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Changing the Oral Tribology of Emulsions Through Crystallization of the Dispersed Triglyceride Phase

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ABSTRACT

Suspoemulsions are used for food, cosmetic and pharmaceutical products, including food such as dairy products and non-dairy alternatives. Product properties, such as flow behavior or sensory perception of non-dairy products differ from those of dairy products and are therefore perceived by consumers as products of inferior quality. One reason for this may be the crystallization behavior of the added triglycerides leading to differences in solid fat content in comparison to cow milk. This is discussed with the solidity of the dispersed phase as a parameter of suspoemulsions. The solidity was varied by using low and high melting triglycerides and measuring at different temperatures. The dispersed phase fraction is $\varphi = 30\%$. The droplet size distribution showed a $x_{50,3}$ of 1.2 and 3.66 µm, mimicking the droplet sizes of milk and dairy cream. Rheological frequency sweeps were carried out within a temperature range from 5°C to 50°C. The differences in solidity of the dispersed phase caused no changes in viscosity at each temperature. In contrast, oral tribology distinguished different solidities of the dispersed phase with changes in the friction coefficient. The friction coefficient was determined for increasing rotational speeds (0.01–100 mm/s), to compare the so called Stribeck curves with each other. In general, with increasing solidity of the dispersed phase, the friction coefficient increases. Comparing the Stribeck curves of pure butter fat suspoemulsion with those of plant-based fat suspoemulsions, different plant-based fats can be mixed, to mimic the friction profile of milk products in plant-based alternatives.

1 | Introduction

Suspensions and emulsions are widely used in the food, pharmaceutical and cosmetics industries. Depending on the temperature, the dispersed phase can change from solid (suspension) to liquid (emulsion) or vice versa. Systems which change between solid and liquid dispersed phase or are partly in emulsion and partly in suspension state are called suspoemulsions. An example for a suspoemulsion is milk, where the emulsified butter fat is partially crystalline and partially liquid over a wide temperature range (Meagher et al. 2007). The distinct mouthfeel of emulsions, as well as the special properties of dairy cream in whipped cream production, are linked to these suspoemulsion properties (Lundin 2013; Schwimmer 2013; Zhou et al. 2022). However, the production of milk is associated with high CO_2-emissions, which is why many consumers have switched to plant-based milk alternatives in recent years (Aschemann-Witzel et al. 2021). Unfortunately, the quality, especially in terms of mouthfeel, is often considered inferior (Cardello et al. 2022). Mouthfeel

Abbreviations: CSS, cream-sized-suspoemulsions; HCG, hydrogen coco-glycerides; MCT, medium chain triglycerides; MSS, milk-sized-suspoemulsions; PBF, pure butter fat is described in the literature as a sensory perception from a combination of viscosity and friction values of the food (Mouritsen 2017).

One big difference between milk and plant-based alternatives is the aggregate state of the dispersed phase. Canola or sunflower oil are the most commonly used triglycerides in plant-based milk alternatives. These milk alternatives are completely in emulsion state. In addition to the dispersed triglycerides, plant-based emulsions contain water as continuous phase, carbohydrates and plant-based emulsifiers. The properties of these ingredients are chosen to mimic the properties of milk. Intensive research is currently being conducted into the use of plant-based emulsifiers for use in emulsions. Plant proteins are generally bigger than milk proteins (Hinderink et al. 2021). Emulsions made with plant proteins are less stable (Jiang et al. 2015) and are more vulnerable to aggregation and coalescence (Hinderink et al. 2020).

Triglycerides are esters of the trivalent alcohol glycerol esterified with three fatty acid chains (Sato 2001). Natural fats are present as a triglyceride mixture, which is highly dependent on the origin (species of origin, environmental influences) and the processing of the triglyceride mixture (Sampels, Strandvik, and Pickova 2009). The fatty acid chain lengths, the number of double bonds and the distribution of esterified fatty acids at the glycerol backbone determine the temperature at which the triglycerides arrange themselves into different crystal lattices. As a result, triglyceride mixtures have a wide melting range, often ranging over several 10 K. At different temperatures, this leads to liquid, semi-crystalline or crystalline phases (Sato 2001). The solid fat content is specified as the amount of crystalline triglyceride divided by the total triglyceride amount and is given as percentage. The solid fat content of pure butter fat at 0°C is approximately 66%, at 20°C it is approximately 21% and at 30°C it is approximately 6% (Meagher et al. 2007). Butter fat is completely molten when exceeding 36.8°C (Keskin 2022; Makhlouf et al. 1987). The melting range of palm fat is $T_M = 36°C–42°C$ (Lida 2018) and of coconut fat it is $T_M = 20°C–26°C$ (Che Man et al. 2003). By mixing low-melting and high-melting triglyceride mixtures, the melting behavior can be adjusted (Motamedzadegan et al. 2020; Piska et al. 2006). However, solid fat content of triglyceride mixtures in bulkphase and in suspoemulsions can differ at the same temperature. In the bulk phase, nuclei are present which induce heterogeneous nucleation and the crystals can grow unhindered. With droplets not every droplet can contain a nucleus and therefore heterogeneous and homogeneous nucleation occur. For homogeneous nucleation higher supercooling is required to crystallize the droplets. In the case of suspoemulsions, this leads to a dispersed phase with supercooled liquid droplets and crystalline structures. The smaller the size of droplets in the dispersed phase, the less likely a nucleus is in each droplet and the formation of supercooled liquid droplets increases. Additionally, the droplet crystallization behavior may be influenced by polydispersity, cooling rate, external forces and additives (Abramov, Ruppik, and Schuchmann 2016; Reiner et al. 2023b).

Tribology is traditionally used to design journal bearings in order to represent the friction between two contact surfaces (Stribeck 1902; Woydt and Wäsche 2010). The friction coefficient μ_f is plotted against the dimensionless speed, the speed or the film thickness ratio to achieve the so-called Stribeck curve (He et al. 2017). The theory can also be applied if particles are present in the lubricant between the contact surfaces (Li et al. 2023).

Oral tribology lubrication properties that occur during chewing are considered resulting in less force, smaller rotational speed than in material sciences and soft surfaces are selected (Pondicherry, Rummel, and Laeuger 2018; Sarkar et al. 2021; Xu, Yu, and Zhong 2022). Oral tribology can be used to measure the friction values of food products, which can then be linked to the perceived mouthfeel (Prakash, Tan, and Chen 2013). The Stribeck curve typically consists of three areas, which are shown in Figure 1a schematically. Area I with high friction is called boundary friction. In area II, mixed friction occurs and in area III, elastohydrodynamic lubrication appears. The Stribeck curve differs depending on the properties of the tribological system (Sarkar et al. 2021). In recent publications, a solid ball pressing on three soft pins or plates is used to investigate liquid systems, see Figure 1b (Pondicherry, Rummel, and Laeuger 2018; Principato et al. 2022), thereby representing the contact surface between the palate and tongue (Sarkar et al. 2019), see Figure 1c. Tribology gives information of friction, wear and lubrication, based on surface phenomena (Sasaki 2023), while rheology gives

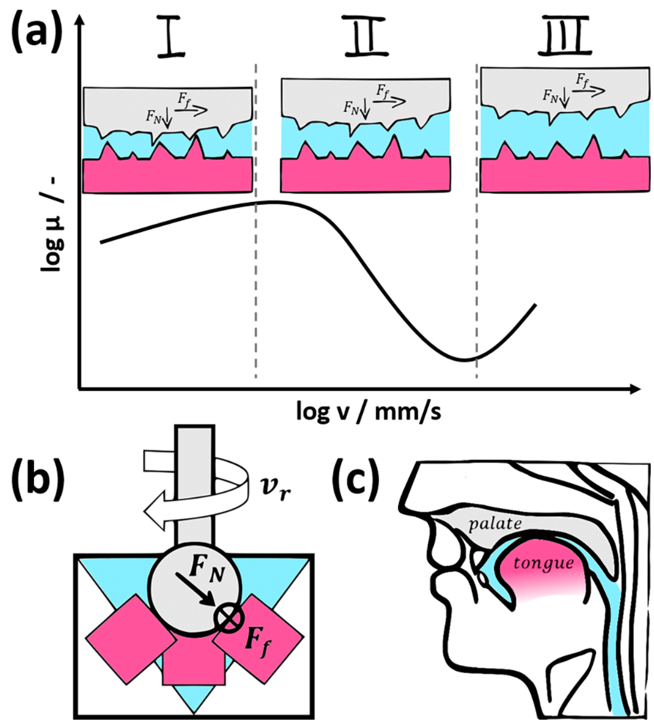

FIGURE 1 | (a) Tribological measurement with friction coefficient μ_f over rotational speed v_r depicted Stribeck curves. The frictional coefficient μ_f a function of the normal Force F_N and of the frictional force F_f and is calculated via $\mu_f = F_f/F_N$. The Stribeck curve consists of the boundary friction area I, mixed friction area II and elastohydrodynamic lubrication area III. (b) Typical configuration for researching liquid systems with oral tribology, using a rotating ball pressing on three polymer pins (c) Schematic representation of the contact area of palate and tongue. Based on (Pondicherry, Rummel, and Laeuger 2018; Prakash, Tan, and Chen 2013; Principato et al. 2022; Sarkar et al. 2019).

information about how a fluid flow or deform under an applied force (Morris 2023).

Sensory panels confirm that there is a correlation between creaminess and dispersed phase proportion. Friction in the oral cavity therefore influences the sensation of processed foods (Chojnicka-Paszun, de Jongh, and de Kruif 2012; Xu, Yu, and Zhong 2022). The perception results from the friction between the food particles and the oral mucosa (Wijk & de Wijk and Prinz 2006).

