Effect of Substituting Sunflower Oil with Starch-Based Fat Replacers on Sensory Profile, Tribology, and Rheology of Reduced-Fat Mayonnaise-Type Emulsions

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Abstract

This study investigates the effect of substituting sunflower oil with two starch-based fat-replacers on the rheological and lubricating properties, as well as the sensory properties of reduced-fat mayonnaise-type emulsions. Lipid-modified maize starch fat replacers containing amylose–lipid complexes, maize starch with 1.5% stearic acid and maize starch with 2% monoglyceride, are used to formulate reduced-fat mayonnaise-type emulsions at 0% (full-fat control), 50%, 80%, and 98% level of oil replacement. Reduced-fat emulsions containing starch/monoglyceride are rated similar ($p > 0.05$) to the full-fat mayonnaise at all the oil replacement levels in terms of smoothness, creaminess, melting, and mouth-coating. They also have similarities in terms of thickness and easy-to-swallow sensory attributes, up to a 50% substitution level. For the corresponding starch/stearic acid emulsions, the smoothness, thickness, creaminess, and mouth-coating attributes are rated lower while the melting and easy-to-swallow attributes are rated higher than for the starch/monoglyceride emulsions. In general, all the reduced-fat emulsions exhibit good lubrication. The ability of the reduced-fat emulsions to support the highly viscous structure provided by the presence of amylose–lipid complexes in the fat replacers is better for the starch/monoglyceride fat replacer than for the starch/stearic acid fat replacer.

1 Introduction

Awareness of the adverse health effects associated with overconsumption of fat has led to the development of reduced-fat products in the food industry. However, fat present in most foods provides unique texture, flavor, and aroma to the food,[1, 2] which is difficult to achieve in the reduced-fat products that often fail to deliver an expected food experience.[3]

Fat reduction through the use of functional fat replacers that would provide the desirable sensory properties would increase the acceptability of the reduced-fat products and contribute to a more balanced diet and well-being of the population. In the formulation of low-fat emulsions, researchers employ non-fat ingredients such as starches, gums, and proteins, to replace some quality attributes that are lost when fat is removed.[4] Identifying the ideal fat replacer that can mimic the multi-functional roles played by fat in food emulsions remains a challenge for the food industry, and research is ongoing. Popular fat replacers are starches that are chemically modified to improve their functionality.[5, 6] However, concerns about consumer and environmental safety related to the use of chemicals,[7] has encouraged the development of ‘clean label’ starches[8] prepared by a novel process that is employing approved food ingredients and additives to modify starch. In this study, such lipid-modified maize starches (maize starch modified with stearic acid or with monoglyceride) were used as fat replacers in mayonnaise-type food emulsions.

Mayonnaise is a semi-solid, oil-in-water emulsion generally described as a high-fat and high-calorie food,[9, 10] that traditionally contains 70–80% fat.[11] The production and quality characteristics of low-calorie mayonnaise emulsions have been studied extensively at different levels of fat reduction, but the challenge of not compromising the desirable sensory properties continues to be critical in the food industry.

Mayonnaise-emulsions with fat replacement levels up to 50% were formulated with fat mimetics based on whey protein isolate and low-methoxy pectin[12] and on modified arrowroot starch.[13] In the related research, reduced-fat mayonnaise was formulated by replacing part of the oil with gelatinized rice starch and xanthan gum, and the effect of their inclusion on rheological properties was investigated.[9] Stable mayonnaise-type emulsions were prepared by replacing up to 30% fat, which resulted in a 23% lower energy content in comparison to full-fat mayonnaise while the emulsions exhibited similar rheological properties as a commercial reduced-fat mayonnaise. In the formulation of reduced-calorie emulsion-based sauces and dressings,[14] demonstrated the effectiveness of using microparticulated whey protein in combination with polysaccharides to formulate emulsions with appearance and consistency similar to their full-fat versions.

The amount of oil substituted by a fat replacer is an important quality characteristic of the low-fat mayonnaise produced. Products with oil substitution levels as high as 80% by teff starch modified with 1.5% stearic acid were studied by Teklehaimanot et al.[15] in their research. They found that replacing sunflower oil at 50% and 80% levels with stearic acid-modified maize starch produced low-calorie mayonnaise type emulsions with higher viscosity and smaller oil droplets compared to full-fat mayonnaise. All the low-calorie mayonnaise type emulsions showed shear-thinning behavior and were more stable to freeze-thaw cycles and high-temperature storage than full-fat mayonnaise. However, the effect of such substantial oil replacement by maize starch/stearic acid on the sensory profile of the resulting low-calorie mayonnaise type emulsions was not researched. Also, no research was conducted on the tribological properties of these emulsions (i.e., thin layer lubrication) that affect mouthfeel and texture perception during oral processing, while these are not fully determined by bulk rheology.

