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

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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.

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, Santos, Casas, & Gómez, 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, Steele, & Duizer, 2018), slimy (Gössinger et al., 2018) and sticky (Yamagata, Izumi, Egashira, Miyamoto, & Kayashita, 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, Nishiyama, Putaux, Vignon, & Isogai, 2006; Spence, Venditti, Rojas, Habibi, & Pawlak, 2010; Stenstad, Andresen, Tanem, & Stenius, 2008; Taipale, Österberg, Nykänen, Ruokolainen, & Laine, 2010), sugar beets (Agoda-Tandjawa et al., 2010; Dinand, Chanzy, & Vignon, 1999), carrots

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(Siqueira, Oksman, Tadokoro, & Mathew, 2016) or fruits (Habibi, Mahrouz, & Vignon, 2009; Jongaroontaprangsee, Chiewchan, & Devahastin, 2018; Pelissari, do Amaral Sobral, & Menegalli, 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, Desloges, Dufresne, & Bras, 2012; Turbak, Snyder, & Sandberg, 1983, p. 374). 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, Gregersen, Moe, & Lenes, 2011; Lowys, Desbrieres, & Rinaudo, 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. (1983, p. 374). The functional and physicochemical properties of MFC have since then been explored in a broad range of foods including bread dough (Ström, Öhgren, & Ankerfors, 2013), hamburgers (Ström et al., 2013), ice cream (Velásquez-Cock et al., 2019), and mayonnaises (Choublab & Winuprasith, 2018; Golchoobi, Alimi, Shokoohi, & Yousefi, 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, Roberts, Fuqua, & Melchion, 1988; Koh & Hayama, 1997; Koppert & Velikov, 2018; Lemmers, Velikov, & Zuidam, 2017; Weibel, 2001, p. 251; 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. Materials & methods

2.1. Sample preparation

2.1.1. 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 3000 rpm for 10 min and afterwards passed twice through a high-pressure homogeniser (Microfluidizer M-110S, MicrofluidicsTM, USA) with a z-shape geometry (\emptyset 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 fibers 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, Mohan, Imhof, & Velikov, 2019; Nomena et al., 2018) and these fibrils form an attractive network.

2.1.2. 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.

2.1.3. Preparation of samples for instrumental measurements and sensory evaluations

MFC dispersions and XG solutions for sensory evaluations (Table 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.

2.2. Rheological characterisation

2.2.1. 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 μ m, 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 $^\circ C$ 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

Table 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				

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 to 100% (1–10000% 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".

2.2.2. 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, Friedman, & Taborek, 2012; Kibbelaar et al., 2020; Louvet, Bonn, & Kellay, 2014). A rheometer (MCR 300, Anton Paar, Austria) was used as the building block of the device. A speed controllable (ν) cylindrical geometry (ϕ 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 MAT-LAB 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, Fig. S1) using $\eta_s = 2^{n-1} n^n \phi_0(n) \gamma [\frac{dh_{min}}{d\tau}]^{-1}$, where $\phi_0(n)$ 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, Suryo, Yildirim, McKinley, & Basaran, 2003; Suryo & Basaran, 2006).

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 to 2200 rpm (equivalent to $4 \cdot 10^{-5} - 10^3$ 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 $^\circ$ 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.

2.4. Sensory evaluation

2.4.1. Participants

Dutch participants between 18 and 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/m²) participated in the study. Participants completed a general questionnaire before starting the sensory evaluation. Participants signed an informed consent and received financial

Table 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;	Custard, milk, water (smooth)			
	absence of lumpiness and graininess.	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	Ice cream, chocolate			
	itself in the mouth.				
Mouthcoating	The feeling that a layer of the sample remains behind in the mouth and	Mayonnaise			
	palate (after swallowing).				
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	Custard, milk, water (smooth)			
	graininess; the sample flows easily in the mouth.	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	Greek yoghurt (thick)			
	flow or deform in the mouth.	Water (not thick)			

reimbursement after completion of the test session.

