.*, 2002). These texture attributes are generally regarded as undesirable (Pellegrino & Luckett, 2020) and results from **Chapter 2** and He *et al.* (2016) suggest that these attributes are associated with high extensional viscosity. Interestingly, both xanthan gum and MFC were perceived as slightly pulpy, albeit in different food matrices. MFC was perceived as pulpy when dispersed in aqueous model foods, which is presumably caused by the presence of insoluble (aggregated) fibrils. Pulpiness was reported in **Chapter 3** for low-fat mayonnaises thickened with xanthan gum, although to the authors' knowledge xanthan gum has not been described as pulpy before. This could be due to the relatively high xanthan gum content in low-fat mayonnaise (1.6-2.0 wt%), while concentrations between 0.1-0.5 wt% are commonly used in foods. **Chapter 2 and 3** furthermore demonstrated that xanthan gum affected the appearance of aqueous model foods to a lower extent than MFC. Differences and similarities regarding the sensory properties of liquid and semi-solid foods thickened with MFC and xanthan gum are visualised in Figure 6.8.

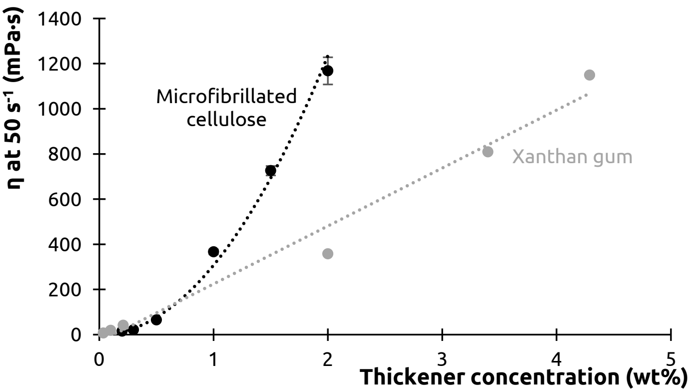

Figure 6.7. *Effect of thickener concentration on shear viscosity η at 50 s^{-1} in aqueous model foods (mean ± standard deviation of duplicate measurements). Black = microfibrillated cellulose (MFC), grey = xanthan gum. Trendlines are displayed to guide the eye.*

Comparison between MFC and starch

Starch is one of the most frequently used thickeners due to its large abundance and its large array of functionalities in foods. Although cellulose and starch both consist of glucose units, the α-(1→4) bonds in starch make this polysaccharide readily digestible in the human gastro-intestinal tract. As native starch has low temperature and pH resistance, physical and chemical modifications are frequently performed to improve its functional properties (Chen *et al.*, 2018). In **Chapter 3** MFC was compared to both native (non-modified) waxy corn starch (WCS) and chemically modified (cross-linked) corn starch (MS). Low-fat mayonnaises thickened with MFC, WCS or MS all show non-Newtonian shear-thinning behaviour and display comparable values for flow index *n*. Addition of MFC resulted in higher storage and loss moduli (*G'*, *G"*) compared to both starches, implying that a stronger network is formed by the cellulose microfibrils. In terms of sensory properties, the use of MFC and native and modified starches results in low-fat mayonnaises with smooth, creamy and melting texture. Sensory texture properties of mayonnaises thickened with MFC displayed strong similarities to those thickened with either chemically modified or native starch (Figure 6.8). The data imply that similar texture properties can be obtained with fewer calories (starch 4 kcal/g; MFC 0 kcal/g) and a clean label when MFC would be used as thickener instead of starches. Whereas starches are often declared on the product label with their respective E-numbers, MFC prepared from citrus fruits can for instance be labelled as 'citrus fibre'. Another advantage of using MFC is the improved temperature stability compared to (non-modified) starch (Agoda-Tandjawa *et al.*, 2010; Lowys *et al.*, 2001). In terms of flavour, results from **Chapter 3** and previous studies demonstrate bitter, metallic, cooked and astringent off-flavours for various starch-thickened foods (Lotong *et al.*, 2003; Matta *et al.*, 2006; Stahlman *et al.*, 2001). It is worth noting that MFC did not impart an off-flavour in lemon-flavoured mayonnaises, whereas an off-flavour was detected in the same mayonnaises thickened with native starch (**Chapter 3**). Although cellulosic material may impart a slight off-flavour, it appears that this is masked by the food matrix whereas this is not the case for starch. All in all, MFC exhibits promising characteristics to be used as a low-calorie, clean label substitute of starch.

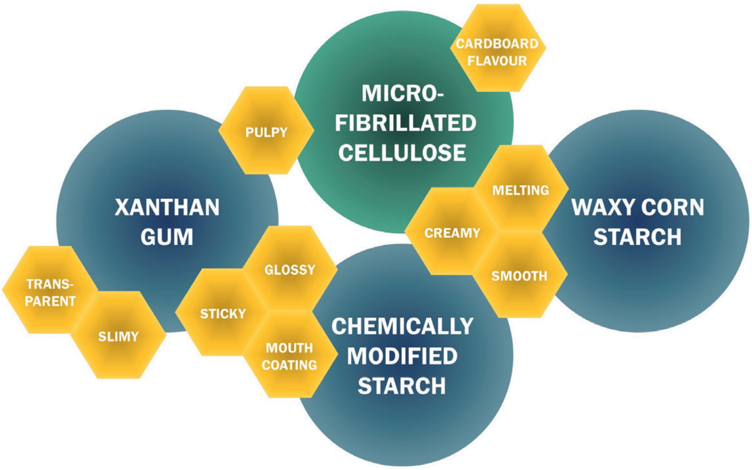

Figure 6.8. *Visual representation of sensory properties of the thickening agents studied in this thesis.*