In a follow-up research, the sensory, tribological and rheological properties of two lipid-modified starch-based fat replacers containing amylose–lipid complexes (maize starch with 1.5% stearic acid and maize starch with 2% monoglyceride) were investigated.[16] The lipid-modified starches were non-gelling and exhibited fat-like properties such as good lubricity, glossiness, smoothness, and creaminess. This study therefore investigates the impact of substituting sunflower oil with the two lipid-modified starch-based fat replacers (maize starch modified with 1.5% stearic acid and maize starch modified with 2% monoglyceride) on the lubricating properties of the reduced-fat mayonnaise-type emulsions in relation to their mouthfeel textural sensory attributes and rheological properties.

2 Experimental Section

2.1 Materials

Potential fat replacers used for the production of the reduced-fat emulsions were produced by modifying maize starches with stearic acid and monoglyceride according to the method by D'Silva et al.[17] Commercial white maize starch (Amyral) was obtained from Tongaat Hullet (Edenvale, South Africa). The proximate composition of the maize starch was approximately 112 g kg $^{-1}$ moisture, 4.5 g kg $^{-1}$ (db) protein, and 1.5 g kg $^{-1}$ (db) crude fat, and 250 g kg $^{-1}$ amylose on a starch basis. Stearic acid (CAS No: 57-11-4) and glycerol monostearate [>90% distilled monoglyceride (CAS No: 31566-31-1)], the monoglyceride used in the study, were obtained from Merck (Pty) Ltd. (Modderfontein, South Africa) and Danisco (now DuPont, Grindsted, Denmark), respectively. Spray-dried egg yolk was purchased from Sunspray Food Ingredients (Pty) Ltd. (Johannesburg, South Africa). Potassium sorbate and sodium benzoate were purchased from Merck Chemicals (Pty) Ltd. (Midrand, South Africa). All other ingredients (sunflower oil, vinegar (5% acidity), sugar, salt, and mustard powder) were purchased from a local supermarket. A commercial citrus-base fat replacer (Citri-Fi 100 FG) was provided by Danlink Ingredients (Pty) Ltd. (Johannesburg, South Africa).

Two commercial mayonnaise products (Crosse & Blackwell high and low-fat mayonnaise): high-fat (52% oil) and low-fat (25% oil) were purchased from a local supermarket and used as references in this study. The commercial high-fat mayonnaise contained thickeners, acidity regulators, colorants, and flavorings. The commercial low-fat mayonnaise (25% oil), in addition to all the above, also contains chemically modified maize starch.

Table 1. Composition (g) of experimental samples

Ingredients	Full-fat[a)]	50% oil replacement	80% oil replacement	98% oil replacement
Sunflower oil	280	140	56	5.6
Spray dried egg yolk	48	48	48	48
Vinegar	60	60	60	60
Salt	4	4	4	4
Sugar	4	4	4	4
Powdered mustard	1.98	1.98	1.98	1.98
Potassium sorbate	0.12	0.12	0.12	0.12
Sodium benzoate	0.12	0.12	0.12	0.12
Fat replacers[b)]	NA	140	224	274.4

NA, Not applicable;

[a)] 70% oil content;

[b)] Maize starch modified with 1.5% stearic acid/maize starch modified with 2% monoglyceride.

2.2 Reduced-Fat Mayonnaise-Type-Emulsions Formulation

Mayonnaise trial formulations were obtained by adjusting the percentage of ingredients from existing literature formulations[15, 18, 19] after several laboratory trials. Tests were done by first varying the levels of ingredients (sunflower oil, egg yolk, vinegar, salt, and sugar) and then keeping them constant to make provision for replacing some percentage of sunflower oil with the freshly produced fat replacers. In the formulation development phase, trial mayonnaise samples (full-fat and low-fat) were informally assessed by sensory scientists of the Department of Consumer and Food Sciences until the samples were generally rated high for likeness. Compositions of the final experimental samples used in this study are presented on a 100 g oil basis (Table 1).

2.3 Preparation of Reduced-Fat Mayonnaise-Type Emulsions

Modified maize starches were prepared by incorporating stearic acid or monoglyceride, into maize starch according to the method by D'Silva et al.[17] Stearic acid and monoglyceride were dissolved in absolute ethanol in a beaker and added to the maize starch at 1.5% and 2% (w/w) respectively (based on recommendations by Maphalla and Emmambux[8] and D'Silva et al.[17]). Maize starch+ monoglyceride or maize starch + stearic acid samples were added to the ethanol in a 1:3 (w:v) starch:ethanol ratio and placed in a shaking water bath at 50 °C for 30 min to ensure complete mixing and incorporation of ingredients. The ethanol was then evaporated in an oven at 40 °C because it only served as a solvent for the solutes (maize starch, monoglyceride, and stearic acid). Suspensions (10% w/w) of the stearic acid-starch and monoglyceride-starch mixtures were pasted using a reactor (IKA LR 1000 control, Staufen, Germany) at 91 °C for 30 min at a stirring rate of 150 rpm.