The use of emulsified droplets and the spreading of a lubricating oil film reduces the friction coefficient (Liu et al. 2016b). The same trends were found with fat particles (Nguyen, Bhandari, and Prakash 2016). Studies showed several trends to enhance the lubrication properties of food emulsions: Addition of saliva (Selway and Stokes 2013; Stokes, Boehm, and Baier 2013), increased dispersed phase fraction (Selway and Stokes 2013) and the influence of different proteins (Kew et al. 2021). Suspensions have also been investigated with oral tribology. Several outcomes are discussed in literature. Solid particles can enter the contact zone and convert sliding of surfaces to rolling of particles, which reduces friction (Garrec and Norton 2013; Rudge et al. 2021; Sarkar et al. 2017; Yakubov et al. 2015). The solid particles can also accumulate in and around the inlet and block the supply of triglycerides into the contact, or be entrained without rolling, either of which leads to increased friction (Chojnicka et al. 2008; Chojnicka-Paszun, Doussinault, and de Jongh 2014; Lee et al. 2003; Liu et al. 2016a; Luengo et al. 1997; Rodrigues et al. 2021).

Rudge et al. investigated how glass beads ranging from 100 to 2000 μm behave as a lubricant between polymer surfaces (Rudge et al. 2021). The authors found that the friction coefficient increases with increasing normal force, decreased number of particles and with smaller particles. The reduction in friction is attributed to the rolling of the beads. The increase of the friction coefficient is explained by the deformation of the pins at higher normal force, causing the polymer surfaces to touch and thus increasing the friction. In a kappa-carrageenan suspension gel particles are drawn into the contact gap between a PDMS bead and a PDMS disc and friction is reduced (Garrec and Norton 2013). Whey-protein microgel particles ($\varphi = 10$–80 vol.%) also reduced the friction coefficient (Sarkar et al. 2017).

Rodrigues et al. investigated the influence of the solid content of melted chocolate by mixing chocolate with different proportions of cocoa butter (Rodrigues et al. 2021). Chocolate acts as a bulk fluid in thick-films. Mixed with saliva an emulsion is formed, where the dispersed triglyceride phase coalesces and coats the interacting surfaces, thereby controlling lubrication in thin-film conditions. The authors observe that particle entrainment increases the friction—this is most likely associated with angular-shaped sugar particles. As the shape of a particle will affect its ability to roll, the use of small spherical particles may reduce the friction. Liu et al. investigated the tribological properties of liquid and semi-solid food systems containing micro-granular rice starch (Liu et al. 2016a). Native and gelatinized rice starch dispersions were compared. The native rice starch showed a high friction coefficient, whereas the friction coefficient of gelatinized rice starch was lower. The reason given was that the native rice particles sticking to the roughness of the surface pairs, agglomerate and thus further increasing the boundary friction, whereas the gelatinized rice starch particles even out the roughness, thus tend to reduce the roughness and thus ensure lower friction. Furthermore, for protein-aggregate dispersions (Chojnicka et al. 2008), polysaccharide-gelled protein suspensions (Chojnicka-Paszun, Doussinault, and de Jongh 2014) and chocolate particles (Luengo et al. 1997) increased friction values were found. This might be due to the differences in the aggregate state of the dispersed phase causing different friction values. The solidity of the dispersed phase, which is directly related to the proportion of crystalline fat present. The solidity of butter fat increases with the amount of crystals present (Ziarno et al. 2023).

In this work, the suspoemulsions with a dispersed phase fraction $\varphi = 30\%$ and a droplet size of around 1.2 μm are first examined for crystallinity using established methods (diffraction scanning calorimetry and polarized microscopy). The rheological properties are measured and discussed. Then the results from oral tribology are discussed and data of emulsions with different dispersed phases are compared, to see if either friction is decreasing or increasing with the existence of dispersed partially solid and solid particles. Further the curve progressions of the different friction profiles are compared and connected to literature. The influence of the droplet size and a comparison to dairy cream are shown.

2 | Materials and Methods

2.1 | Materials

Hydrogenated-Coco-Glycerides (HCG, Softisan 100, $T_M \approx 34$°C, 70% C12/14, 30% C16/18) and Medium-chain-triglycerides (MCT, Witarix 60:40, $T_M < 0$°C, 55%–65% C8, 35%–45% C10) were provided by IOI Oleo GmbH (Germany). Pure butter fat (PBF, Butaris, $T_M \approx 28$°C–33°C, 63% saturated fatty acids, 29% unsaturated fatty acids) as obtained from a local supermarket (Butaris, Dairy Fine Food GmbH, Uelzen, Germany). Powdered whey protein isolate was provided from Fonterra Co-operative Group Limited (New Zealand). Dairy cream was obtained from a local supermarket (Rewe Beste Wahl, 32% fat). All materials were used without further purification. All samples were prepared with ultrapure water (Thermo Scientific Barnstead MicroPure, Waltham, USA).

2.2 | Sample Preparation

2.2.1 | Suspoemulsion Preparation

A stock solution was prepared by diluting 2 wt% whey protein isolate in water under stirring at room temperature. The solution then was transferred into a 10 mL Luer Lock syringe (Omnifix, B. Braun SE, Melsungen, Germany). The HCG, the HCG/MCT-mixture and the PBF were melted 10 K above the melting range during stirring on a heat plate. The different triglycerides were then transferred into a second 10 mL Luer Lock syringe according to the amount in Table 1 for the according emulsions.

TABLE 1 | Formulation parameters of the suspoemulsions. The suspoemulsions are named after the used triglyceride phase. Medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG), a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) and pure butter fat (PBF) were used.

Suspoemulsions	Whey protein isolate solution/wt%	HCG/wt%	MCT/wt%	PBF/wt%	Dispersed phase solidity at room temperature
MCT	70	30	—	—	soft
MCT/HCG		15	15	—	pasty
HCG		—	30	—	hard
PBF		—	—	30	pasty

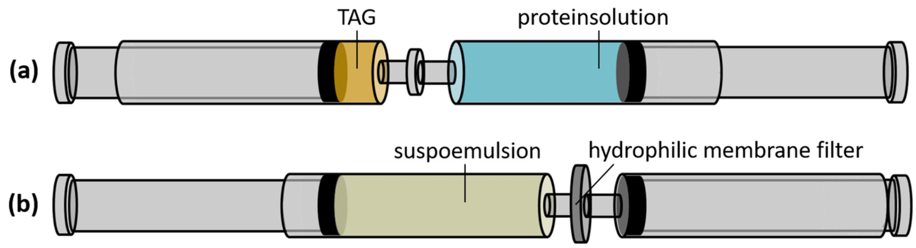

FIGURE 2 | Premix membrane emulsification process with (a) processing triglyceride and whey protein isolate solution without filter and (b) processing by using a customized pneumatic membrane emulsification process and a hydrophilic membrane filter with pore size 10 μm for suspoemulsions with big droplets (cream-sized-suspoemulsion, CSS), and a filter with pore size 0.8 μm for suspoemulsions with small droplets (milk-sized-suspoemulsion, MSS).

All syringes were kept at 60°C in a waterbath for the triglycerides to remain liquid until processing in the premix membrane emulsification process. Changing the dispersed phase causes changes in solidity of the dispersed droplets at room temperature, resulting in soft, pasty and hard solidity. The dispersed phase fraction is $\varphi = 30\%$ in order to obtain distinct measurements. All suspoemulsions were freshly prepared on the day of the measurement.

2.2.2 | Premix Membrane Emulsification

For Premixing the syringes were connected with a Luer Lock adapter (Rotilabo, Carl Roth GmbH + Co. KG, Karlsruhe, Germany) for pushing the liquids 20 times back and forth between the syringes, further named number of passes. Afterwards, a hydrophilic syringe filter was inserted between the syringes, see Figure 2. To ensure a reproducible production, a customized pneumatic membrane emulsification process was used for the second step (Reiner et al. 2023a). Both syringes were constantly kept at over 60°C with hot air streams to keep the dispersed phase liquid. The inlet pressure for the production of all suspoemulsions was 0.3 MPa. Suspoemulsions with droplets around 3.66 μm mimicking the droplet size of dairy cream (cream-sized-suspoemulsion (CSS)) were produced using hydrophilic syringe filter with a pore size of 10 μm (PSF Acrodisc, diameter = 25 mm, Pall Corporation, New York, USA) and 20 passes. Milk-sized-suspoemulsions (MSS) with small droplets around 1.2 μm were produced using hydrophilic syringe filter with a pore size of 0.8 μm (PSF Acrodisc, diameter = 25 mm, Pall Corporation, New York, USA) and 10 passes. After the emulsification process, all samples were cooled down at room temperature.

2.3 | Suspoemulsion Characterization

To achieve different solidity of the dispersed phases, different temperatures were used for the measurements. Measurement at 5°C resembles the cooled storage, at 20°C resembles the room temperature, at 37°C the maximum temperature in the oral cavity and at 50°C, for a cooked product when the dispersed phase for all suspoemulsions is completely liquid.

2.3.1 | Droplet Size Measurements

The droplet size distributions of emulsions were determined by a laser diffraction particle size analyzer in a flow measuring cell (Horiba LA-940, Retsch Technology, Haan, Germany) at the same day as they were produced with membrane emulsification. Emulsions were diluted prior to the analysis and measured three times at room temperature. The characteristic droplet $x_{50,3}$ and the span for showing the narrow droplet size distributions were determined. The distributions are monomodal and particles between 0.3 and 8.0 μm were measured. Regardless of which triglyceride is present in the dispersed phase, similar droplet sizes are achieved, as can be seen in Table 2. The shop-bought dairy cream had an $x_{50,3} = 3.552\,\mu m \pm 0.069$ with a span of 1.167.

2.3.2 | Polarized Microscopy

The behavior of emulsions was examined with a polarizing microscope (Eclipse LV100ND, Nikon, Shinagawa, Japan) equipped with an optically accessible temperature-controlled stage (LTS 420, Linkam Scientific, Tadworth, UK). The images were evaluated with regard to the crystallization of the differently sized

TABLE 2 | Droplet size distributions of the milk-sized-suspoemulsions (MSS) and the cream-sized-suspoemulsions (CSS). Medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG), a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) and pure butter fat (PBF) were used as the dispersed phase. To describe the emulsion the $x_{50,3}$ is shown, which is defined as the volume-based diameter of a sphere, at which 50% of the measured particles are smaller, and 50% are bigger. Also the span is given, which provides information about the width of an emulsion. Droplet sizes are the same, therefore an influence of the droplet size due to the different dispersed phases can be excluded.