2.4.2. 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). 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 1 h 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).

2.5. 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, Brockhoff, & Christensen, 2017; Lenth, 2019). Principal Component Analysis (PCA) was performed with 95% confidence ellipses for the twelve samples (Kassambara & Mundt, 2020; Lê, Josse, & Husson, 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 $\alpha = 0.05$ was used.

3. Results & discussion

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 Fig. 1. At high MFC (1.0, 1.5 and 2.0 wt%) and XG concentrations (2.0,



Fig. 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). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.) 3.4 and 4.3 wt%), shear viscosities of both thickeners matched over a broad range of shear rates $(1-1000 \text{ s}^{-1})$. 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 $(1-10 \text{ s}^{-1})$ which might be due to flow instabilities (see also Supp. Material, Fig. S2). All MFC dispersions and XG solutions displayed shear thinning behaviour. As expected, shear and extensional viscosities increased with increasing thickener concentrations (Lundahl, Berta, Ago, Stading, & Rojas, 2018; Martín-Alfonso, Cuadri, Berta, & Stading, 2018; Moberg, Rigdahl, Stading, & Bragd, 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, Wiklund, Altskär, Ekberg, & Stading, 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, Turcanu, Ekberg, & Gallegos, 2017; Hadde & Chen, 2019; Hadde, Cichero, Zhao, Chen, & Chen, 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, Velikov, & Bonn, 2018). Correspondingly, an increase in the apparent extensional viscosity is observed (Fig. 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.

As anticipated, viscoelastic moduli (G' & G") and yield stress increased with increasing concentrations of MFC and XG (Table 3, Fig. 2) (Agoda-Tandjawa et al., 2010; Iotti et al., 2011; Lowys et al., 2001; Lundahl et al., 2018; Martín-Alfonso et al., 2018; Ross, Tyler, Borgognone, & Eriksen, 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'

Table 3

Mean (\pm 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' (P	a)	G" (Pa)		
	MFC	XG	MFC	XG		
1	3.1 ± 0.3	0.0 ± 0.0	0.6 ± 0.1	0.3 ± 0.0		
2	$\textbf{6.9} \pm \textbf{0.5}$	$\textbf{0.2}\pm\textbf{0.0}$	1.1 ± 0.1	0.5 ± 0.0		
3	19.3 ± 0.2	0.5 ± 0.1	$\textbf{2.7} \pm \textbf{0.0}$	$\textbf{0.8} \pm \textbf{0.0}$		
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	$\textbf{76.2} \pm \textbf{16.3}$	13.3 ± 0.1		
6	996.2 ± 21.8	62.1 ± 0.6	142.1 ± 1.7	17.4 ± 0.2		

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, Flores, Torres, & Galindo, 1991; Song, Kim, & Chang, 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, Ishioka, & Matsumoto, 2002).

In accordance with literature, hysteresis was observed for MFC dispersions (Agoda-Tandjawa et al., 2010; Martoïa et al., 2015; Schenker, Schoelkopf, Gane, & Mangin, 2018) and XG solutions (Alghooneh, Razavi, & Kasapis, 2018; Ghannam, Selim, Zekri, & Esmail, 2019; Silva & Lucas, 2018) (Table 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 and 20 s^{-1} , whereas hysteresis of XG solutions occurred at a larger range of shear rates (1-500 s^{-1} ; Supp. Material, Fig. 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 ((AUCupward-AUCdownward)/AUCupward) 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 (Fig. 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, Fig. 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.