The method by Teklehaimanot et al.[15] was used with some modifications. The aqueous phase (vinegar, egg yolk, salt, sugar, mustard, and preservatives) was homogenized for 1 min at 4500 rpm using a Silverson high shear homogenizer L5T (Silverson Machines Ltd. Waterside, Chesham Bucks. England). Sunflower oil was then added gradually to the mix for 2 min while increasing the homogenizing speed from 4500 to 8000 rpm. The final emulsion was then homogenized at 8000 rpm for 2 min. In the case of the reduced-fat mayonnaise-type emulsions, 10% (w/w) suspensions of each fat replacer [maize starch with 1.5% stearic acid and maize starch with 2% monoglyceride (w/w)] mixed thoroughly with the aqueous phase before adding the oil. The emulsions were then poured into glass bottles and stored at 4 °C for further analysis within two weeks.

2.4 Descriptive Sensory Evaluation

Ethical approval for this study was obtained from the ethics committee of the Faculty of Natural and Agricultural Sciences, University of Pretoria, South Africa (EC170425-111). Ten trained panellists (6 females and 4 males) aged from 20 to 54 years, 8 of whom were part of the panel that previously assessed the sensory attributes of the starch-lipid complexes,[16] participated in this study.

Two sessions (2 h each) were used for the development of descriptors, definitions, agreement on references, and training on the use of scales. Panellists were given a list of potential mayonnaise descriptors (obtained from existing literature and focus group discussion using students from the Department of Consumer and Food Sciences, University of Pretoria) and asked to check attributes that best describe the mayonnaise samples. They were also asked to include additional attributes that might have been omitted. For the development of descriptive terms, the two commercial samples of mayonnaise and four experimental samples were used (Table 2). Once all samples were evaluated by the panellists, sessions of discussions were held to reach consensus on the list of terms with their definitions. Reference samples were also presented to panellists and consensus reached on anchoring the attributes on 10 point structured scales. A list of mouthfeel attributes, definitions, references, and scale used for evaluating the mayonnaise samples is given in Table 3.

Table 2. Oil content (%) of reduced-fat mayonnaise-type emulsions and commercial mayonnaise (reference) used in the study

Mayonnaise samples	Oil content %[a)]
Full-fat (control–no fat replacer)	70
50%[b)] (maize starch + 1.5% stearic acid)	35
80%[b)] (maize starch + 1.5% stearic acid)	14
98%[b)] (maize starch + 1.5% stearic acid)	1.4
50%[b)] (maize starch + 2% monoglyceride)	35
80%[b)] (maize starch + 2% monoglyceride)	14
98%[b)] (maize starch + 2% monoglyceride)	1.4
50%[b)] (commercial fat replacer mayonnaise)	35
Commercial high-fat (reference)	52
Commercial low-fat (reference)	25

[a)] Oil content as a weight fraction of the whole emulsion;

[b)] Percentage of oil replaced by fat replacer with respect to oil content in the full-fat formulation.

Table 3. Sensory attributes used by panel for evaluation of mayonnaise samples

Attributes[a)]	Definition	Scale (from 0 to 10)
	Flavor and in-mouth texture	
Bitter	Intensity of basic taste of which caffeine in water is typical	Not bitter–very bitter
Thick	Force required to compress a sample between the tongue and palate	Not thick–very thick
Smooth	The absence of detectable particles in the sample	Not smooth–very smooth
Creamy	The intensity of creaminess	Not creamy–very creamy
Mouth-coating	The extent to which sample forms a coating in the mouth during and after oral processing	Not mouth-coating–very mouth-coating
Melting	How easily the sample melted in the mouth	Melted slowly–melted very fast
Ease-to-swallow	Amount of effort required to swallow	Not easy-to-swallow–very easy-to-swallow

[a)] In-mouth textural sensory attributes discussed.

A randomized complete block design was used for the actual product evaluation. Eight experimental samples were used in addition to the two commercial samples (Table 2). About 2 mL of each sample was presented monadically at a temperature of 4–6 °C in glass ramekins covered with foil with stainless steel spoons. Samples were coded with a three-digit number.

Panellists were given lukewarm water (40 °C), recommended for products that leave an oily residue (The ASTM E1871 Standard, 2006) and carrots as palate cleansers.

All evaluations were performed by panellists seated in individual evaluation booths with white daylight illumination. The panellists were instructed to evaluate each sample by first lifting the side of the foil covering the ramekins and smelling to assess the sample for aroma attributes followed by appearance attribute. Samples were then placed in the mouth, and the flavor, mouthfeel, and aftertaste attribute assessed. A structured 10-point line scale was used to measure the intensities of the different attributes for each sample. Zero indicated the absence of the attribute being measured, while 10 indicated a high intensity of perception. Panellists evaluated all 10 samples in triplicate over three sessions lasting 2 h per day.

Compusense cloud (Compusense Inc., Guelph, Canada) was used to design the test setup, generate random codes and to capture responses from the trained panel.

2.5 Tribology Measurements (Friction Coefficient)

Tribology measurements were performed with a Mini-Traction Machine (MTM2. PCS Instruments Ltd. UK), using an elastomeric tribo-pairs PDMS (polydimethylsiloxane) disk and ball.[20] The instrument was coupled with an external water bath keeping the temperature in the measurements chamber at 35 ± 1 °C during the experiments. A load of 2 N was applied in all experiments while sliding-to-roll ratio, was set to 50% to mimic the relative movement of the tongue and soft palate in the oral cavity.