	MSS			CSS		
	$x_{50,3}$/µm	Standard deviation/µm	Span/-	$x_{50,3}$/µm	Standard deviation/µm	Span/-
MCT	1.208	0.016	1.126	3.793	0.193	0.933
MCT/HCG	1.279	0.005	1.242	3.552	0.309	0.926
HCG	1.054	0.021	1.123	3.584	0.080	0.967
PBF	1.261	0.007	1.175	3.723	0.052	1.027

Note: MSS will be further referred to having an average droplet size of 1.2 µm, and CSS having an average droplet size of 3.66 µm.

TABLE 3 | Young's modulus for the different measurable triacylglycerides cylinders to compare the solidity of the dispersed phases. Medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG), a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) and pure butter fat (PBF) were measured at 5°C and 20°C.

	5°C		20°C	
	Young's modulus/MPa	Standard deviation/MPa	Young's modulus/MPa	Standard deviation/MPa
HCG	5.5419	0.3231	3.004	0.0729
PBF	2.6301	0.3164	0.0195	0.0004
MCT/HCG-mixture	2.2532	0.3085	0.5615	0.1124

drops. Partially crystalline droplets show with a green color in these images. The samples were prepared with object slides, cover slips and glue according to Abramov et al. (2016).

2.3.3 | Calorimetry Measurements

The melting and crystallization behavior of the triglycerides was analyzed using a double-furnace differential calorimeter DSC 8500 (Perkin Elmer Inc., Waltham, USA). The calorimeter was calibrated against indium ($T_M = 156.6$°C), water ($T_M = 0.000$°C) and n-decane ($T_M = -29.65$°C) at a rate of ±10 K/min. 10 mg of sample was loaded into aluminum pans. The pans were cooled to −5°C and after 2 min holding time heated to 60°C. After 2 min holding time, they were cooled back to −5°C. The total melting enthalpy ΔH_m was determined from the total area of the peak between T_{onset}, when the DSC signal leaves the baseline, and T_{end}, when it reaches the baseline.

2.3.4 | Young's Modulus

Uniaxial compression tests were performed on different triglyceride cylinders (8 mm height, 25 mm diameter) using a rheometer with parallel plates geometry (Haake Mars 60, Thermo Scientific, Karlsruhe, Germany), based on Liu et al. (Liu et al. 2024). For producing the triglyceride cylinders, HCG, the triglyceride mixture MCT/HCG and PBF were melted, solidified in the fridge and cut out with a metal tool. The cylinders were compressed at the temperatures 5°C and 20°C. At 37 and 50°C no Young's modulus is obtained. The tests were performed at a compression speed of 0.05 mm/s to 75% of initial height. The Young's modulus was extracted from the initial slope of the stress–strain curves within the strain region of approximately 0.05–0.10, see results in Table 3. Measurements were carried out in triplicates.

2.3.5 | Rheological Properties

For measuring the viscosity a double-gap measurement system (DG26.7SN47667, 3.8 mL per sample) on a rheometer (Physica MCR 101 rheometer, Anton Paar, Graz, Austria) was used. The viscosity was measured on the day the emulsion was produced. A total of 20 measuring points are recorded with a measuring point duration of 15 s. Shear rate starts at 1 s^{-1} and goes up to 1000 s^{-1}. The measurements were carried out in triplicates. The shear rate at 50 s^{-1} is used for the evaluation, as this is similar to the shear rates in the mouth (Wood and Goff 1973).

2.3.6 | Tribological Properties

The friction coefficient was measured at the four temperatures to obtain a friction profile for each suspoemulsion. The friction coefficient was measured using a customized tribology-cell with a glass ball-on-three-polymer-Pin geometry mounted on a Rheometer (Haake Mars 60, Thermo Scientific, Karlsruhe, Germany). The volume of 900 μL of each suspoemulsion was equilibrated at each temperature in the measurement cell before analysis. The measurement parameters were a 180 s duration in log mode, a constant applied force of 1 N, 1000 data points, a integration time of 2 s for each data point and a rotational speed varying from 0.001 to 100 mm/s (data shown: conditions in the oral cavity: 10–100 mm/s (Olivares, Shahrivar, and de Vicente 2019; Pondicherry, Rummel, and Laeuger 2018)). The friction coefficient was plotted versus rotational speed to achieve Stribeck curves. Every measurement was carried out in triplicates. One measurement existing out of five passes, first two passes were used for run-in, last three passes were averaged and used for evaluation.

The polymer-pins were produced with a customized polytetrafluoroethylene template in a temperature controllable vakuum-chamber using two-component resin (Polydimethylsiloxan, Sylgard 184 Silicone Elastomer Kit, Dow Europe GmbH, Wiesbaden, Germany). For the pins a Young's modulus of $E = 2.15 \pm 0.05\,\mathrm{N\,mm^{-2}}$ was measured with a pressure test.

3 | Results and Discussion

3.1 | Crystallization and Melting Behavior of Hydrogenated Coconut Glycerides of Bulk and in Suspoemulsion

The general microstructure of the droplets was assessed directly after production. Exemplary polarized light microscopy images at 5°C of HCG as bulk-phase, as CSS and as MSS are depicted in Figure 3a. The bulk phase appears green under polarized light, meaning it is mostly crystalline. Some droplets at the HCG-CSS image appear as green and yellow, meaning that most of the droplets were at least partially crystalline. In case of the HCG-MSS, droplets are small and not much color is detectible. Because submicron-sized particles are not visible under light microscopy, no conclusions about their solid fat content can be drawn. In Figure 3b exemplary melting thermograms from differential scanning calorimetry of HCG-bulk and HCG-MSS are compared. The y-axis of the diagrams is omitted as the analysis was performed qualitatively to identify the peak positions and compare them to the researched temperatures. With increasing temperature, the HCG-bulk melts at around 10°C and has its first endothermic peak at 17.9°C. The second peak is at 29.6°C and the third peak is at 35.5°C indicating that at 40.0°C the HCG-bulk is fully liquid. The dispersed phase of the HCG-MSS melts at around 15°C and has its first endothermic peak at 21.8°C. The second peak is at 31.9°C indicating that at 39.5°C the emulsified HCG is fully liquid. Both thermograms have an exothermic peak at 21.9°C (HCG-bulk) and 23.5°C (HCG-MSS). Peaks from the HCG-bulk are higher than from the HCG-MSS.

Microscopic images showed, that at 5°C the HCG-bulk is solid, HCG-CSS is at least partially solid. The thermograms indicate that at 5°C the HCG in bulk and MSS is crystalline, partially crystalline from around 10 to 40°C for HCG-bulk and around 15 to 40°C for HCG-MSS and fully molten with temperatures higher 40°C. The measurements from diffraction scanning calorimetry show, that even when not visible in polarized-light microscopic images, in MSS a phase transition of the triglycerides occurs. Crystallization of the HCG in bulk and in emulsion with microscopic imaging was as expected. With a submicron sized dispersed phase the crystallization is more complicate to analyze (Abramov, Ruppik, and Schuchmann 2016).

The solid fat content could not be quantified because of more complex fats and smaller droplets than shown in literature (Reiner et al. 2023a). Different Peaks in diffraction scanning calorimetry indicating different polymorphs that are build up out of different fatty acids in the triglyceride mixture (Márquez, Pérez, and Wagner 2013).

It is concluded that even if the particles are small and crystallization is less likely due to the low probability of nucleation, the dispersed phase is at least partially solid in CSS and MSS when measuring below 40°C.

3.2 | Influence of the Crystallized Dispersed Phase on Rheological Measurements

The viscosity (at shear rate $50\,\mathrm{s^{-1}}$) for the different MSS (30 wt% and a droplet size of around 1.2 μm) over the temperature is depicted in Figure 4. For comparison, the viscosity of the continuous phase and the PBF-MSS is shown. With rising temperature, all samples show the expected decrease in viscosity. The viscosity of the PBF-MSS at 5°C is slightly higher than of the other suspoemulsions. Besides that, the viscosity of the MSS is the same at each temperature, despite the differences in aggregate state

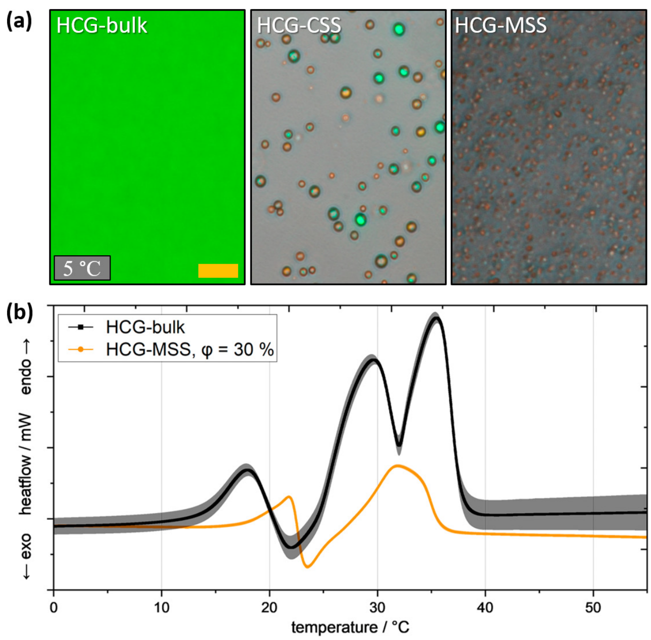

FIGURE 3 | (a) Polarization microscope images at 5°C of hydrogenated coco-glycerides (HCG) as bulk (left), as cream-sized-suspoemulsion (CSS) (middle) and as milk-sized-suspoemulsion (MSS) (right). Partially crystalline droplets show with a green color. When molten, the color intensity decreases. For smaller droplets, the green color is not visible anymore. Length of the scale bar is 20 μm and applies to all three images. (b) Differential scanning calorimetry thermograms by heating up with 10 K/min comparing HCG-bulk and HCG-MSS showing different endothermic melting peaks.