3.2. Tribological properties

Mean friction coefficients of MFC dispersions and XG solutions as a function of sliding speed are displayed in Fig. 3. Friction coefficients of



Fig. 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 \sim 2) and a linear fit for XG. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 4

Absolute and relative hysteresis area of microfibrillated cellulose (MFC) dispersions and xanthan gum (XG) solutions (mean \pm 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 hyste	resis area (Pa· s ⁻¹)	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	$\textbf{4.2}\pm\textbf{0.1}$				
6	2726 ± 1228	6852 ± 440	$\textbf{2.8} \pm \textbf{1.1}$	$\textbf{8.0}\pm\textbf{0.5}$				

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, Stokes, & Spikes, 2006, 2005; Selway, Chan, & Stokes, 2017; Stokes, Boehm, & Baier, 2013). On the other hand, our results show no clear reduction in friction for increasing concentrations of MFC. This confirms recent work by Kinoshita, Inada, and Matsumoto (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, Saarinen, Salmela, & Seppälä, 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.

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 5. To summarise the results of the sensory evaluation, Fig. 4 shows the Principal Component Analysis bi-plot positioning the twelve samples in the sensory space.

3.3.1. Effect of thickener concentration on sensory perception

A significant main effect of thickener concentration on all sensory attributes was found (Table 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.

3.3.1.1. 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



Fig. 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.