For each sample, two tribology experiments were done while using a new PDMS tribo-pair, a ball, and a disk, each time. Mayonnaise samples (15 g) were poured into the mini-traction machine measurement chamber onto the disk. The ball and the disk were brought into a loaded contact, and the entrainment speed was ramped from 750 to 1 mm s^{-1} and then back to 750 mm s^{-1}, while friction was accessed at 40 logarithmically spaced speeds during each ramp. Three ramping cycles immediately after each other were performed. The results for each speed are presented as an average of six friction coefficient values obtained during the three down-speed and the three up-speed ramps.

2.6 Flow Property Measurement

Flow properties were determined, according to Teklehaimanot et al.[15] with modifications. The shear behavior of the experimental samples was determined with a Physica MCR 101 Rheometer (Anton Paar, Ostfildern, Austria) using a vane and cup method. The shear rate was increased from 0.01 to 1000 s^{-1} and reduced back from 1000 to 0.01 s^{-1} at 25 °C. The data was analyzed using Cross rheology equations

$$\mu = \frac{\mu_o + \mu_\infty \cdot \alpha\gamma^n}{1 + \alpha\gamma^n} \quad (1)$$

as described by Xie and Jin.[21] Where γ is the shear rate, μ_o is the viscosity when the shear rate is close to zero, μ_∞ is the viscosity when the shear rate is infinity, n is the flow behavior index, and α is the consistency index. For shear-thinning materials, the value of n is between 0 and 1.

2.7 Texture Measurements

The texture of mayonnaise type emulsions was determined with an EZ-test texture analyzer (EZ– L, Shimadzu Tokyo, Japan). Samples were scooped into cylindrical containers (40 mm × 55 mm). Using a 35 mm probe, samples were compressed at a speed of 1 mm s^{-1}, to a sample depth of 40 mm, and then retracted. From the resulting force–time curve, the values for texture attributes, that is, firmness and adhesiveness, were obtained. Firmness was calculated as the maximum force reached, before the probe penetrates the sample and adhesiveness as the area below the negative force versus distance curve, representing the work necessary to pull the compressing probe away from the sample.

2.8 Statistical Analysis

A multivariate analysis of variance was used to determine the effect of the independent variables [oil replacement levels (50, 80, and 98%) and different potential fat replacers (maize starch + 1.5% stearic acid, maize starch + monoglyceride, and a commercial fat replacer)] on the rheological, tribological and textural properties of the mayonnaise-type emulsions and the least significant difference test (LSD) was used to separate the means ($p \leq 0.05$). Statistical analysis was performed using IBM SPSS version 20 (SPSS, Inc., 1998, Chicago, IL). For the sensory ratings, a two way ANOVA without interactions was conducted on the independent variables (samples and panellists) and the LSD test used to separate the means using XLSTAT 2014. Principal component analysis (PCA) was used to show the relationship between tribology, rheology, and sensory properties of the different mayonnaise-type emulsions.

3 Results

3.1 Mouthfeel Textural Characteristics of Reduced-Fat Mayonnaise-Type Emulsions

Mean scores of the mouthfeel textural attributes are shown in Table 4. Eighteen different attributes were evaluated, but only the in-mouth textural sensory attributes were tested for correlation to tribology and rheology in our PCA analysis are discussed.

Panellist rated oiliness (fattiness) of all the reduced-fat emulsions significantly lower ($p < 0.05$) compared to the full-fat mayonnaise control (Table 4). For the reduced-fat emulsions, there is a trend, although not significant, for a reduction in the perceived oiliness with the increasing level of fat-replacement.

The thickness of the reduced-fat emulsion containing starch/stearic acid was significantly lower ($p < 0.05$) compared to the mayonnaise formulated with starch/monoglyceride and the full-fat mayonnaise (Table 4). Oil replacement level (50%, 80%, and 98%) did not affect ($p > 0.05$) the thickness of emulsions containing starch/stearic acid. For the starch/monoglyceride fat-replacer, the level of fat replacement affected the thickness of emulsions; the perceived thickness of 50% fat-replaced sample was similar to that of the full-fat emulsion, while the thickness of 80% and 98% fat-replaced samples was significantly higher ($p < 0.05$) (Table 4).