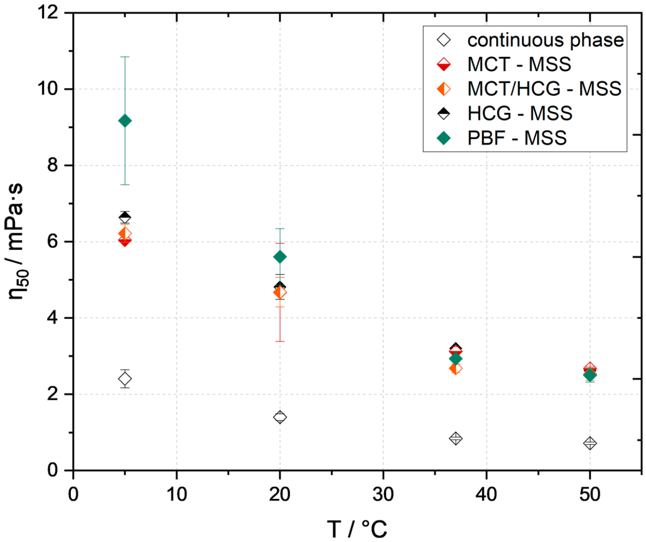

FIGURE 4 | Rheological measurements with viscosity over temperature at 50 s^{-1} of milk-sized-suspoemulsions (MSS) are depicted. For dispersed phase medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG) and a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) were used (half-filled squares). For comparison also pure butter fat milk-sized-suspoemulsions (PBF-MSS) (full filled squares) and continuous phase (empty squares) are shown. Viscosity of the MSS showed no differences, despite the differences of the aggregate states of the dispersed phase. Whole curves were measured and showed shear-thinning behavior as expected (see Appendix 1).

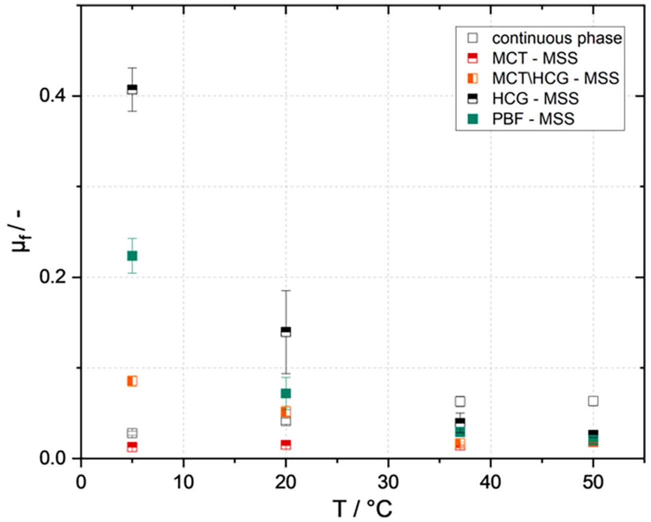

FIGURE 5 | Tribological measurements with friction coefficient over temperature at $v_r = 100$ mm/s of milk-sized-suspoemulsions (MSS) are depicted (for cream-sized-suspoemulsions see Appendix 2). For dispersed phase medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG) and a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) were used (half-filled squares). For comparison also pure butter fat milk-sized-suspoemulsions (PBF-MSS) (full filled squares) and continuous phase (empty squares) are shown. As the temperature rises, the dispersed phase of the different MSS melts and the friction coefficient decreases.

of the dispersed phase. The viscosity for all suspoemulsions is higher than for the continuous phase at each temperature.

At high dispersed phase content, the viscosity of the entire emulsion increases, which is consistent with the MSS shown (Köhler and Schuchmann 2012, 197). The decrease of viscosity with temperature is according to literature, see Arrhenius equation (Peleg, Normand, and Corradini 2012). At the same temperature, the dispersed phase did not influence the viscosity of the suspoemulsions.

Internal stresses are passed on to the dispersed phase via the continuous phase, which increases the viscosity and explains the difference of the continuous phase compared to the suspoemulsions. Whether the droplets are liquid or partially solid does not make any difference in these measurements. This is consistent with the relevant literature which states that relevant properties of food-grade emulsions cannot be described using rheology alone (Giasson, Israelachvili, and Yoshizawa 1997; Lee et al. 2003; Malone, Appelqvist, and Norton 2003; de Wijk and Prinz 2006). Since the forces on the droplets are relatively weak and the droplets small, the interfacial tension of the droplets is sufficient to make them non-deformable, even when they are liquid. When no deformation is to be expected, the only parameters influencing the viscosity are the droplet sizes and the dispersed phase ratio.

Rheological measurements at conditions relevant to oral consumption give no indication regarding the solidity of the dispersed phase, but with presence of dispersed phase viscosity can be adjusted.

3.3 | Influence of the Crystallized Dispersed Phase on Tribological Measurements

3.3.1 | Considerations of Milk and Dairy Cream Alternatives in Oral Conditions

The friction coefficient for the different MSS (30 wt% and a droplet size of around 1.2 µm) is plotted against the temperature in Figure 5. For the processing of liquid in the oral cavity there is a wide range of values in literature, for example 50 mm/s (Chojnicka-Paszun, Doussinault, and de Jongh 2014), below 150 mm/s (Chojnicka et al. 2008) and up to 200 mm/s (Hiiemae and Palmer 2003). Due to the wide range of speeds in literature and the data available, decision was made to compare the friction coefficients at $v_r = 100$ mm/s. The values were extracted from whole Stribeck curves. For comparison, the friction coefficients of the continuous phase is shown.

With increasing temperature the friction coefficient of the continuous phase is slightly increasing. The friction coefficients of the MCT-MSS do not change with the temperature and lie below those of the continuous phase. For the other three MSS the dispersed phase melts as the temperature rises, which results in decreasing friction coefficients. This can be seen, for example, in HCG-MSS, where the dimensionless friction coefficient decreases from 0.407 at 5°C to 0.02 at 50°C. Between 37°C and 50°C there are no significant difference in the measured friction coefficients for all MSS, since all dispersed phases are completely liquid.

The bulk triglyceride phase Young's moduli match the friction coefficients of the suspoemulsions—with decreasing Young's modulus the friction coefficient is decreasing. For example at 5°C HCG produces the highest measured $E=5.54\pm0.32$ MPa with the HCG-MSS causing the highest measured $\mu_f=0.407\pm0.024$, followed by PBF with the second highest $E=2.63\pm0.32$ MPa with the PBF emulsion causing the second highest $\mu_f=0.224\pm0.019$.

The range of the friction coefficient (20°C: $\mu_f=0.015$–0.14) is as expected. For example, a emulsion systems with a high amount of fat in dispersed phase have a friction coefficient around $\mu_f\approx0.06$ (cheese sauce) and $\mu_f=0.14$ (choco cream with saliva) (Pondicherry, Rummel, and Laeuger 2018). At 37°C the friction coefficient ($\mu_f=0.014$–0.039) is in the same range when compared to a whey protein isolate solution with a friction coefficient of 0.012 (Kew et al. 2021) and different thickened creams at 35°C with $\mu_f=0.01$–0.06 (Selway and Stokes 2013). It is also consistent with the literature that the friction coefficients of all MSS at 50°C is lower than the continuous phase, because the emulsified oil droplets act as a lubricating film (Liu et al. 2015; Nguyen, Bhandari, and Prakash 2016). The higher friction coefficient at 5°C for the MSS with partially crystalline dispersed phase ($\mu_f=0.085$–0.407) can also be seen for choco cream without saliva with $\mu_f\approx0.26$ (Pondicherry, Rummel, and Laeuger 2018), due to crystalline triglyceride particles that have a melting range of 17°C–39°C. Different chocolate nut pastes at 36.5°C showed a friction coefficient around $\mu_f\approx0.1$ (Principato et al. 2022). Examples for a comparable system with different temperatures causing different solidity in dispersed phase were not found in literature. However, the use of particles in emulsions were researched before. The partially crystallized dispersed phase in the MSS did not reduce the friction like the carrageenan- or whey-protein suspensions (Garrec and Norton 2013; Sarkar et al. 2017). They also didn't behave like the spherical particles researched by Rudge et al. (2021) presumably due to the difference that the minimum investigated size of 100 μm is significantly larger than the maximum possible size of the crystallized dispersion phase of 1.2 μm. Also the partially crystalized phase did not behave like the microparticulated whey proteins which did not change the friction coefficient and could imitate liquid dairy fat (Olivares, Shahrivar, and de Vicente 2019). The partially crystallized dispersed phase in the MSS seem to behave like the native starch particle suspensions described by Liu et al., where the friction coefficient could be reduced by gelling the dispersed phase (Liu et al. 2016a). Rodruigez et al. explained in their model system that the dispersed triglyceride phase in the emulsion coalesces and coats the interacting surfaces, thereby controlling lubrication in thin-film conditions. The authors also observed that particle entrainment increases the friction (Rodrigues et al. 2021). The suspoemulsions with the differences in Young's modulus of the dispersed phase behave the same way. Thin film lubrication takes place, with the liquid triglyceride phase lubricating and reducing the friction whereas the solid triglyceride phase acts like particles and thus through particle entrainment friction is increased. Chojnicka-Paszun et al. also found that the friction coefficient increased from 0.3 to 0.75 in xanthan solutions with added protein particles (Chojnicka-Paszun, Doussinault, and de Jongh 2014). The increase in friction of the continuous phase can be explained by the unfolding of the proteins when the temperature rises.

The results underline the importance of the aggregate state of the dispersed phase on the tribology. When measuring above the melting range of HCG and PBF ($T_M>37$°C) all MSS show the same measurement values. Below the melting point, the droplets are expected to be crystalline and partially crystalline and the friction coefficients differ according to the measured Young's moduli. The higher the solid fat content is expected from the chemistry of the dispersed phase, the higher the values for the friction coefficients are.

3.3.2 | Behavior of Suspoemulsions Based on Stribeck Curves

For better understanding the friction coefficients for the samples from Figure 5 are plotted against the rotational speed, resulting in Stribeck curves at four different temperatures in Figure 6. The progression of the MCT-MSS Stribeck curve is sigmoidal at all temperatures, first constant over the rotational speed, with a decrease to lower friction coefficients at higher rotational speed. The progression of the friction coefficient of the HCG-MSS is very different. At 5°C the friction coefficient increases as the rotational speed increases, at 20°C the curve also increases and only at 37°C and 50°C is the progression of the Stribeck curve similarly sigmoidal as with the MCT-MSS. This also applies to the PBF-MSS, where the Stribeck curve rises at low temperatures and then follows a similar progression as the MCT-MSS at higher temperatures. The change in the curve due to the temperature can also be seen in the MCT/HCG-MSS.