Comparison between MFC and other nanocelluloses

Untreated native cellulose is generally not used in food products due to its incompatibility with water. To improve its functionality numerous cellulose derivatives have been developed, such as MFC. Some other well-known cellulosics include cellulose nanocrystals (CNC, also called cellulose whiskers), microcrystalline cellulose (MCC), carboxymethyl cellulose (CMC), methyl cellulose (MC), hydroxypropyl cellulose (HPC) and hydroxypropyl methyl cellulose (HPMC). Preparation of several cellulose derivatives involves hydrolysis and yields smaller molecules, including cellulose nanocrystals and microcrystalline cellulose. These molecules are less effective in network formation due to the reduction in molecular weight (*e.g.* MCC typically has a DP < 400 (Wüstenberg, 2014)). Networks formed by MFC are considerably stronger than for instance microcrystalline cellulose networks and therefore the effect of MFC on rheological properties is larger. Compared to MFC, higher concentrations of cellulose nanocrystals or microcrystalline cellulose are required to acquire the same increase in viscosity. Chemical modification through esterification or etherification yields intact cellulose molecules with vastly improved water solubility, including carboxymethyl cellulose, methyl cellulose, HPC and HPMC. Carboxymethyl cellulose is widely used in foods due to its viscosifying effect (Wüstenberg, 2014), but has been reported to be highly sticky, mouthcoating and difficult to swallow (Ong *et al.*, 2018a; Ross *et al.*, 2019). An additional disadvantage of these types of cellulose derivatives is that these are not considered clean label additives due to the chemical modification performed. MFC on the other hand provides essential functional properties, yet only requires physical modification. It can therefore be specified as ‘citrus fibre’ in food labelling, which enables its use in clean label foods.

6.1.3. Correlating sensory perception to instrumental properties

Throughout this thesis, results from rheology and tribology experiments have been linked to sensory properties of (model) foods to explore possible correlations between instrumental and sensory parameters (Figure 6.1). Such correlations could theoretically be used to predict from instrumental data how specific foods are perceived sensorially. This would evidently reduce the financial costs associated with sensory evaluation procedures and could accelerate product development. For decades, researchers have therefore attempted to develop models to predict sensory characteristics of foods based on instrumental analyses. So far, however, most sensory (texture) properties cannot be predicted properly from instrumental measurements.

Thickness

Although a complete understanding of any of the sensory texture attributes is lacking, the texture attribute that is currently best understood is thickness. Research on predicting perception of thickness in the mouth from instrumental parameters traces back more than 50 years. Wood (1968), being the first to study this topic, concluded that oral evaluation of thickness of liquids occurs at a shear rate of approximately 50 s^{-1}. Despite the fact that most subsequent studies agree that the stimulus related to oral perception of thickness is the shear stress applied to the tongue (Christensen, 1979; Cutler *et al.*, 1983; Dickie & Kokini, 1983; Kokini, 1987; Kokini *et al.*, 1977; Stanley & Taylor, 1993), debate has been ongoing about which specific shear rate is most relevant for thickness perception. According to Shama and Sherman (1973) the shear rate associated with evaluation of oral thickness depends on the specific food and its flow characteristics, and therefore covers a range of shear rates extending from 10 to 1000 s^{-1}. The majority of the studies modelling sensory thickness from rheological parameters used relatively simple models that do not fully represent the complex processes that foods undergo during oral processing (Dickie & Kokini, 1983; Janssen *et al.*, 2007; Kokini, 1987; Kokini *et al.*, 1977). The generally accepted model of Kokini *et al.* (1977) was recently adapted and extended by Deblais *et al.* (2021), who included a parameter to account for dynamic squeezing flow occurring when orally processing liquid foods instead of approximating flow by static squeezing. According to this model the relation between sensory thickness and calculated shear stress follows the Weber-Fechner law, which describes a logarithmic relation between the strength of the stimulus and its perceived intensity (Figure 6.9). This is not very surprising taking into account that the Weber-Fechner law is known to apply to several other senses, including sight and hearing (Gescheider, 2013). Indeed, others have demonstrated that oral thickness perception follows the Weber-Fechner law (Camacho *et al.*, 2015; Terpstra *et al.*, 2005). Likewise, a logarithmic relation is observed between MFC concentration and perceived thickness and creaminess of aqueous model foods (Figure 6.10).

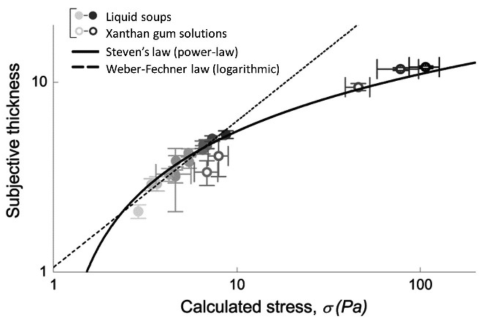

Figure 6.9. *Power law dependence between subjective thickness and calculated shear stress on the tongue. Adopted from Deblais et al. (2021).*

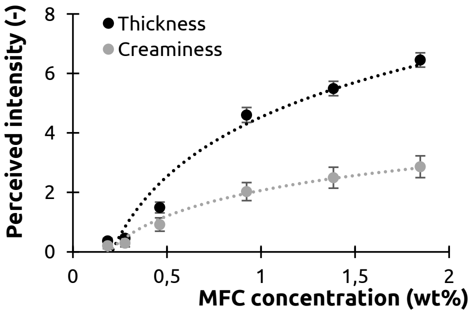

Figure 6.10. *Relation between MFC concentration in aqueous model foods (**Chapter 2**) and perceived thickness (black) and creaminess (grey).*