Table 4. Mean ratings for flavor and texture (in-mouth feel) of reduced-fat mayonnaise-type emulsions

	Full-fat (control, 70% oil)	**Decreasing oil concentration**			**Decreasing oil concentration**			***p*-value**	***F*-value**
		Maize starch + 1.5% stearic acid			**Maize starch + 2% monoglyceride**				
		50%	80%	98%	50%	80%	98%		
Overall-flavor	$8.3^{a} \pm 1.9$	$7.6^{ab} \pm 2.5$	$7.1^{b} \pm 2.7$	$6.7^{b} \pm 2.8$	$7.5^{ab} \pm 2.8$	$8.1^{a} \pm 2.0$	$8.2^{a} \pm 2.2$	0.00	14.53
Saltiness	$6.3^{a} \pm 2.9$	$4.9^{b} \pm 3.1$	$4.8^{b} \pm 3.6$	$4.8^{b} \pm 3.1$	$5.3^{ab} \pm 3.3$	$5.2^{b} \pm 3.3$	$4.8^{b} \pm 3.0$	0.05	23.27
Thickness	$3.4^{b} \pm 3.0$	$1.1^{c} \pm 1.8$	$1.2^{c} \pm 2.0$	$0.5^{c} \pm 1.0$	$4.3^{b} \pm 2.6$	$5.7^{a} \pm 3.0$	$5.7^{a} \pm 3.4$	0.00	18.79
Smoothness	$8.9^{a} \pm 2.0$	$8.4^{ab} \pm 2.7$	$8.3^{b} \pm 2.6$	$8.3^{b} \pm 2.6$	$8.5^{ab} \pm 2.1$	$8.8^{ab} \pm 1.9$	$8.7^{ab} \pm 1.9$	0.13	52.80
Bitterness	$3.1^{bc} \pm 3.7$	$2.5^{c} \pm 3.1$	$2.9^{bc} \pm 3.1$	$3.8^{ab} \pm 3.7$	$2.9^{bc} \pm 2.9$	$3.2^{abc} \pm 3.1$	$4.3^{a} \pm 3.8$	0.06	14.30
Starchy	$2.9^{a} \pm 3.3$	$2.3^{a} \pm 2.8$	$3.2^{a} \pm 3.3$	$2.6^{a} \pm 3.4$	$2.9^{a} \pm 3.0$	$3.3^{a} \pm 3.5$	$3.6^{a} \pm 3.6$	0.52	10.30
Eggy	$4.8^{a} \pm 2.9$	$4.5^{a} \pm 2.7$	$4.4^{a} \pm 2.6$	$4.8^{a} \pm 3.6$	$4.5^{a} \pm 2.7$	$4.6^{a} \pm 2.7$	$4.5^{a} \pm 2.7$	0.99	7.66
Oiliness	$6.9^{a} \pm 2.8$	$4.4^{bc} \pm 3.4$	$4.3^{bc} \pm 3.0$	$3.7^{c} \pm 2.8$	$5.2^{b} \pm 2.8$	$5.1^{b} \pm 3.2$	$4.9^{b} \pm 3.3$	0.00	21.47
Vinegar flavor	$6.5^{a} \pm 3.0$	$5.9^{a} \pm 2.8$	$6.0^{a} \pm 2.4$	$6.4^{a} \pm 2.7$	$6.2^{a} \pm 2.3$	$7.1^{a} \pm 2.3$	$6.9^{a} \pm 2.1$	0.42	2.60
Creaminess	$7.3^{a} \pm 2.7$	$6.2^{bc} \pm 3.4$	$5.2^{cd} \pm 3.9$	$5.0^{d} \pm 4.0$	$7.3^{a} \pm 2.7$	$7.1^{ab} \pm 2.9$	$6.7^{ab} \pm 3.3$	0.00	23.92
Sweet	$2.7^{a} \pm 3.1$	$2.1^{ab} \pm 2.0$	$1.9^{abc} \pm 2.2$	$1.5^{bc} \pm 2.0$	$2.2^{ab} \pm 2.5$	$1.5^{bc} \pm 1.7$	$1.2^{c} \pm 1.6$	0.01	12.61
Mustard	$3.2^{a} \pm 3.4$	$3.1^{a} \pm 2.4$	$3.4^{a} \pm 2.9$	$3.5^{a} \pm 3.2$	$3.0^{a} \pm 2.4$	$3.4^{a} \pm 2.8$	$4.1^{a} \pm 3.4$	0.66	7.33
Mouth-coating	$4.4^{a} \pm 3.0$	$2.6^{b} \pm 2.9$	$3.0^{b} \pm 3.2$	$2.8^{b} \pm 3.1$	$3.6^{ab} \pm 3.4$	$3.7^{ab} \pm 3.5$	$4.3^{a} \pm 3.5$	0.00	18.65
Melting	$8.7^{bc} \pm 1.8$	$9.2^{ab} \pm 1.3$	$9.3^{a} \pm 1.2$	$9.4^{a} \pm 0.8$	$8.5^{c} \pm 1.9$	$8.7^{bc} \pm 1.8$	$8.1^{c} \pm 2.4$	0.00	15.97
Ease-to-swallow	$9.1^{a} \pm 1.3$	$9.2^{a} \pm 1.2$	$9.0^{ab} \pm 1.6$	$9.0^{ab} \pm 1.9$	$9.0^{ab} \pm 1.4$	$8.8^{bc} \pm 1.6$	$8.5^{c} \pm 1.8$	0.00	70.50
Astringent	$3.4^{b} \pm 3.0$	$3.7^{b} \pm 3.1$	$4.5^{ab} \pm 3.4$	$5.2^{a} \pm 4.1$	$4.6^{ab} \pm 3.8$	$5.4^{a} \pm 3.9$	$5.6^{a} \pm 4.0$	0.00	24.81
Artificial	$2.2^{d} \pm 3.1$	$2.7^{cd} \pm 3.0$	$3.3^{bc} \pm 3.6$	$3.9^{ab} \pm 4.3$	$2.9^{bcd} \pm 3.4$	$3.6^{bc} \pm 3.6$	$4.7^{a} \pm 4.0$	0.00	27.02
Tangy	$6.2^{abc} \pm 3.3$	$5.9^{abc} \pm 3.1$	$5.8^{bc} \pm 3.4$	$4.8^{c} \pm 3.7$	$5.8^{bc} \pm 3.1$	$6.6^{ab} \pm 3.4$	$7.3^{a} \pm 2.6$	0.04	5.87