Friction coefficients of all MSS despite different dispersed phases is decreasing with increasing temperature. The progression of the MCT-MSS is the same at all four temperatures, whereas the progression of the HCG-MSS rises at low temperatures and decreases at high temperatures.

The MCT-MSS for all temperatures behave like milk in literature (Nguyen, Bhandari, and Prakash 2016). First, the friction is constant with increasing rotational speed, then it decreases in a sigmoidal curve. In literature this is explained with the increasing gap caused by the hydrodynamic lift that occurs between the contact areas of glass ball and polymer pins, which reduce the friction (Prakash, Tan, and Chen 2013). The strong rise of the friction coefficients at 5°C of the HCG-MSS and of the PBF-MSS, when the droplet crystallize is not yet described in literature. Olivares et al. measured skimmed milk at 25°C and 37°C and found no difference in the general curve progression. However, at 25°C and 37°C there is only a small amount of crystalline triglyceride, especially in the relatively high temperature range for butter fat (Olivares, Shahrivar, and de Vicente 2019).

Explanation for the increasing friction coefficient with decreasing temperature is most likely the crystallization of the different triglyceride mixtures. The higher the solid fat content is expected from the utilized triglycerides, the higher the friction coefficient was measured. However, the amount of crystallized particles cannot be qualified with solid fat content like in previous papers (Abramov, Ruppik, and Schuchmann 2016; Reiner et al. 2023a) because of partially crystallized droplets. Based on the differential scanning calorimetry, the melting ranges for the bulk triglycerides and the measured Young's moduli, the highest solidity of the dispersed phase is the HCG-MSS, then the PBF- and MCT/HCG-MSS and the lowest in

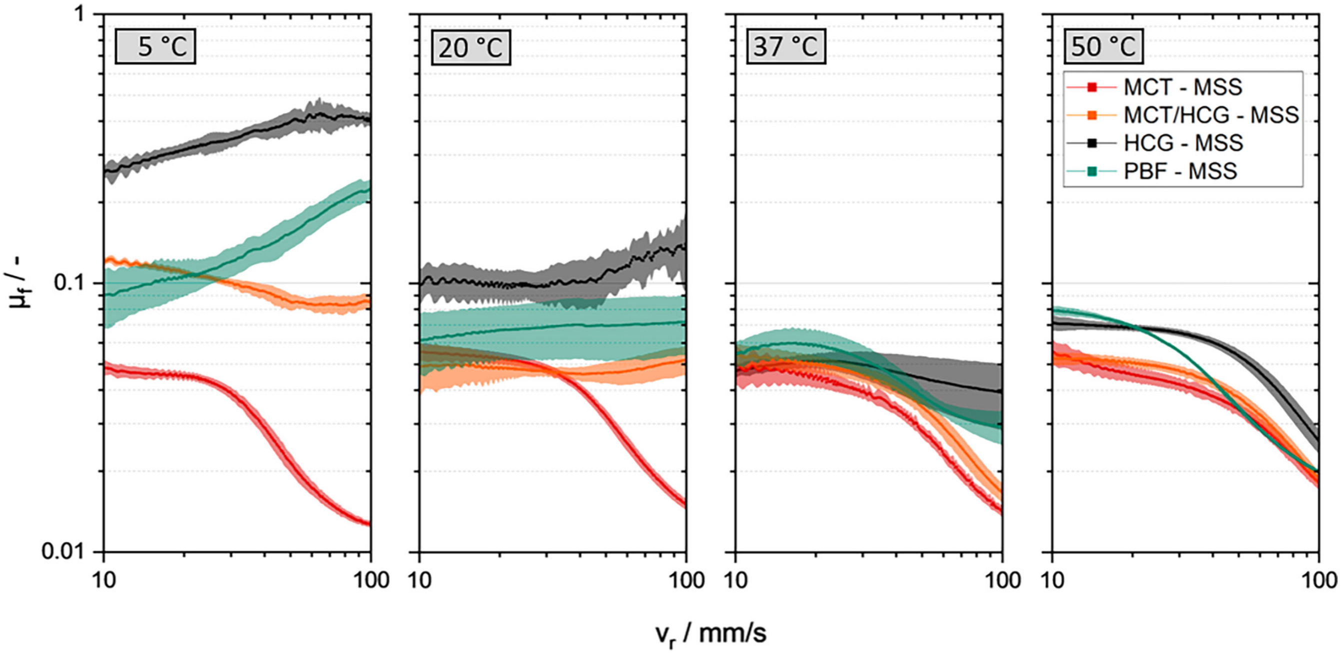

FIGURE 6 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for suspoemulsions with dispersed phase out of medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange) and pure butter fat (PBF, green) are depicted at the temperatures left: 5°C, middle left: 20°C, middle right: 37°C, right: 50°C. Friction coefficients of all milk-sized-suspoemulsions (MSS) despite different dispersed phases is decreasing with increasing temperature. The progression of the MCT-MSS is the same at all four temperatures, whereas the progression of the HCG-MSS rises at low temperatures and decreases at high temperatures. Curves with $v_r = 0.01–100$ mm/s are depicted in Appendix 3.

the MCT-MSS. The data show, that the friction coefficient increases as the solidity of the dispersed phase increases. This is also shown by the progression of the curve, which can be assigned to an area of the Stribeck-curve, as depicted in Figure 7.

At 5°C and with the HCG-MSS, the partially solid droplets in the measuring gap probably accumulate with increasing rotational speed and cause the Stribeck curve to rise. This is also the case with the PBF-MSS, albeit at lower friction coefficients. This suggests that there is a high solidity, which fits to the measured Young's moduli ($E_{HCG,5°C} = 5.54 \pm 0.32$ MPa; $E_{PBF,5°C} = 2.63 \pm 0.32$ MPa) and the values of the solid fat content in the literature (65% for pure fat butter in bulk). The fact that the friction coefficients of the HCG-MSS are higher could be due to the narrower melting range or the different distribution of the crystals within the droplets. On one hand the general height of the friction coefficients of the MCT/HCG-MSS is similar to the PBF-MSS. This corresponds to the Young's moduli of the bulk triglycerides with $E_{MCT/HCG,5°C} = 2.25 \pm 0.31$ MPa and $E_{PBF,5°C} = 2.63 \pm 0.32$ MPa. On the other hand the curve progression of the MCT/HCG-MSS can be assigned to area II, whereas the PBF-MSS can be assigned to area I. It remains to be investigated whether the shape of the Stribeck curve plays a role in the oral sensation of emulsions, or whether only the level of the friction coefficient is decisive. The MCT-MSS behave as in the literature and can be clearly assigned to area II. At 20°C, the friction coefficients of all MSS are lower, which can be explained by the lower crystallinity and thus the lower solidity of all MSS. The HCG-, the PBF- and the MCT/HCG-MSS are largely constant and thus correspond to typical Stribeck curves in the literature and are assigned to area II. It is possible that the partially crystalline droplets in the dispersed phase are soft enough due to the lower crystallinity to be drawn through the gap with increasing rotational speed and thus no longer accumulate. At 37°C the solidity has decreased further and this can also be seen in the curves as PBF-MSS and MCT/HCG-MSS can be assigned to area II. HCG-MSS can be assigned in between area I and II—the dispersed phase is still the most solid. However, the order of the friction values at v_r still corresponds to the order of the assumed solidity. At 50°C, all curves can be assigned to area II. At 50°C the dispersed phase of all MSS is liquid, the droplets in the gap can be deformed, do not accumulate and the friction between the surfaces is reduced.

With oral tribology, a higher friction coefficient can be determined with increasing solidity of the dispersed phase. This was caused by the crystallinity of the different triglyceride mixtures used. Therefore, if all crystals are melted by increasing the temperature, the crystallinity of the dispersed phase decreases and the friction of the systems is equalized. By mixing HCG and MCT the Stribeck curves of MCT/HCG-MSS can mimic the Stribeck curves of PBF-MSS. A method for researching the tribological behavior of different aggregate state dispersed phase suspoemulsions was found and is coherent with existing literature.

3.4 | Influence of the Droplet Size on the Tribological Behavior of Suspoemulsions

In Figure 8 the Stribeck curves of MSS with a droplet size of around 1.2 µm are compared to the Stribeck curves of CSS