Creaminess

The sensory texture attribute that has received most attention in the scientific world is creaminess, due to the fact that it is generally strongly liked (Ares *et al.*, 2010; Richardson-Harman *et al.*, 2000). Numerous research and review papers have been written trying to understand creaminess (e.g. Bom Frøst & Janhøj, 2007; de Wijk *et al.*, 2006; Dickinson, 2018; Upadhyay *et al.*, 2020), but so far no model exists that can accurately predict creaminess for a variety of foods. General consensus prevails about the fact that creaminess is a multi-modal sensory attribute, meaning that multiple senses are involved in the perception of creaminess (Chen & Eaton, 2012). More specifically the level of creaminess of foods is determined by a combination of gustatory, olfactory and tactile cues. Although flavour has been found to be an important factor contributing to the creaminess of foods (e.g. Jervis *et al.*, 2014), throughout this thesis the focus has been on the tactile perception of creamy mouthfeel. One of the most important factors contributing to creaminess is the food's viscosity, as more viscous foods are generally perceived as creamier (e.g. Akhtar *et al.*, 2006; Akhtar *et al.*, 2005; Daget & Joerg, 1991; Janhøj *et al.*, 2008; van Aken *et al.*, 2011). Since viscosity only reflects the material's bulk properties and cannot fully predict creaminess, friction properties have been postulated to play a role in creaminess as these represent surface properties of the food (de Wijk *et al.*, 2006; Dickinson, 2018; Kokini, 1987; Kokini & Cussler, 1983; Upadhyay *et al.*, 2020). Indeed several researchers found a negative correlation between creaminess and friction properties, suggesting that improved lubrication (*i.e.* lower friction) enhances creaminess (Chojnicka-Paszun *et al.*, 2012; Laiho *et al.*, 2017; Sonne *et al.*, 2014). Fat is hypothesised to play a major role in providing lubrication-assisted creaminess, since coalescence of fat on the tongue during oral processing facilitates the formation of a lubricating oil film that provides creaminess (Dresselhuis *et al.*, 2008). Since viscosity has an effect on friction properties, it was tried to disentangle the effects of viscosity and friction properties on creaminess in **Chapter 5**. The results of this chapter imply that viscosity is the main contributor to creaminess of liquid foods. Laguna *et al.* (2017) on the other hand demonstrated that discrimination between full-fat and fat-free dairy products was often due to differences in creaminess, and was better predicted by the material's friction properties than their rheology. It should be noted that friction and lubrication are system properties instead of intrinsic characteristics of the food (Sarkar & Krop, 2019). This means that friction properties strongly depend on the contact surfaces and experimental conditions used, and that establishing universally applicable correlations may be challenging. Correlations between viscosity, friction properties and creaminess may furthermore depend on the type of food studied, and friction properties may have a larger effect on certain foods compared to others. Creaminess is for instance easier discriminated in liquid foods compared to solid foods, suggesting that the food's physical state (*i.e.* liquid, semi-solid or solid) affects its perceived creaminess (Drewnowski *et al.*, 1989). The foods studied in **Chapter 5** were low viscous liquids with low fat content, which may have affected the general perception of creaminess of these foods. The multi-modal nature of creaminess and the poor generalisability of models from one food to another make it difficult to determine the relative contribution of factors involved in creaminess perception.

Other sensory texture attributes

While thickness and creaminess have so far received most scientific attention, correlations between other sensory texture attributes and rheological (recently reviewed in Joyner, 2018) or tribological properties (see Sarkar & Krop, 2019 for a comprehensive review) have been reported as well. The pioneering work of Kokini and colleagues (Kokini, 1987; Kokini & Cussler, 1983; Kokini *et al.*, 1977) for instance describes how smoothness and slipperiness can be predicted from rheological and tribological measurements by determining viscous and friction forces on the tongue. Figure 6.1 summarises the correlations between instrumental analyses and sensory perception that were identified in this thesis. It should be noted that the validity of these correlations should be tested in other foods before these can be generalised to other biopolymers and food types. Establishing universal relations between sensory properties and instrumental analyses that are valid for a broad range of foods is challenging, as the type of food influences oral processing and consequently the way in which foods are perceived. Kokini and colleagues proposed a black-box model in which all sensory texture attributes can be predicted from sensory thickness, smoothness and slipperiness scores (Kokini, 1987; Kokini *et al.*, 1977), illustrating the high level of interdependence of sensory texture attributes. This further clarifies why elucidation of the mechanisms behind oral texture perception is complex, as many texture attributes and underlying physical properties are correlated with one another.

6.2. Methodological considerations

6.2.1. Preparation of MFC and sustainability

One of the advantages of MFC is the fact that a large variety of plant sources can be used for its production, including by-products and waste from for example fruit or vegetable cultivation. As such, MFC can be considered a sustainable food ingredient that can aid in valorisation of agricultural waste streams. While the microfibrillated material studied throughout this thesis was prepared from citrus fibre many other vegetal sources could be considered for production of MFC, such as sugar beet leaves and corn waste. Nevertheless the sustainability of MFC is disputed due to the large amount of energy needed for its production (Lavoine *et al.*, 2012). Although the preparation process of MFC from primary cell wall material from fruits or vegetables requires less energy compared to MFC from wood pulp (Lavoine *et al.*, 2012; Nechyporchuk *et al.*, 2016), energy demands can reach up to 30,000 kWh/tonne (Siró & Plackett, 2010). The energy consumed during the production of MFC is 100 times higher than that of native starch, which is estimated to be around 200-400 kWh/tonne (Assawamartbunlue & Luknongbu, 2020). The degree of fibrillation of cellulose fibres and consequently the functional properties of MFC depend on the number of homogenisation passes and the applied pressure. Food manufacturers should therefore take into consideration whether the benefits of using MFC outweigh the amount of energy consumed during its production. Valorisation of waste materials is desirable and contributes to improving the sustainability of food supply chains, however the overall sustainability of MFC can be questioned as a result of the high energy input during production. Possibilities to reduce energy consumption include the use of pre-treatments, different types of starting material or other types of fibrillation equipment. Finally, *in situ* preparation of MFC can provide a way to save energy, in particular for foods that require homogenisation for preparation such as mayonnaise.