Values are means of panel ratings; Mean values in the same row with different superscripts differ significantly ($p < 0.05$).

Panellist rated all the reduced-fat emulsions high for smoothness, melting, and easy-to-swallow. The different fat replacers and oil replacement levels did not significantly affect ($p > 0.05$) the smoothness, melting, and ability to swallow. On the other hand, choice of fat-replacer significantly affected ratings for the creaminess and mouth-coating attributes; the emulsions formulated with starch/monoglyceride were statistically similar to that of the full-fat mayonnaise samples, but emulsions formulated with starch/stearic acid were rated significantly less creamy and mouth-coating than the full-fat reference (Table 4). For both fat-replacers, mean scores for creaminess indicated decrease with the increasing level of fat replacements; however, this trend was not statistically significant.

There is also an interesting trend in astringency ratings, where the mean scores are increasing above those obtained for the full-fat sample with the increasing level of fat replacement. The significant difference in comparison to the full-fat reference was reached at 98% fat replacement level by starch/stearic acid and for 80% and 98% fat replacement level for starch/monoglyceride. The perception of astringency might be possibly related to the sensory attribute "artificial" that shows a similar trend.

3.2 Tribology of Reduced-Fat Mayonnaise-Type Emulsions

The Stribeck curves of all samples were relatively similar. They feature mixed lubrication regime up to entrainment speed of about 15–20 mm s^{-1}, characterized by friction coefficient decreasing with speed, followed by full elastohydrodynamic regime, characterized by an increase of friction coefficient with the entrainment speeds. A distinctive additional feature of the Stribeck curves of all reduced-fat emulsions containing fat-replacers (Figure 1A,B) is the presence of "hump" at (high) entrainment speeds of about 200 mm s^{-1}, while the corresponding feature is absent for the full-fat reference with no fat-replacer.

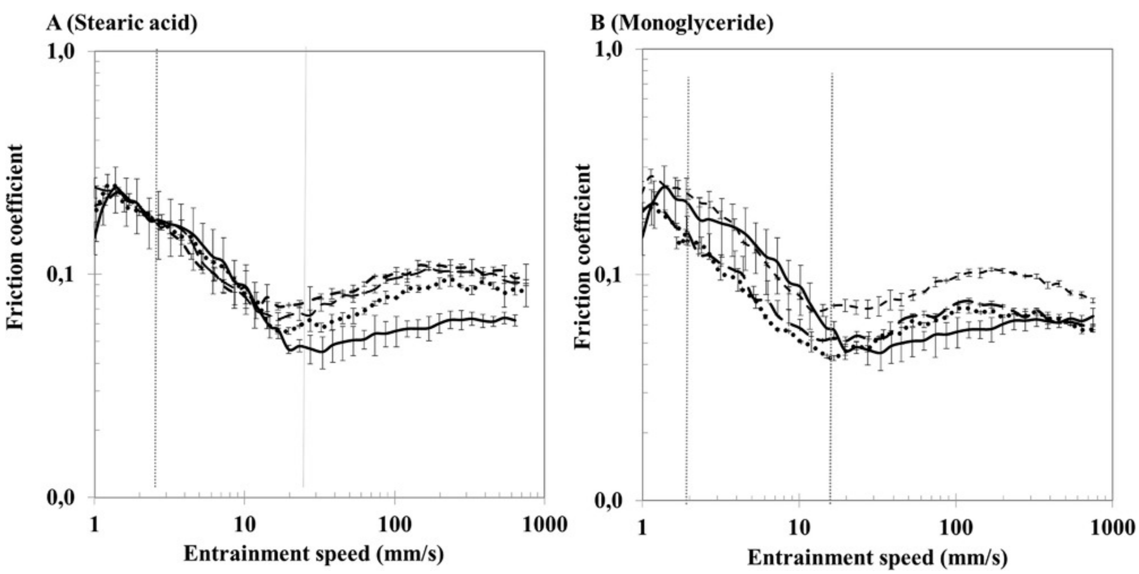

Figure 1. Effect of lipid-modified maize starch fat replacers on the friction coefficient of reduced-fat mayonnaise-type emulsions containing different levels (full fat — , 50% … , 80% – – – , 98% − − −) of maize starch modified with 1.5% stearic acid (A) and maize starch modified with 2% monoglyceride (B).