Table 5

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Mean intensities (\pm 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														Thicker	ner ation	Thickener	type	Interaction	a effect
		MFC-2	MFC-3	MFC-4	MFC-5	MFC-6	XG-1	XG-2	XG-3	XG-4	XG-5	XG-6	F-value	<i>p</i> - value	F-value	<i>p</i> - value	F-value	<i>p</i> - value		
Appearance																				
Glossiness	$6.0~\pm$ $0.3^{ m ab}$	$\begin{array}{c} 5.5 \ \pm \\ 0.3^{ab} \end{array}$	$\begin{array}{c} 5.6 \ \pm \\ 0.3^{ab} \end{array}$	$5.1~\pm$ $0.3^{ m bc}$	$\begin{array}{c} \textbf{4.3} \pm \\ \textbf{0.3}^{cd} \end{array}$	$3.4 \pm 0.3^{ m d}$	$\begin{array}{c} 5.5 \ \pm \\ 0.3^{ab} \end{array}$	$\begin{array}{c} 5.8 \pm \\ 0.3^{ab} \end{array}$	$\begin{array}{c} \textbf{4.2} \pm \\ \textbf{0.3}^{cd} \end{array}$	$\begin{array}{c} 6.3 \pm \\ 0.2^{a} \end{array}$	$\begin{array}{c} 6.3 \pm \\ 0.2^{\rm a} \end{array}$	$\begin{array}{c} 5.9 \pm \\ 0.3^{ab} \end{array}$	F(5,789) = 9.3	***	F(1,789) = 29.1	***	F(5,789) = 23.1	***		
Red colour intensity	$6.7~\pm$ $0.2^{ m a}$	$\begin{array}{c} 6.3 \pm \\ 0.2^{ab} \end{array}$	$5.1 \pm 0.2^{ m cd}$	4.6 ± 0.2^{de}	$\begin{array}{c} 3.6 \ \pm \\ 0.2^{\rm f} \end{array}$	$\begin{array}{c} 3.5 \pm \\ 0.2^{\rm f} \end{array}$	$\begin{array}{c} 6.5 \pm \\ 0.2^a \end{array}$	$6.7 \pm 0.2^{\mathrm{a}}$	$6.5~\pm$ $0.2^{ m a}$	$\begin{array}{c} 5.6 \ \pm \\ 0.2^{bc} \end{array}$	$\begin{array}{c} 4.6 \pm \\ 0.2^{de} \end{array}$	$\begin{array}{c} \textbf{4.4} \pm \\ \textbf{0.2}^{e} \end{array}$	F(5,787) = 148.3	***	F(1,787) = 97.5	***	F(5,787) = 8.7	***		
Sliminess	$\begin{array}{c} \textbf{0.9} \pm \\ \textbf{0.2}^{de} \end{array}$	$\begin{array}{c} 1.2 \pm \\ 0.2^{de} \end{array}$	$2.7~\pm$ $0.3^{ m c}$	$6.2~\pm$ $0.3^{ m ab}$	$\begin{array}{c} 5.9 \ \pm \\ 0.4^{b} \end{array}$	$\begin{array}{c} 5.8 \ \pm \\ 0.4^{\rm b} \end{array}$	$\begin{array}{c} 0.1 \ \pm \\ 0.0^{\rm e} \end{array}$	$\begin{array}{c} 0.4 \ \pm \\ 0.1^{de} \end{array}$	$\begin{array}{c} 1.2 \ \pm \\ 0.2^{\rm d} \end{array}$	$\begin{array}{c} 5.9 \ \pm \\ 0.3^{b} \end{array}$	$7.1~\pm$ $0.2^{ m a}$	$7.1~\pm$ $0.2^{ m a}$	F(5,788) = 342.4	***	F(1,788) = 1.3	n.s.	F(5,788) = 12.6	***		
Smoothness	$\begin{array}{c} \textbf{7.8} \pm \\ \textbf{0.2}^{\textbf{a}} \end{array}$	$\begin{array}{c} 7.5 \ \pm \\ 0.2^{a} \end{array}$	$\begin{array}{c} \textbf{6.9} \pm \\ \textbf{0.3}^{ab} \end{array}$	$5.6~\pm$ 0.3^{c}	$5.1~\pm$ $0.3^{ m cd}$	$4.1 \pm 0.4^{ m d}$	$\begin{array}{c} 8.0 \ \pm \\ 0.2^{\rm a} \end{array}$	$\begin{array}{c} 8.0 \ \pm \\ 0.2^{a} \end{array}$	$\begin{array}{c} \textbf{5.8} \pm \\ \textbf{0.4}^{bc} \end{array}$	$\begin{array}{c} 5.2 \pm \\ 0.4^{cd} \end{array}$	$\begin{array}{c} 4.3 \pm \\ 0.4^d \end{array}$	$\begin{array}{c} 4.2 \pm \\ 0.4^d \end{array}$	F(5,789) = 81.2	***	F(1,789) = 3.2	n.s.	F(5,789) = 2.8	*		