6.2.2. Rheology

Although shear rheology is assessed by the majority of studies on MFC, few include extensional viscosity. In this thesis, extensional rheology was performed on aqueous model foods thickened with MFC (**Chapter 2**). Since extensional viscosity is known to depend on the type of thickener used (Hadde & Chen, 2019), extensional rheological properties of MFC and xanthan gum were examined. Extensional viscosity has been related to duration and ease of swallowing and knowledge on this parameter could therefore be crucial in dysphagia management (Hadde & Chen, 2019; Hadde *et al.*, 2019; Mackley *et al.*, 2013; Theocharidou *et al.*, 2021). Foods with higher extensional viscosity are more cohesive and will be more resistant to bolus stretch during elongation upon swallowing the food. This reduces the risk of bolus disintegration after swallowing and thus the risk of aspiration or asphyxiation. **Chapter 2** demonstrates that xanthan gum solutions had higher extensional viscosity at high shear rates compared to iso-viscous MFC dispersions. MFC's lower extensional viscosity compared to other hydrocolloids is expected to be related to its water insolubility, since the presence of fibrous particles makes the bolus more prone to break-up. Although higher extensional viscosity and bolus cohesiveness can improve safe swallowing of low viscous fluids, it may not be preferred for high viscous foods. In fact it could induce swallowing difficulty, as foods highly resistant to stretching deformation require more muscle strength to be swallowed. High extensional viscosity may moreover evoke generally unfavourable sensations of mouthcoating, slimy and sticky texture (Chapter 2, He *et al.*, 2016). Future studies should further explore how and which sensory properties are correlated with extensional rheology parameters, as relatively few studies have focused on this.

6.2.3. Tribology

Based on the tribological results in this thesis, determining lubricating properties of liquid foods thickened with MFC was challenging using the present tribological set-up. It is speculated that the entangled networks formed by dispersed MFC fibrils were excluded from the tribometer's contact zone due to their large hydrodynamic volume. The continuous phase of the sample was presumptively measured instead, which resulted in samples that contained varying amounts of MFC all displaying the same friction curves. Spray-dried MFC particles suspended in milks and soups yielded comparable results. High shear rates in the tribological gap probably disrupted the spherical structure of MFC particles and promoted the formation of aggregated MFC networks of a size exceeding the height of the tribological contact zone. An attempt was made to enlarge the contact zone by decreasing the normal force (F_N) applied to the samples. As this yielded higher rather than lower friction coefficients, it was concluded that the intended increase in gap size was still not sufficient to entrain the microfibrillated material. Other researchers were able to determine friction properties of MFC dispersions using a ball-on-plate tribometer employing reciprocal motion (Kinoshita *et al.*, 2020). These authors tested several sliding balls from several materials, which were moved against a stainless steel or polyoxymethylene plate. The choice of surface material, the device used and the type of movement may explain why Kinoshita and colleagues were able to determine friction properties of MFC while the material was excluded from the contact zone in our experiments. The choice of tribological set-up used in this thesis might not be optimal for this type of material and using another set-up might enable the assessment of friction properties of MFC dispersions. The use of larger surface areas or increasing the surface roughness could improve assessment of lubrication by dispersed MFC. The relatively small contact area of the ball-on-three pins set-up may be the cause of exclusion of MFC from the contact zone, and tribometers with larger surface areas such as the Mini Traction Machine by PCS Instruments or the Bruker UMT TriboLab might allow entrainment of MFC (Rudge *et al.*, 2019). Rougher surfaces on the other hand better represent the surface of the human tongue and might facilitate inclusion of MFC between the tribological surfaces. The larger asperities resulting from increased roughness yield larger voids that could potentially enclose MFC.

6.2.4. Sensory evaluation

As previous studies mainly focused on the functional properties of MFC, the primary aspect of this thesis was to elucidate the effect of MFC on sensory properties of foods. Considering the limited knowledge on sensory properties of MFC, the Rate-All-That-Apply (RATA) method was selected for sensory evaluation of MFC-thickened (model) foods. This method makes use of an extensive list of attributes of which panellists select and rate only those attributes they consider applicable to the food sample (Ares *et al.*, 2014). This method accommodates determination of the sensory attributes relevant to MFC-containing foods, while preventing dumping effects or research bias. RATA is suitable to be used by naive consumers and does not require prior training of the panellists. On the other hand, rank-rating was performed in **Chapter 4 and 5** as the focus in those studies was on specific sensory attributes rather than development of a sensory profile on MFC-thickened foods. Using this method panellists evaluate the intensity of an attribute by direct comparison of all samples, which accommodates the detection of smaller sensory differences (Cleaver, 2018; Kim & O'Mahony, 1998).

Sensory evaluation was performed by non-trained subjects throughout this thesis. Sensory assessment by trained panels may be preferred in case high panel agreement is required and small sensory differences should be detected. It is questionable whether results from trained panels represent the way consumers sensorially perceive foods. Since this thesis aimed to assess the suitability of MFC as a texture modifier in foods, sensory evaluation by naive consumers was considered more relevant. The consumer panels were able to distinguish relatively small differences in viscosity (**Chapters 2-5**), which demonstrates that such panels yield reliable results. These results reflect those of Oppermann *et al.* (2017), who concluded that untrained panellists had similar discriminative ability using RATA compared to Descriptive Analysis by a trained panel.

6.3. Future research

6.3.1. Use of MFC as texture modifier in foods

A first step for future research would be to extend the number and type of foods in which MFC is applied and studied. The work in this thesis has mainly focused on liquids (aqueous model foods, milk, soup) and only includes a limited selection of semi-solids (aqueous model foods, low-fat mayonnaises). It is recommended to validate the results of this thesis in other foods such as other (low-fat) sauces or dairy products (*e.g.* yoghurt, custard, pudding) to establish which sensory attributes characterise MFC and to allow generalisation of the results. Special attention should be given to (masking) the cardboard-like flavour that was identified in **Chapter 2**, as its intensity may depend on the type of food and the presence of other flavours. The use of MFC might be particularly interesting for development of low-calorie sauces and dressings, considering the number of products in this category that contain thickening agents. Its vegetal origin makes MFC highly suitable for use in vegan food products, including plant-based dairy substitutes.