In the mixed regime, the surfaces are in partial contact and friction is affected both by lubricant fluids that are partly entrained between surfaces and by interfacial phenomena such as surface roughness, surface chemistry, and adsorption of the components of lubricant fluid onto surfaces. In the elastohydrodynamic lubrication regime, the surfaces are fully separated by a lubricant film, and the friction is determined by the bulk properties of lubricant fluids.

Friction coefficient values obtained for the mayonnaise emulsions in the mixed regime, at the speed of 5 mm s^{-1}, and in the elastohydrodynamic regime, at the speed of 100 mm s^{-1}, are presented in Table 5. These speeds represent the points at which the lubricant fluids were effectively entrained in the mixed and elastohydrodynamic regime, respectively. In the mixed regime, the friction coefficient values measured for the reduced-fat emulsions formulated with starch/stearic acid fat replacers were not significantly different ($p > 0.05$) from those for the full-fat sample, however, significantly lower friction was measured for the reduced-fat emulsions formulated with starch/monoglyceride at 50% and 80% fat replacer levels. In the elastohydrodynamic regime, it was, on the other hand, that the starch/monoglyceride emulsions with lower friction were similar to the full-fat emulsion, while the friction coefficient values for the starch/stearic acid emulsions were significantly higher ($p < 0.05$) than for full-fat sample in this regime. Only at the highest level of fat substitution, 98%, by starch/monoglyceride, the friction in the elastohydrodynamic regime became higher than for the full-fat sample and similar to that measured for the starch/stearic acid emulsions. The choice of lipid used to modify maize starch as the fat replacer apparently affected lubrication properties of the reduced-fat emulsions. In both mixed and elastohydrodynamic lubrication regimes, the reduced-fat emulsions formulated with starch/stearic acid showed a rise to higher friction (i.e., were less lubricious) than the corresponding emulsions formulated with starch/monoglyceride.

Table 5. Friction coefficient of the different reduced-fat mayonnaise-type emulsions at sliding speeds 5 mm s^{-1} (mixed lubrication regime) and 100 mm s^{-1} (hydrodynamic regime)

Mayonnaise samples	Fat replacement [%]	Friction coefficient (μ) 5 mm s^{-1}	Friction coefficient (μ) 100 mm s^{-1}
Full-fat (control, 70% oil)	NA	$0.14^{a} \pm 0.03$	$0.06^{c} \pm 0.01$
Maize starch + 1.5% stearic acid	50	$0.12^{a} \pm .00$	$0.08^{b} \pm 0.00$
	80	$0.12^{a} \pm 0.01$	$0.10^{a} \pm 0.01$
	98	$0.11^{ab} \pm 0.00$	$0.10^{a} \pm 0.01$
Maize starch + 2% monoglyceride	50	$0.08^{b} \pm 0.01$	$0.07^{bc} \pm 0.00$
	80	$0.08^{b} \pm 0.01$	$0.08^{bc} \pm 0.00$
	98	$0.13^{a} \pm 0.01$	$0.10^{a} \pm 0.00$

Values are means of replicate readings with standard deviation; Mean values in the same column with different superscripts differ significantly ($p < 0.05$).

In general, all reduced-fat emulsions exhibited good lubrication and the tribological differences to the full-fat control were relatively small.

3.3 Flow Properties of Reduced-Fat Mayonnaise-Type Emulsions

All the emulsions exhibited shear thinning behavior ($n < 1$) with their flow behavior index, n, ranging from 0.64 to 0.78 (Table 6). Addition of the modified starches increased the hysteresis area of the reduced-fat emulsions compared with the full-fat mayonnaise. Reduced-fat emulsions formulated with starch/monoglyceride fat replacer had significantly higher ($p < 0.05$) hysteresis values compared to the emulsions formulated with starch/stearic acid fat replacer (Table 6).

Table 6. Effect of lipid-modified maize starch fat replacers on the flow-properties [consistency coefficient (*K*) and flow behavior index (*n*)] and zero shear viscosity determined from the Cross equation, hysteresis, and textural properties [firmness and adhesiveness] of reduced-fat mayonnaise-type emulsions