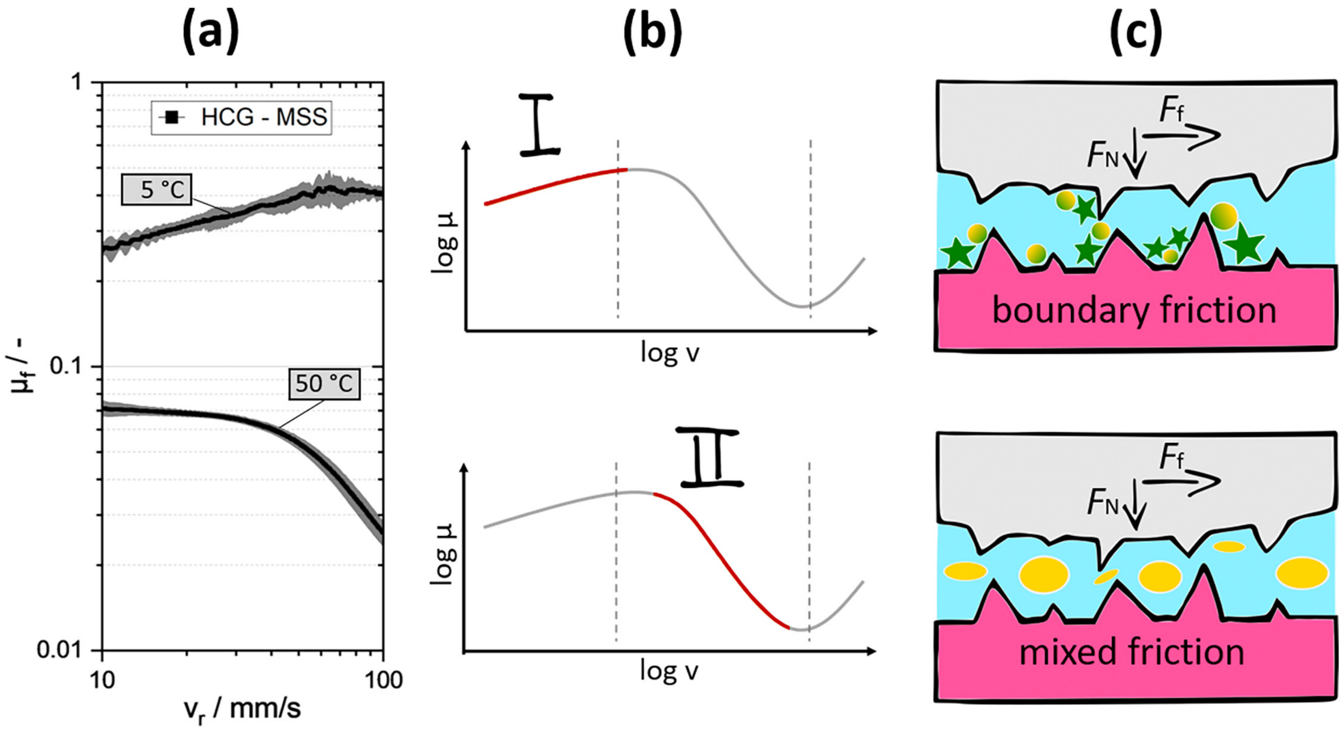

FIGURE 7 | (a) Exemplary Stribeck curves of milk-sized-suspoemulsions with hydrogenated coco-glycerides as dispersed phase (HCG-MSS) at 5°C and 50°C (b) Schematic Stribeck curves to illustrate how the HCG-MSS can be assigned to the respective areas. At 5°C, the friction is analogous to the boundary friction area I. At 50°C, the friction is analogous to the mixed friction area II. (c) Possible schematic explanation why the HCG-MSS behave different. At 5°C the dispersed phase is partially crystalline, therefore the droplets and particles can coalesce, aggregate and accumulate in the gap. This would lead to physical changes of the emulsion. The dispersed phase with high solidity acts as a new boundary and therefore ensures the high friction which is seen as boundary friction area I in the Stribeck curves. At 50°C the dispersed phase is fully liquid, deformable, will not accumulate and resulting in less friction which is seen as mixed lubrication area II in the Stribeck curves. To support the theory, exemplary Stribeck curves were recorded up to rotational speed of 300 mm/s, showing the start of area II for HCG-MSS and the start of area III for medium chain triglycerides-MSS (see Appendix 4). This explanatory approach based on solidity provides a good tool for interpreting measurement curves, regardless of whether the differences are caused by changes in temperature or choice of triglycerides.

(droplet size around 3.66 μm) known from Section 3.3. The progression of the MCT-MSS Stribeck curve is the same as with the MCT-CSS at low (5°C) and high (50°C) temperature. Also the progression of the MCT/HCG-MSS and MCT/HCG-CSS are analogous, staying constant over the rotational speed. At 5°C, the friction coefficients of the MCT/HCG-MSS and the MCT-MSS are slightly lower than the corresponding MSS. At 50°C, no differences between the different droplet sizes can be identified.

Stribeck curve progressions of MSS and CSS are the same. At low temperature the dispersed phases vary in solidity and big differences of the curve progressions are visible, at high temperature the dispersed phase of all CSS is melted and therefore the curve progressions of all CSS are the same. Within the examined range, the influence of the droplet size on the friction coefficient is neglectable.

For CSS the microscopy images showed the crystallization that occurs at 5°C. The Stribeck curves at 5°C also showed, that the HCG-MSS and HCG-CSS behaving similarly, indicating that the solidity of the dispersed phases seems to be the same. The same behavior in oral tribology indicates, that the small droplets behave like the big droplets. Because the disperse phase content of the MSS and CSS is the same, the number of droplets in the MSS is much higher. The solidity of the dispersed phase is therefore important for the progression of the Stribeck curve, and not the number of droplets. With oral tribology the crystallinity of the disperse phase can be validated, even with small droplets in dispersed phase.

MSS was also measured with the lower dispersed phase content of 3.5% *with the aid of dilution after production to have a system close to milk or milk alternatives. The data confirms the discussed trends of MSS with* $\varphi = 30\%$ and does not add additional findings. *The graphs are therefore shown in* Appendixes 5 and 6. The MSS with $\varphi = 3.5\%$ showed in total higher friction coefficients than the MSS with $\varphi = 30\%$, since less triglyceride means less lubrication. It is indicated that oral tribology can also be applied for small dispersed phase contents.

3.5 | Application of the Findings of the Suspoemulsions to Dairy Cream

In order to compare the behavior of the plant protein stabilized suspoemulsions produced with membrane emulsification in this work with dairy cream, the Stribeck curves of CSS and dairy cream are plotted in Figure 9. At 5°C the friction coefficient of dairy cream is as high as the MCT/HCG-CSS and constant over increasing rotational speed. At 50°C the Stribeck curve of the dairy cream and the CSS are both decreasing with increasing

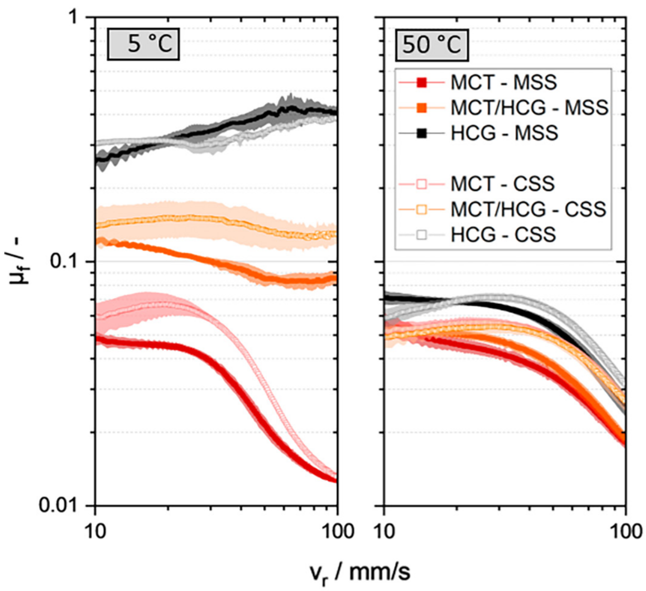

FIGURE 8 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for milk-sized-suspoemulsions (MSS, filled squares) and cream-sized-suspoemulsions (CSS, empty squares) with medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange) as dispersed phase at the temperatures 5°C (left) and 50°C (right). The friction coefficients and curve progressions of the CSS and MSS are similar.

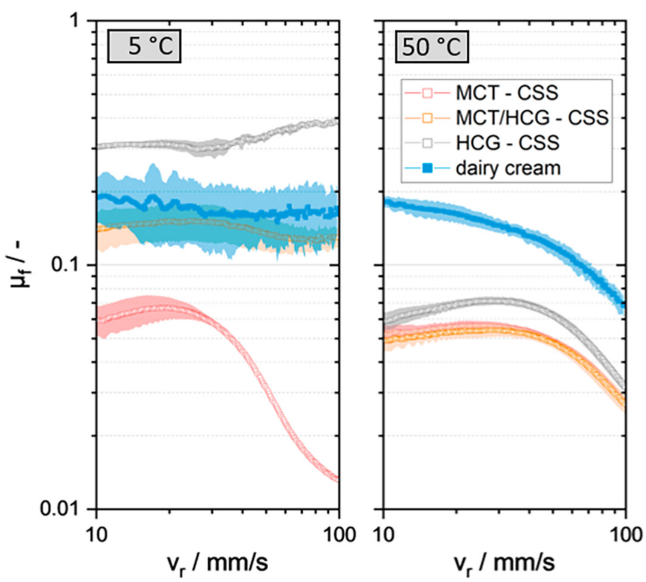

FIGURE 9 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for dairy cream (filled squares, blue) and cream-sized-suspoemulsions (CSS, empty squares) with medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange) as dispersed phase at the temperatures 5°C (left) and 50°C (right). At 5°C the behavior of the MCT/HCG-CSS and dairy cream are similar. At 50°C the progression of the Stribeck curve is similar, with all emulsions decreasing, however dairy cream having a higher friction than all CSS.

rotational speed, however the progression of the Stribeck curves is at much higher friction coefficients.

The difference of the friction coefficients at high temperatures may be because of the additional components in dairy cream. Additionally to the stabilizing whey proteins, casein and dissolved lactose are present in the dairy cream, increasing the viscosity roughly four times compared to the suspoemulsions (see Appendix 7). In other works, with different polysaccharides changes in friction coefficient were observed (Chojnicka-Paszun, Doussinault, and de Jongh 2014). At 35°C Selway and Stokes had a similar friction coefficient with cream ($\varphi = 13\%$) (Selway and Stokes 2013), however, there the Stribeck curve is assigned to area I. Also the difference might be because of casein micelles. Fan et al. that is researched the influence of the casein whey protein ratio (Fan et al. 2021). They suggest that under pH neutral conditions, interaction of casein with saliva leads to increased friction. Ji et al. found, that addition of large particles like casein micelles had limited effect on the lubrication properties of emulsions, independently of the lubrication mechanism (Ji et al. 2022).

At 5°C the MCT/HCG-CSS and the dairy cream are constant over rotational speed and both can be assigned to area I, boundary friction. At 50°C the dispersed phase is completely molten, which can be seen as Stribeck curves that can be assigned to mixed friction area II.

The behavior of the curve profiles of the mixture is analogue to dairy cream, indicating a similarity to real application products. When comparing the Stribeck curves of the dairy cream with those of the plant-based fat suspoemulsions, different plant-based fats could be mixed, to mimic the friction profile of milk products in plant-based alternatives.