Future research could furthermore focus on the gel-forming capacity of MFC and its suitability as thickening or gelling agent in solid foods could be explored. Concentrations above 2.0 wt% MFC should be used to produce gels but such high concentrations may elicit problems during microfibrillation, including clogging of the homogenisation equipment. Higher MFC concentrations could alternatively be achieved by water removal from dilute dispersions, for instance by evaporation or freeze-drying. It is worth noting that hornification should be monitored in such processes, as this is one of the challenges associated with high MFC concentrations. MFC could furthermore be used as a texture modifier in solid foods, including (vegan) meat analogues. Preliminary results from pilot experiments suggest that MFC can be used as a binder or texture modifier in plant-based burgers, while potentially enhancing their juiciness due to improved water holding capacity.

6.3.2. Composition of spray-dried MFC particles

Chapter 4 demonstrates that the effect of MFC particles on rheological and sensory properties of foods depends on the MFC:maltodextrin of the particles. Shear viscosity, perceived thickness and perceived creaminess of liquids containing 0.3 wt% MFC emerging from high MFC:maltodextrin ratio particles were similar to those containing 0.15% MFC from low MFC:maltodextrin ratio particles. A 50% reduction in MFC content can thus be achieved by lowering the ratio between MFC and maltodextrin, while preserving desirable physico-chemical and sensory properties of the resulting food. This implies that the texture-modifying properties of spray-dried MFC particles can be tuned by the ratio of MFC to maltodextrin. An increase in the relative amount of material preventing hornification (in this case maltodextrin) reduces the number of entanglements between microfibrils that form the network of the particles, resulting in the formation of less rigid particles. Because the particles contain lower quantities of microfibrillated material, at constant MFC content larger quantities of these particles can be obtained compared to high MFC:maltodextrin ratio particles. The most effective increase in rheological and sensory properties is thus obtained at a lower ratio between network-forming and hornification-preventing material (*i.e.* MFC and maltodextrin). Yet one can envision that at a certain point the ratio becomes too low for MFC to form an entangled network that stays intact upon dispersion in water, and thus for proper particles to be formed. Future research should therefore focus on determining the optimum MFC:maltodextrin ratio.

Another aspect that should be considered is the type of material used to improve dispersibility of the powder, as maltodextrin is a calorie-providing sugar. The question arises whether MFC particles containing >85% maltodextrin are suitable as fat replacer in low-fat foods. The high maltodextrin content contributes to a higher energy density of the final product, as maltodextrin provides 4 kcal/g. The use of maltodextrin to prevent aggregation of microfibrils thus compromises the intended energy reduction by the fat reduction that follows from MFC being a low caloric dietary fibre. The possibilities of using other materials, such as soluble fibres, gums, starch or salt should be further explored. Although drying MFC facilitates dispersion, storage and transport of the material and extends its shelf life, the dispersibility of MFC particles could be further optimised. The current powder forms a viscous gel upon contact with water, which impedes further diffusion of water and rapid dispersion of the powder in liquids. Improving the dispersibility of the powder could create potential for its use as a thickener or creamer by consumers, for example in diets for dysphagia patients.

6.3.3. Other applications of MFC

The results presented in this thesis suggest that MFC can be used as texture modifier in foods, yet this versatile material may have other applications that can be studied. The emulsifying and suspension stabilising properties of MFC have been reported elsewhere (e.g. Lu *et al.*, 2019; Winuprasith & Suphantharika, 2013; 2015) and have been attributed to microfibrils behaving as Pickering particles. Confocal images in **Chapter 3** also show the presence of cellulosic material at the oil-water interface. These functional properties should be verified in a wider range of foods, preferably in studies that include the effect of the emulsifier on sensory properties. Preliminary results furthermore revealed that MFC has excellent foaming properties, as was observed from the highly stable foam that was formed upon high-shear mixing of skimmed milk with MFC particles (**Chapter 4**). Such foaming properties could be favourable in dairy products such as (chilled) iced coffees, or even in plant-based dairy substitutes. Apart from its applications in food, MFC can be used to increase the strength of nanocomposites (Siró & Plackett, 2010; Zimmermann *et al.*, 2004). Such nanocomposites could be of use in packaging materials, including food packaging. Several high-tech functional applications for MFC have been summarised by Heise *et al.* (2021), including its use in shape-memory or self-healing materials or photosynthetic cell factories.

6.3.4. Effect of MFC on satiety, energy intake and lipid digestion

Although only a small range of liquid and semi-solid (model) foods was studied, the results from the current thesis suggest that foods with favourable rheological and sensory properties can be prepared when MFC is used as a texture modifier. Addition of polymers such as MFC enhances foods' viscosity, which consequently lowers the eating rate of these foods and extends oral processing time (Bolhuis & Forde, 2020). Oro-sensory exposure time is known to induce feelings of satiety (de Graaf, 2012), and as such MFC can have an effect on energy intake. In addition, MFC can be used as a fat replacer to compensate for the loss in viscosity as a result of fat reduction in low-fat foods. In this way MFC can have a positive effect on reducing the energy density of foods, especially since it is composed of insoluble fibre and thus provides fewer calories than several common thickening agents. Being a dietary fibre, MFC could moreover hypothetically have a positive effect on satiety and reduce appetite and long-term energy intake (Wanders *et al.*, 2011). Even though MFC will typically only constitute a small component of food as a texture modifier, its effect on satiety and energy intake could be examined in studies assessing *ad libitum* intake.