Thickness	$\begin{array}{c} 1.0 \pm \\ 0.2^{\rm fg} \end{array}$	$\begin{array}{c} 1.5 \pm \\ 0.2^{\rm f} \end{array}$	$2.9 \pm 0.2^{ m e}$	$7.0~\pm$ $0.2^{ m c}$	$\begin{array}{c} 8.2 \pm \\ 0.1^{\rm ab} \end{array}$	8.5 ± 0.1^{a}	$\begin{array}{c} 0.3 \pm \\ 0.1^{ m g} \end{array}$	$\begin{array}{c} 0.4 \pm \\ 0.1^{g} \end{array}$	$\begin{array}{c} 1.2 \pm \\ 0.2^{\rm f} \end{array}$	$\begin{array}{c} 5.6 \ \pm \\ 0.2^{d} \end{array}$	$7.6~\pm$ $0.1^{ m bc}$	$\begin{array}{c} 8.0 \pm \\ 0.1^{ab} \end{array}$	F(5,787) = 1246.9	***	F(1,787) = 150.6	***	F(5,787) = 5.9	***		
Transparency	$\begin{array}{c} 1.5 \pm \\ 0.2^{c} \end{array}$	$\begin{array}{c} 1.2 \pm \\ 0.2^{cd} \end{array}$	$\begin{array}{c} 1.0 \pm \\ 0.2^{cde} \end{array}$	$\begin{array}{c} 0.6 \ \pm \\ 0.1^{def} \end{array}$	$\begin{array}{c} 0.4 \pm \\ 0.1^{ef} \end{array}$	$\begin{array}{c} 0.3 \pm \\ 0.1^{\rm f} \end{array}$	$7.5~\pm$ $0.2^{ m a}$	$7.1~\pm$ $0.2^{ m a}$	$\begin{array}{c} \textbf{4.4} \pm \\ \textbf{0.3}^{b} \end{array}$	$\begin{array}{c} 0.6 \ \pm \\ 0.1^{def} \end{array}$	$\begin{array}{c} 0.7 \pm \\ 0.2^{def} \end{array}$	$\begin{array}{c} 0.6 \ \pm \\ 0.1^{def} \end{array}$	F(5,788) = 371.3	***	F(1,788) = 1121.4	***	F(5,788) = 213.7	***		
Flavour																				
Cardboard flavour	$1.9~\pm$ $0.3^{ m c}$	$\begin{array}{c} 2.0 \ \pm \\ 0.3^{bc} \end{array}$	$\begin{array}{c} 3.0 \pm \\ 0.4^{\rm bc} \end{array}$	$4.5~\pm$ 0.4 ^a	$5.1~\pm$ 0.4^{a}	$5.2 \pm 0.4^{\mathrm{a}}$	$0.4~\pm~$ 0.1^{d}	0.3 ± 0.1^{d}	$0.3~\pm$ $0.1^{ m d}$	$\begin{array}{c} 2.0 \ \pm \\ 0.3^{bc} \end{array}$	$\begin{array}{c} \textbf{2.4} \pm \\ \textbf{0.3}^{bc} \end{array}$	$\begin{array}{c} 3.0 \ \pm \\ 0.3^b \end{array}$	F(5,789) = 61.5	***	F(1,789) = 247.0	***	F(5,789) = 2.2	*		
Strawberry flavour	$\begin{array}{c} \textbf{4.8} \pm \\ \textbf{0.3}^{abc} \end{array}$	$\begin{array}{c} 5.0 \ \pm \\ 0.3^{ab} \end{array}$	$\begin{array}{c} \text{4.5} \pm \\ \text{0.3}^{\text{bc}} \end{array}$	$3.4~\pm$ $0.3^{ m de}$	$\begin{array}{c} \textbf{2.9} \pm \\ \textbf{0.3}^{\text{def}} \end{array}$	$\begin{array}{c} \textbf{2.8} \pm \\ \textbf{0.3}^{\rm ef} \end{array}$	$5.5~\pm$ $0.3^{ m a}$	$\begin{array}{c} \textbf{5.4} \pm \\ \textbf{0.3}^{ab} \end{array}$	$5.3~\pm$ $0.2^{ m ab}$	$\begin{array}{c} 3.8 \pm \\ 0.3^{cd} \end{array}$	$\begin{array}{c} \textbf{2.6} \pm \\ \textbf{0.3}^{\text{ef}} \end{array}$	$\begin{array}{c} \textbf{2.2} \pm \\ \textbf{0.3}^{\rm f} \end{array}$	F(5,788) = 70.8	***	F(1,788) = 4.2	*	F(5,788) = 3.5	**		
Sweetness	$6.1~\pm$ $0.2^{ m ab}$	$\begin{array}{c} 5.7 \ \pm \\ 0.3^{ab} \end{array}$	$\begin{array}{c} \textbf{5.2} \pm \\ \textbf{0.2^b} \end{array}$	4.2 ± 0.3^{c}	3.5 ± 0.3^{cde}	$\begin{array}{c} 3.4 \pm \\ 0.3^{cde} \end{array}$	$6.4 \pm 0.2^{\mathrm{a}}$	$6.5 \pm 0.2^{ m a}$	$\begin{array}{c} 5.6 \ \pm \\ 0.2^{\rm ab} \end{array}$	$\begin{array}{c} 4.0 \ \pm \\ 0.3^{cd} \end{array}$	$\begin{array}{c} \textbf{3.2} \pm \\ \textbf{0.2}^{\text{de}} \end{array}$	$\begin{array}{c} \textbf{2.7} \pm \\ \textbf{0.2}^{\text{e}} \end{array}$	F(5,788) = 104.6	***	F(1,788) = 0.2	n.s.	F(5,788) = 4.1	**		