4 | Conclusion

The oral tribology of milk and milk alternatives with liquid oil droplets differ significantly from products with solid triglyceride particles. It was demonstrated that adapting the solidity of the dispersed phase by replacing liquid tiglycerides with blends of plant-based triglycerides could mimic the friction coefficient of pure butter fat. While the rheological properties remained unaltered by the change of the triglyceride mixtures, the presented tribology method could detect clear differences. We showed how suspoemulsions with different solidity in the dispersed phase gave different progressions of the Stribeck curves. At low temperature (5°C) a high friction was caused by the high solidity of the dispersed phase, caused by butter or plant fat. At higher temperatures (37 and 50°C) and mouth-typical processing speeds (10–100 mm/s), a similar friction ($\mu_f = 0.02$) was found for all suspoemulsions regardless of the melting point of the dispersed phase. Therefore with oral tribology the occurrence of crystalline dispersed droplets can be detected. This was found for the typical droplet sizes of milk ($x_{50,3} = 1.2\,\mu$m) and dairy cream ($x_{50,3} = 3.66\,\mu$m). In comparison with dairy cream, the correlation with the Stribeck curves in the literature could also be demonstrated.

The suspoemulsion with pasty dispersed phase (MCT/HCG) had comparable absolute values with the pure butter fat suspoemulsion, but did not behave in the same range. It remains to be investigated to what extent the shape of the friction curve plays a role in the perception in the mouth, or whether the absolute value of the friction coefficient plays a greater role.

An investigation such as the one conducted in this study has not yet been carried out. There have been isolated studies on particles in the dispersed phase, but these do not consist of fat but of proteins, microgel particles, starch and sugars or glass. The study was able to demonstrate how sensitive the measuring device is for investigating emulsions with a partially crystalline dispersed phase, varied with triglyceride formulation and temperature.

Author Contributions

Conceptualization, P.R.S. and N.L.; methodology, P.R.S.; formal analysis, P.R.S. and L.L.; investigation, P.R.S. and L.L.; resources, H.P.K. and N.L.; data curation, P.R.S., and L.L.; writing – original draft preparation, P.R.S.; writing – review and editing, H.P.K. and N.L.; visualization, P.R.S.; funding acquisition, N.L.

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Ethics Statement

The authors have nothing to report.

Consent

Written informed consent was obtained from all participants.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data available on request from the authors.

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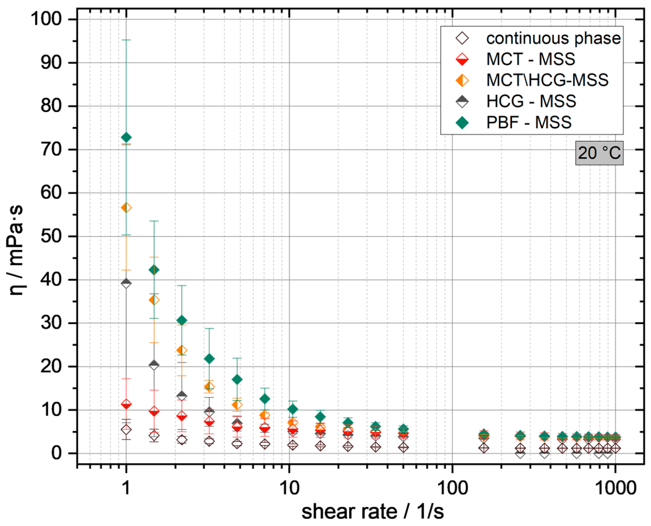
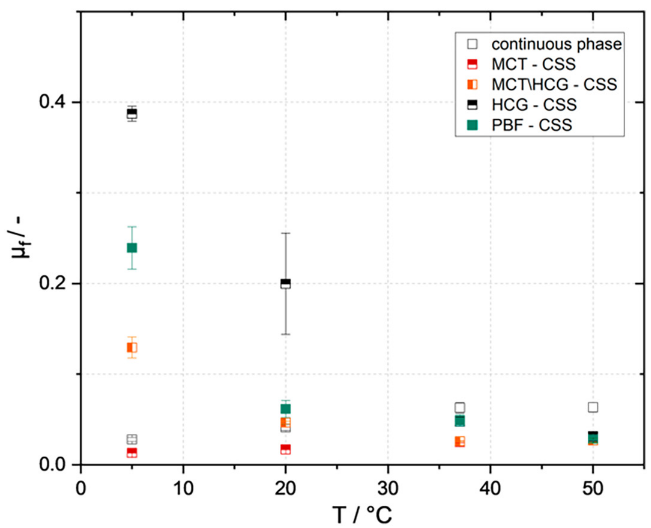
Appendix A

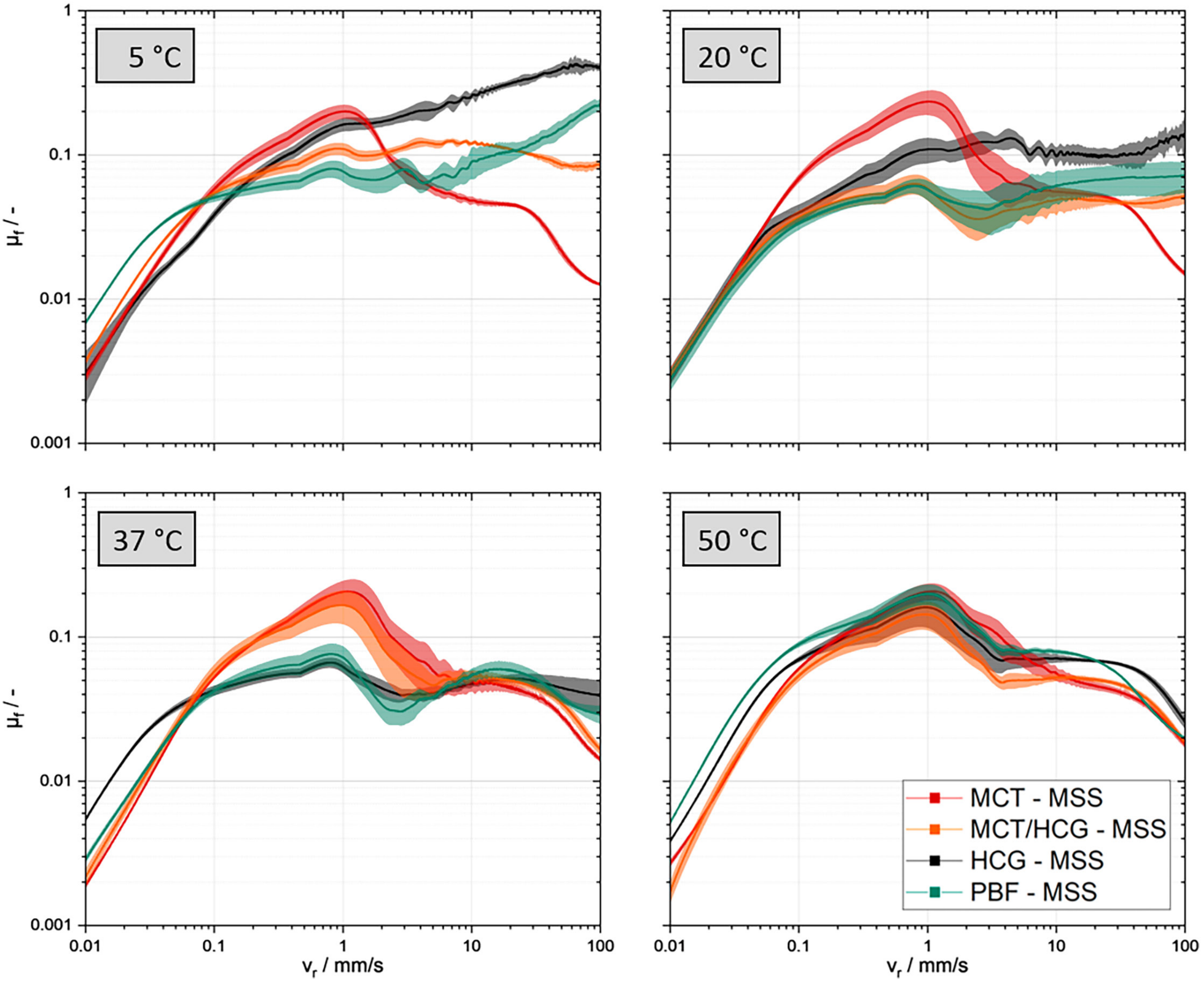
Appendices



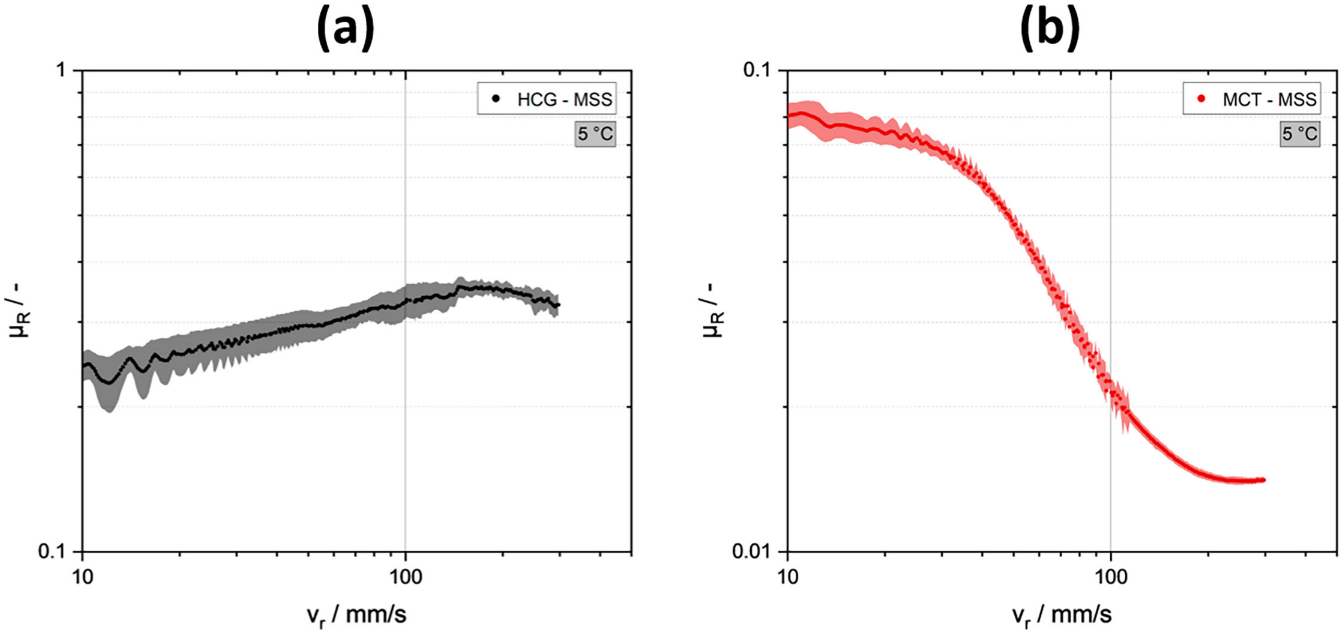
APPENDIX 1 | Exemplary rheological measurements with viscosity over shear rate at 20°C of milk-sized-suspoemulsions (MSS) are depicted. For dispersed phase medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG) and a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) were used (half-filled squares). For comparison also pure butter fat milk-sized-suspoemulsions (PBF-MSS) (full filled squares) and continuous phase (empty squares) are shown. With increasing shear rate, the viscosity decreases in all suspoemulsions, showing shear-thinning behavior as expected.