MFC has furthermore been reported to impair lipid digestion, resulting in fat being excreted instead of being absorbed by the human body. This reduction in the amount of fat that is absorbed from fat-rich foods could potentially facilitate weight loss management. Studies using *in vitro* digestion have shown that the rate of lipid digestion and total lipid uptake can be reduced by the presence of MFC in oil-in-water emulsions (DeLoid *et al.*, 2018; Liu & Kong, 2019a; Winuprasith *et al.*, 2018). Three mechanisms for this reduction in intestinal lipid digestion have been proposed: (i) formation of a complex polymer network that acts as a physical barrier for lipase (Liu *et al.*, 2019; Liu & Kong, 2019a, 2019b; Winuprasith *et al.*, 2018), (ii) coalescence of oil droplets (DeLoid *et al.*, 2018), and (iii) binding of bile salts by MFC (DeLoid *et al.*, 2018; Winuprasith *et al.*, 2018). As shown throughout this thesis, at sufficiently high concentrations of MFC an entangled polymer network is formed with excellent water holding capacity. A higher viscosity is consequently obtained, which lowers the diffusion rates and therefore the activity of lipase. The second mechanism involves MFC-induced oil droplet coalescence or flocculation through bridging mechanisms. The surface area of the oil droplet that is accessible for lipase is reduced and the lipid digestion rate decreases. Thirdly, MFC has been found to interact with bile salts, which lowers the availability of bile salts and impairs lipid digestion (DeLoid *et al.*, 2018). Since the speculated effect of MFC on fat digestion has thus far only been studied *in vitro*, future research should focus on establishing this effect *in vivo*.

6.4. Main conclusions

Ever increasing concerns about climate change and the obesity pandemic cause food manufacturers and researchers to explore new sustainable and healthy food ingredients. The suitability of fibrous material from citrus peels as low-caloric clean label food thickener was the focus of the current thesis. Two types of MFC were studied (dispersed MFC fibrils and spray-dried MFC particles) and their effect on sensory, rheological, tribological and morphological properties was determined in several liquid and semi-solid (model) foods. These properties were compared to those of a selection of commonly used thickeners. Potential correlations between sensory and physico-chemical properties were explored in an attempt to elucidate mechanisms behind their perception.

The results presented imply that MFC prepared from citrus fibre can thicken liquid and semi-solid foods already at relatively low polymer concentration (0.2-2.0 wt%). The material provides rheological properties that make MFC highly suitable for use in foods. A wide range of shear viscosities can be obtained, the material exhibits a yield stress and its shear-thinning behaviour is similar to that of commonly used food thickeners. Foods thickened with MFC demonstrate sensory texture properties similar to foods thickened with chemically modified starch, a polymer that is frequently used in commercial (low-fat) products. Compared to xanthan gum, MFC can thicken foods in a similar way while avoiding sticky, slimy and mouthcoating texture. MFC can moreover be spray-dried in the presence of maltodextrin to improve its implementation in foods, without losing its ability to enhance viscosity, sensory thickness and creaminess. MFC thus shows potential as a low-caloric clean label food thickener, which can moreover be obtained from (agricultural) plant sources.

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References

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S

Summary

Summary

Growing global concerns about obesity and the environment have generated a demand for healthy and sustainable food products. Many foods nowadays have a low-fat counterpart, and attention of food manufacturers increasingly focuses on reducing the environmental impact of foods and ingredients. The use of waste materials offers a promising strategy to pursue a more circular economy. An example of a material that has shown potential as a functional ingredient in foods is microfibrillated cellulose (MFC), which can be prepared from agricultural waste materials. Mechanical treatment of primary cell walls, for example of citrus fruits, disintegrates the structure of native cellulose and yields a three-dimensional entangled polymer network that can form stable dispersions in water. In this thesis the potential of MFC as a texture modifier in liquid and semi-solid foods was studied, thereby focusing on its effect on rheological, tribological and sensory properties of the resulting food. In other words, this thesis aimed to '*unravel the unravelled'* by studying sensory and physico-chemical properties of MFC and thereby examine its suitability as a texture modifier. Sensory properties were furthermore correlated to instrumental properties to elucidate mechanisms underlying texture perception.

Chapter 2 studied MFC in simple aqueous dispersions and compared its effect on sensory, rheological and tribological properties to those of iso-viscous xanthan gum solutions. An increase in concentration of dispersed MFC fibrils caused an increase in shear viscosity, storage modulus G', loss modulus G'' and yield stress of the dispersion. Similar effects were observed for xanthan gum solutions, although these showed a linear relationship between concentration and yield stress as opposed to the power-law dependence observed for MFC. Xanthan gum solutions moreover exhibited higher yield stresses than MFC dispersions at similar shear viscosity. In addition to the effect of MFC concentration on rheological properties, the intensity of several sensory attributes increased at higher MFC concentration. These included thickness, creaminess, cardboard flavour and melting texture, whereas smoothness and transparency decreased with increasing MFC concentration. Glossiness and transparency of the aqueous dispersions declined at higher concentrations of MFC, which is presumably caused by the insolubility of MFC. Although mouthcoating, sliminess and stickiness intensities increased with MFC concentration, considerably higher intensities were reported for xanthan gum solutions. Perception of these generally unfavourable texture attributes was correlated with extensional viscosity at high shear rates. In terms of extensional rheology, MFC dispersions exhibited extensional shear thinning behaviour over a large range of deformation rates. Varying the MFC concentration did not affect friction properties of the dispersions under the chosen experimental tribological conditions. It was hypothesised that MFC

fibrils were excluded from the tribological gap, due to the hydrodynamic volume of the network formed by MFC. To summarise, MFC is capable of thickening liquids similar to xanthan gum while avoiding perception of some unfavourable texture attributes including sliminess, stickiness and mouthcoating.