Texture																				
Creaminess	$0.2 \pm 0.1^{ m b}$	$\begin{array}{c} 0.3 \pm \\ 0.1^{b} \end{array}$	$\begin{array}{c} 0.9 \pm \\ 0.2^{b} \end{array}$	$2.0~\pm$ $0.3^{ m a}$	$2.5~\pm$ $0.4^{ m a}$	$2.9~\pm$ $0.4^{ m a}$	$\begin{array}{c} 0.1 \pm \\ 0.1^{ m b} \end{array}$	$\begin{array}{c} 0.2 \pm \\ 0.1^{\mathrm{b}} \end{array}$	$0.6~\pm$ $0.2^{ m b}$	$2.5~\pm$ $0.3^{ m a}$	$\begin{array}{c} 2.3 \pm \\ 0.3^{ m a} \end{array}$	$\begin{array}{c} 2.5 \pm \\ 0.4^a \end{array}$	F(5,789) = 62.0	***	F(1,789) = 0.9	n.s.	F(5,789) = 1.0	n.s.		
Melting	$0.8~\pm$ $0.2^{ m c}$	$\begin{array}{c} 0.8 \pm \\ 0.2^{ m c} \end{array}$	$1.0~\pm$ $0.2^{ m bc}$	$2.3~\pm$ $0.3^{ m a}$	$2.0~\pm$ $0.3^{ m ab}$	$2.3~\pm$ $0.3^{ m a}$	$0.6 \pm 0.2^{ m c}$	$\begin{array}{c} 0.8 \pm \\ 0.2^{ m c} \end{array}$	$1.4~\pm$ $0.3^{ m abc}$	$1.6~\pm$ $0.3^{ m abc}$	$rac{1.1 \pm 0.2^{ m bc}}{}$	$1.5 \pm 0.3^{ m abc}$	F(5,789) = 12.7	***	F(1,789) = 9.5	**	F(5,789) = 3.0	*		
Mouthcoating	$0.9 \pm 0.2^{ m e}$	$1.1 \pm 0.2^{ m e}$	$1.8 \pm 0.2^{ m de}$	$\begin{array}{c} \textbf{2.8} \pm \\ \textbf{0.3}^{d} \end{array}$	4.0 ± 0.3^{c}	4.6 ± 0.3 ^c	$\begin{array}{c} 0.8 \pm \\ 0.2^{ m e} \end{array}$	$1.2 \pm 0.2^{ m e}$	$1.4 \pm 0.2^{ m e}$	$^{ m 4.9~\pm}_{ m 0.3^{bc}}$	$\begin{array}{c} 5.8 \pm \\ 0.3^{ab} \end{array}$	${6.0} \pm {0.3^{a}}$	F(5,789) = 148.3	***	F(1,789) = 36.1	***	F(5,789) = 11.1	***		
Pulpiness	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	$1.0 \pm 0.2^{\rm bc}$	$1.6 \pm 0.3^{\mathrm{b}}$	$2.8 \pm 0.4^{\mathrm{a}}$	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	$\begin{array}{c} 0.0 \pm \\ 0.0^{ m d} \end{array}$	0.5 ± 0.2^{cd}	$1.0 \pm 0.3^{ m bc}$	1.1 ± 0.3^{c}	F(5,788) = 42.1	***	F(1,788) = 21.3	***	F(5,788) = 7.2	***		
Sliminess	$0.3 \pm 0.1^{\rm f}$	$0.4 \pm 0.1^{\rm f}$	1.5 ± 0.2 ^e	3.5 ± 0.4^{d}	4.5 ± 0.4 ^c	4.5 ± 0.3 ^c	$0.1 \pm 0.1^{\rm f}$	$0.2 \pm 0.1^{\rm f}$	0.6 ± 0.1 ^{ef}	6.0 ± 0.3 ^b	7.5 ± 0.2^{a}	7.9 ± 0.2 ^a	F(5,788) = 421.0	***	F(1,788) = 122.5	***	F(5,788) = 46.8	***		
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	$6.2 \pm 0.3^{ m bc}$	5.9 ± 0.4 ^{bc}	F(5,787) = 56.1	***	F(1,787) = 0.5	n.s.	F(5,787) = 1.3	n.s.		
Stickiness	0.4 ±	0.6 ±	0.7 ±	1.0 ± 0.2 ^c	2.2 ± 0.3^{b}	2.2 ± 0.3^{b}	0.2 ±	0.4 ±	0.5 ±	3.0 ± 0.3 ^b	4.8 ± 0.4^{a}	5.4 ±	F(5,787) =	***	F(1,787) = 102.1	***	F(5,787) = 29.6	***		
Thickness	0.4 ± 0.1^{e}	$0.2 \pm 0.1^{ m e}$	${}^{0.2}_{1.5\pm}_{0.2^{ m d}}$	0.2 4.6 ± 0.3 ^c	$5.5 \pm 0.2^{\rm b}$	6.5 ± 0.2^{a}	$0.2\pm 0.1^{ m e}$	$0.3 \pm 0.1^{ m e}$	$0.8 \pm 0.1^{ m de}$	$4.9 \pm 0.3^{ m bc}$	$6.7\pm 0.2^{\mathrm{a}}$	6.9 ± 0.2^{a}	F(5,789) = 651.0	***	F(1,789) = 3.4	n.s.	F(5,789) = 8.1	***		