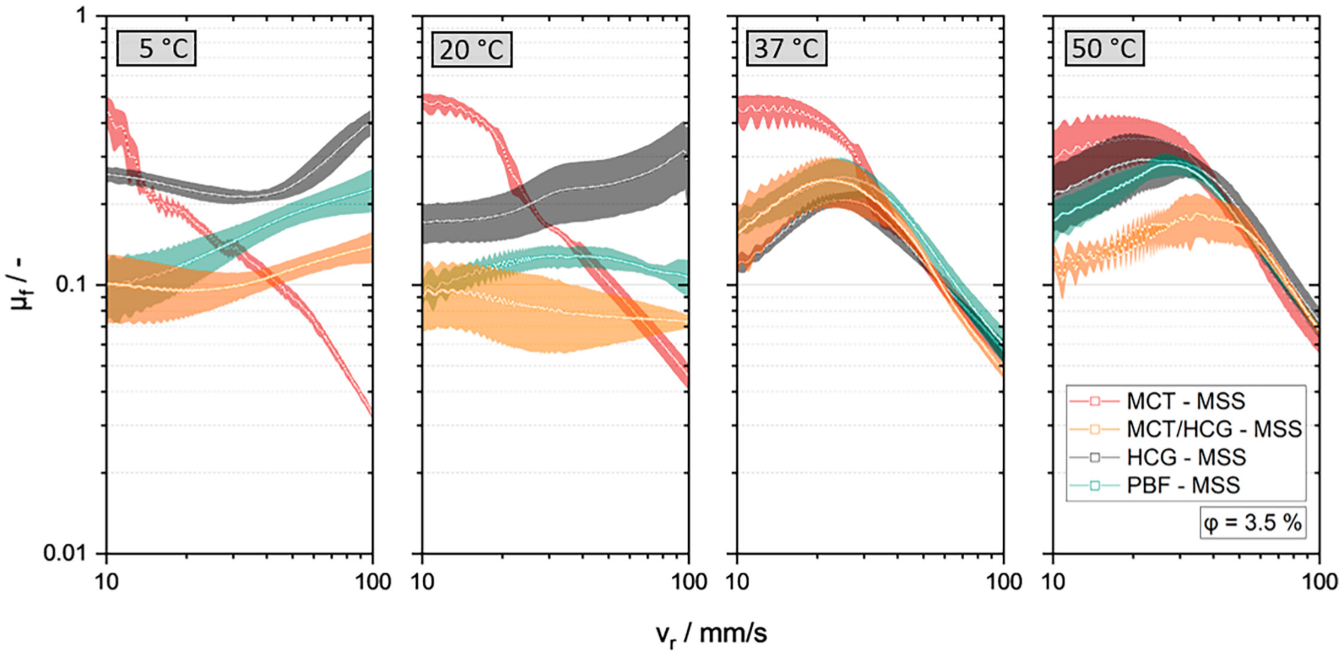
APPENDIX 2 | Tribological measurements with friction coefficient over temperature at $v_r = 100$ mm/s of cream-sized-suspoemulsions (CSS) are depicted. For dispersed phase medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG) and a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) were used (half-filled squares). For comparison also pure butter fat milk-sized-suspoemulsions (PBF-MSS) (full filled squares) and continuous phase (empty squares) are shown. As the temperature rises, the dispersed phase of the different CSS melts and the friction coefficient decreases. Whole Stribeck curves were measured (data not shown).



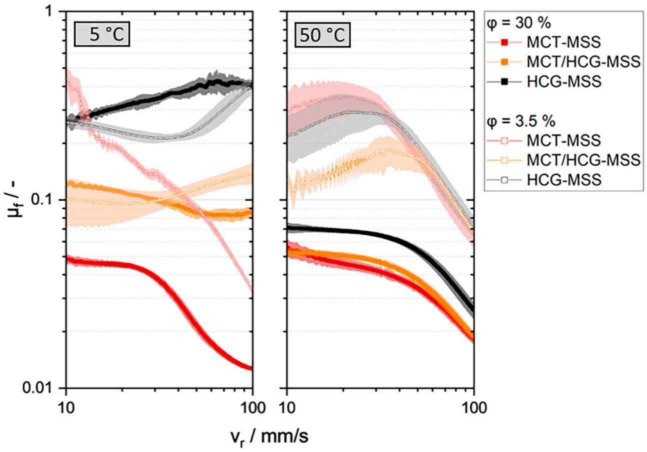
APPENDIX 3 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for suspoemulsions with dispersed phase out of medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange) and pure butter fat (PBF, green) are depicted at the temperatures left: 5°C, middle left: 20°C, middle right: 37°C, right: 50°C. With increasing solidity of the dispersed phase of the milk-sized-suspoemulsions (MSS) the friction coefficients are increasing with increasing rotational speed.



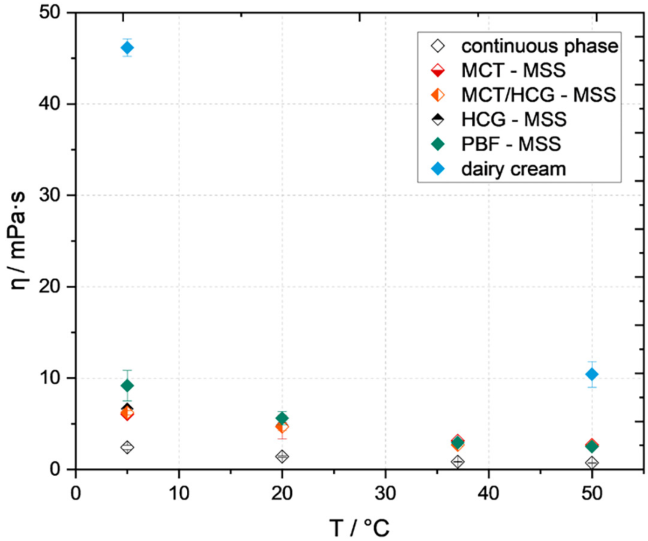
APPENDIX 4 | Friction coefficient over rotational speed at 5°C of (a) a hydrogenated coco-glycerides milk-sized-suspoemulsion (HCG-MSS) and (b) a medium chain triglyceride milk-sized-suspoemulsion (MCT-MSS) up the maximum possible rotational speed (300 mm/s). A maximum has been reached in the Stribeck curve progression of the HCG-MSS, indicating that at around 200 mm/s with decreasing friction coefficients at around 200 mm/s mixed friction area II starts to form. With the MCT-MSS Stribeck curve the minimum is reached at around 200 mm/s indicating that hydrodynamic lift area II starts to form.



APPENDIX 5 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for suspoemulsions with a dispersed phase content of φ = 3.5%. The dispersed phase consists of medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange) and pure butter fat (PBF, green). The Stribeck curves are depicted at the temperatures left: 5°C, middle left: 20°C, middle right: 37°C, right: 50°C. The progression of the MCT-MSS is the same at all four temperatures, whereas the Stribeck curves of the HCG-MSS rises at low temperatures and decreases at high temperatures.



APPENDIX 6 | Tribological measurements with friction coefficient over rotational speed depicted as Stribeck curves for milk-sized-suspoemulsions with a dispersed phase content of $\varphi = 30\%$ (filled squares) and milk-sized-suspoemulsions (MSS) with a dispersed phase content of $\varphi = 3.5\%$ (empty squares) are shown. The dispersed phase consists of medium chain triglycerides (MCT, red), hydrogenated coco-glycerides (HCG, black) and a mixture out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG, orange). The Stribeck curves are shown at the temperatures 5°C (left) and 50°C (right). At 50°C the MSS with $\varphi = 3.5\%$ showed much higher friction coefficients than the MSS with $\varphi = 30\%$, caused by reduced lubrication due to fewer triglycerides present. At 5°C the progressions of the Stribeck curves of the MSS with $\varphi = 3.5\%$ are in general very similar to the MSS with $\varphi = 30\%$. The influence of the solidity on the Stribeck curves can also be seen with suspoemulsions with dispersed phase contents of $\varphi = 3.5\%$.



APPENDIX 7 | Rheological measurements with viscosity over temperature at $50\,s^{-1}$ of milk-sized-suspo-emulsions (MSS) are depicted. For dispersed phase medium chain triglycerides (MCT), hydrogenated coco-glycerides (HCG) and a combination out of medium chain triglycerides and hydrogenated coco-glycerides (MCT/HCG) were used (half-filled squares). For comparison pure butter fat milk-sized-suspoemulsions (PBF-MSS) (full filled squares), continuous phase (empty squares) and dairy cream (full filled blue squares) are shown. Viscosity of the dairy cream at 5 and 50°C is higher than of any suspoemulsion.