In a continuation of **Chapter 2**, the suitability of MFC as a food thickener in fat-containing semi-solid foods was examined in **Chapter 3**. MFC was applied at three concentrations in low-fat mayonnaises and its effect on sensory, rheological and tribological properties was compared to mayonnaises thickened with varying concentrations of xanthan gum, native waxy corn starch and chemically modified corn starch. Rheological properties such as shear viscosity, storage modulus *G'*, loss modulus *G"* and yield stress increased with biopolymer concentration. Highest storage and loss moduli were observed for low-fat mayonnaises thickened with MFC, whereas these mayonnaises also exhibited lowest shear viscosities and yield stresses. Friction in the boundary regime primarily depended on the type of thickener and the ability to form a lubricating film, whereas viscosity of the mayonnaise was the predominant factor determining friction in the mixed regime. The microstructure of the low-fat mayonnaises was examined using CLSM and demonstrated that low-fat mayonnaises thickened with MFC or xanthan gum contained larger oil droplets than mayonnaises thickened with native waxy corn starch and chemically modified corn starch. Mayonnaises with MFC or xanthan gum also had a polydisperse droplet size distribution as opposed to the small and uniformly sized droplets observed in mayonnaises with native or chemically modified starch. The microscopy images furthermore revealed that MFC did not only form a viscosifying network in the continuous phase, but was also present at the oil-water interface where it is expected to stabilise the oil droplets. In terms of sensory properties, the type of thickener used affected all sensory attributes except sweetness and visual thickness. Irrespective of thickener type, increasing the biopolymer concentration generated higher intensities of thickness, mouthcoating and stickiness, whereas melting and smooth texture were reduced. Sticky texture was negatively correlated with friction in the boundary regime, which might be caused by adsorption of certain thickeners on the tribological surface. A negative correlation was found between slimy texture and flow index *n*, which suggests that more shear-thinning mayonnaises were perceived as slimier. Low-fat mayonnaises thickened with MFC had a less glossy and yellower appearance than mayonnaises thickened with xanthan gum, native starch or modified starch. Perception of sensory texture of MFC-thickened mayonnaises was highly similar to mayonnaises thickened with native or chemically modified starch, which implies that MFC can replace native and modified starch as a thickener in low-fat mayonnaises.

One of the main obstacles preventing widespread use of MFC in foods is the high water content of MFC dispersions (> 98%). This is (financially) disadvantageous in terms of transport and storage of the dispersions, and makes them susceptible to microbial spoilage. Preparation of dry MFC powders could potentially solve these issues, when irreversible aggregation occurring upon dehydration of MFC dispersions is avoided. This so-called hornification results in a loss of functional properties and should therefore be prevented, for example by dehydration in the presence of other compounds. **Chapter 4** showed that a mixture of citrus fibre and maltodextrin can be homogenised and spray-dried to obtain a powder consisting of spherical MFC-maltodextrin particles. While maltodextrin prevented aggregation of the microfibrils during dehydration, it dissolved in the liquid continuous phase upon suspension of the particles. MFC formed a mixture of spherical MFC networks and individual fibrils, and the former remained intact after suspension. The MFC particles enhanced the viscosity of skimmed milk and instant tomato soup, which in turn positively affected sensory thickness and creaminess. No effect of varying the MFC concentration on tribological properties was observed, which is presumably caused by exclusion of the particles from the contact zone. It was furthermore demonstrated that using a higher ratio of MFC:maltodextrin before spray-drying does not further enhance viscosity compared to lower MFC:maltodextrin ratios, which is attributed to the formation of fewer but denser networks. Using lower MFC:maltodextrin ratios to prepare dehydrated MFC can therefore be favourable, as these particles are more effective at increasing viscosity, thickness and creaminess. To summarise, spray-dried MFC powders can be prepared that function as texture modifiers (thickener and creamer) upon resuspension in liquid foods.

The results of **Chapter 2-4** suggest that MFC can potentially be used as a food thickener or even as a creamer. In **Chapter 5** an attempt was made to improve our understanding of the factors underlying creaminess. Multiple studies postulated that viscosity and friction are the two main physical properties contributing to creaminess perception. However, as viscosity itself has an effect on a material's friction properties, it remains unclear what the relative and independent contributions of viscosity and friction properties are on creaminess of foods. **Chapter 5** attempted to disentangle the effects of viscosity and friction on creaminess of iced coffees. Iced coffees were prepared with either low or high viscosity and friction coefficients by addition of maltodextrin to increase viscosity and polyethylene glycol (PEG) to reduce friction. Sensory evaluation of thickness, slipperiness and creaminess of the iced coffees was performed both orally and haptically (in hand). High viscous iced coffees were significantly more creamy, thick and slippery compared to low viscous iced coffees. Reducing the friction of low viscous iced coffees surprisingly resulted in lower creaminess and thickness, but enhanced slipperiness. Haptic evaluation of thickness was congruent with oral evaluation of thickness. Although increasing viscosity resulted in enhanced creaminess, reduced friction properties did not induce a similar effect. The results suggest that viscosity is the predominant factor in eliciting creaminess, while friction properties are of subordinate importance. **Chapter 5** revealed how challenging it is to disentangle the effect of viscosity from the effect of friction on creaminess in an experimental study. Further studies are needed to gain better understanding of the relative and independent contributions of rheological and friction properties of foods on creaminess perception.