Fig. 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. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

concentrations has been reported by others (Kim, Hwang, Song, & Lee, 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 (Fig. 1) and viscosity is related to visual thickness (Christensen & Casper, 1987) and sliminess (Brandenstein, Busch-Stockfisch, & Fischer, 2015).

3.3.1.2. 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, Hollowood, Linforth, & Taylor, 2002; Gössinger et al., 2018; Hollowood, Linforth, & Taylor, 2002; Kim et al., 2017; Malone, Appelqvist, & Norton, 2003; Matta, Chambers IV, Garcia, & Helverson, 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.

3.3.1.3. Texture. Increasing the thickener concentration resulted in large differences in shear viscosity (Fig. 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, Morris, & Taylor, 1983). Since creaminess, sliminess and stickiness are correlated with perceived thickness (Frøst & Janhøj, 2007; He, Hort, & Wolf, 2016; Lyly et al., 2003; Morris, Richardson, & Taylor, 1984; Upadhyay, Aktar, & Chen, 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, Busch-Stockfisch, & Fischer, 2015Brandenstein, Busch-Stockfisch, & Fischer, 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.

3.3.2. 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 5). The differences between the two thickeners is reflected by the second dimension of the PCA bi-plot (Fig. 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.

3.3.2.1. 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 (Fig. 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 (Fig. 1).

3.3.2.2. 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 $125 \,^{\circ}$ C for $15 \,\text{min}$) 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.

3.3.2.3. 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, Murray, & Dickinson, 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, Stenzel, Murray, & Dickinson, 2005; Frøst & Janhøj, 2007).

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 (Fig. 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 (Fig. 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, Ichiba, Silveira, Albano, & Nicoletti, 2018; Krzeminski et al., 2013; Sonne, Busch-Stockfisch, Weiss, & Hinrichs, 2014).

The right side of the MFA plot shows that the texture attributes





Fig. 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. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

mouthcoating, slimy and sticky are correlated (Fig. 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, vield 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".

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.

CRediT authorship contribution statement

Annelies E. Blok: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft. Dieuwerke P. Bolhuis: Conceptualization, Methodology, Writing – review & editing. Heleen V. M. Kibbelaar: Methodology, Investigation, Formal analysis, Writing – review & editing. Daniel Bonn: Methodology, Writing – review & editing. Krassimir P. Velikov: Conceptualization, Methodology, Writing – review & editing. Markus Stieger: Conceptualization, Methodology, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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A.E. Blok et al.

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