In **Chapter 6** the results of the preceding chapters were integrated and discussed, and dispersed MFC fibrils were compared to spray-dried MFC particles. Increasing the concentration of MFC fibrils or particles caused higher values of shear viscosity and storage modulus G'. Viscosity and MFC concentration displayed a power law dependence for dispersed MFC fibrils whereas an exponential dependence was observed for MFC particles. For storage modulus G' on the other hand, power law relationships were established for dispersed MFC fibrils and spray-dried MFC particles. Methodological considerations such as the exclusion of MFC from the tribological contact zone and the sustainability of MFC are discussed in more detail in this chapter, as well as areas for future research on MFC. MFC has a limited caloric value since it is an insoluble dietary fibre, and can therefore be used to reduce the energy density of foods. Addition of MFC to foods yields sensory texture properties similar to those provided by commercially used xanthan gum and starches, while limiting mouthcoating, slimy and sticky mouthfeel. All in all, MFC showed favourable effects as a texture modifier in liquid and semi-solid foods at relatively low concentrations. It can be concluded that MFC can be used as an efficient clean label texture modifier in liquid and semi-solid foods.

A

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About the author

About the author

Annelies Emma Blok was born on August 31st, 1992 in Borger, the Netherlands. She started a BSc in Nutrition & Health at Wageningen University in 2010. During her BSc thesis, she wrote a literature review on the effect of food odour on food intake. After obtaining her BSc degree in 2013, Annelies continued her studies at Wageningen University with a MSc specialised in Sensory Science, which was performed partly at the University of Copenhagen. As a result of the courses taken during this study programme, her interests in food science and technology grew and she decided to pursue a second MSc programme in Food Technology. For this programme she wrote a thesis at the laboratory of Food Chemistry on the binding affinity of phlorotannins in seaweed for β-casein. Her second MSc thesis was written at the department of Nutrition and Health and focused on the role of odour, taste, flavour and texture in the recognition of vegetables. She performed a 6-month internship at Arla Foods Nijkerk, the Netherlands, where she examined the influence of composition and process parameters on the flavour profile of yoghurt. Annelies graduated from both MSc programmes in May 2017. After completing her studies she continued to work at Arla Foods before starting as a PhD candidate at the Food Quality & Design group at Wageningen University in 2018. Her PhD project focused on the effect of microfibrillated cellulose on sensory, rheological and tribological properties of liquid and semi-solid foods. Her work, as presented in this thesis, was part of the project 'BioMolecule2Food'.

Annelies can be contacted by email at *anneliesblok@live.nl*.

A

List of publications

This thesis

Annelies E. Blok, Dieuwerke P. Bolhuis, Heleen V. Kibbelaar, Daniel Bonn, Krassimir P. Velikov & Markus Stieger (2021). *Comparing rheological, tribological and sensory properties of microfibrillated cellulose dispersions and xanthan gum solutions*. Food Hydrocolloids, 121, 107052.

Annelies E. Blok, Dieuwerke P. Bolhuis & Markus Stieger (2020). *Contributions of viscosity and friction properties to oral and haptic texture perception of iced coffees*. Food & Function, 11(7), 6446-6457.

Others

Antoine Deblais, Elyn den Hollander, Claire Boucon, **Annelies E. Blok**, Bastiaan Veltkamp, Panayiotis Voudouris, Peter Versluis, Hyun-Jung Kim, Michel Mellema, Markus Stieger, Daniel Bonn & Krassimir P. Velikov (2021). *Predicting thickness perception of liquid food products from their non-Newtonian rheology*. Nature Communications, 12, 6328.

Vera L. van Stokkom, **Annelies E. Blok**, Olaf van Kooten, Cees de Graaf & Markus Stieger (2018). *The role of smell, taste, flavour and texture cues in the identification of vegetables*. Appetite, 121, 69-76.

Anne M. Vissers, **Annelies E. Blok**, Adrie H. Westphal, Wouter H. Hendriks, Harry Gruppen & Jean-Paul Vincken (2017). *Resolubilization of protein from water-insoluble phlorotannin–protein complexes upon acidification*. Journal of Agricultural and Food Chemistry, 65(44), 9595-9602.

Overview of completed training activities

Discipline specific activities		* Poster presentation ** Oral presentation
2021	6th International Conference on Food Oral Processing *	Valencia, Spain *[online]*
	4th Food Structure & Functionality Symposium *	Cork, Ireland *[online]*
	Course Healthy Food Design (VLAG)	Wageningen, the Netherlands
2020	34th EFFoST International Conference **	Tel Aviv, Israel *[online]*
	9th European Conference on Sensory and Consumer Research (EuroSense) *	Rotterdam, the Netherlands *[online]*
2019	13th Pangborn Sensory Science Symposium *	Edinburgh, United Kingdom
	33th EFFoST International Conference **	Rotterdam, the Netherlands
	Course Chemometrics (VLAG)	Wageningen, the Netherlands
2018	5th International Conference on Food Oral Processing	Nottingham, United Kingdom
	Course Sensory Perception & Food Preference: The role of context (VLAG)	Wageningen, the Netherlands
	Course Rheology: The do's and don'ts (VLAG)	Wageningen, the Netherlands

General courses		
2021	Adobe InDesign (WGS)	Wageningen, the Netherlands
	Career Assessment (WGS)	Wageningen, the Netherlands
2020	Scientific Artwork, Data Visualisation and Infographics with Adobe Illustrator (WGS)	Wageningen, the Netherlands
	Posters & Pitching (WGS)	Wageningen, the Netherlands
	Reviewing a scientific manuscript (WGS)	Wageningen, the Netherlands
2019	Scientific Writing (WGS)	Wageningen, the Netherlands
	Introduction to R (VLAG)	Wageningen, the Netherlands
	Applied Statistics (VLAG)	Wageningen, the Netherlands
	PhD Carousel (WGS)	Wageningen, the Netherlands
2018	Competence Assessment (WGS)	Wageningen, the Netherlands
	PhD Carousel (WGS)	Wageningen, the Netherlands
	PhD week (VLAG)	Wageningen, the Netherlands

Other activities		
2022	PhD study tour	Spain
2018	Research proposal writing	Wageningen, the Netherlands
2018-2022	Group meetings Food Quality & Design	Wageningen, the Netherlands
	Project meetings with project partners	Amsterdam, the Netherlands

Colophon

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