Mayonnaise samples	Fat replacement [%]	*n*	*K* [Pa s^n]	Hysteresis [Pa s]	Zero shear viscosity [Pa s]	Firmness [N]	Adhesiveness [N]
Full-fat (control, 70% oil)	NA	0.74^c ± 0.01	50.28^g ± 1.84	20201^a ± 4804	322.47^{bc} ± 16.49	0.16^b ± 0.01	−0.04^d ± 0.01
	50	0.64^e ± 0.01	12.14^j ± 0.28	23524^{ab} ± 1521	149.6^{de} ± 34.82	0.11^a ± 0.00	−0.03^d ± 0.00
Maize starch + 1.5% stearic acid	80	0.76^b ± 0.01	33.93^h ± 1.56	25991^{abcd} ± 793	66.83^e ± 27.12	0.21^{cd} ± 0.02	−0.05^d ± 0.00
	98	0.73^c ± 0.02	16.88^i ± 0.66	24471^{abc} ± 1682	29.53^e ± 11.83	0.17^b ± 0.01	−0.04^d ± 0.01
	50	0.78^a ± 0.02	56.84^f ± 4.79	35213^d ± 1246	284.8^{bc} ± 21.98	0.19^{bc} ± 0.00	−0.06^d ± 0.01
Maize starch + 2% monoglyceride	80	0.78^a ± 0.01	77.99^d ± 2.48	34520^{cd} ± 983	150.83^{de} ± 59.6	0.27^{ef} ± 0.01	−0.11^c ± 0.03
	98	0.76^{ab} ± 0.01	95.75^c ± 2.98	31120^{bcd} ± 528	376.17^b ± 113.7	0.28^f ± 0.01	−0.13^{ab} ± 0.03
Commercial fat replacer mayo	50	0.76^b ± 0.01	121.03^a ± 3.6	79225^f ± 10320	912.33^a ± 139.37	0.38^h ± 0.01	−0.17^a ± 0.02
Commercial high-fat (52% oil)	NA	0.70^d ± 0.01	113.37^b ± 1.86	47672^e ± 2107	209.13^{cd} ± 56.44	0.34^g ± 0.00	−0.13^{ab} ± 0.01
Commercial low-fat (25% oil)	NA	0.71^d ± 0.01	71.28^e ± 1.76	28788^{abcd} ± 1970	132.33^{de} ± 130.37	0.24^{cd} ± 0.01	−0.06^d ± 0.00

Values are means of triplicate readings with standard deviation; Mean values in the same column with different superscripts differ significantly ($p < 0.05$).

The plot of viscosity versus shear rate of the reduced-fat emulsions at different levels of oil replacement (Figure 2) shows that the emulsions prepared using starch/monoglyceride had significantly higher ($p < 0.05$) zero-shear viscosities compared with reduced-fat emulsions formulated with starch/stearic acid as fat replacer. Reduced-fat emulsions formulated with starch/stearic acid fat replacer had the lowest consistency coefficient values, while the mayonnaise formulated with the commercial fat replacer, on the other hand, recorded the highest consistency coefficient. Firmness and adhesiveness of the reduced-fat emulsions formulated with starch/monoglyceride fat replacers were significantly higher ($p < 0.05$) than for their counterparts formulated with starch/stearic acid fat replacers and for the full-fat emulsion.

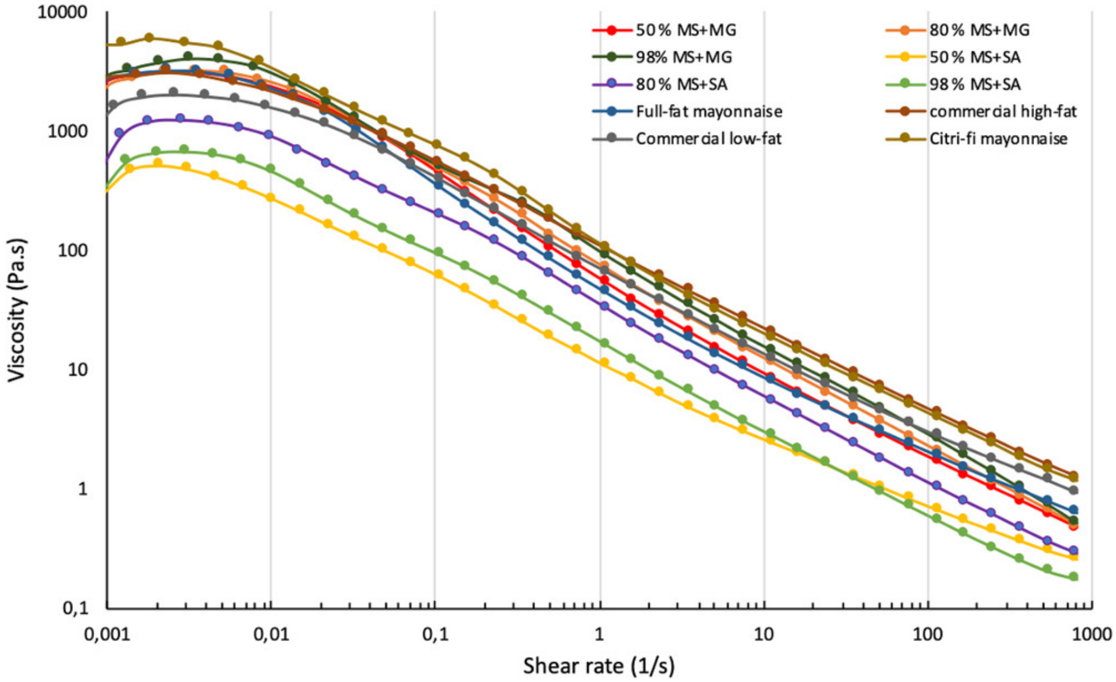

Figure 2. Effect of lipid-modified (SA, stearic acid; MG, monoglyceride) maize starch fat replacers on the viscosity of reduced-fat mayonnaise-type emulsions compared with commercial mayonnaise samples.

Mayonnaise formulated by replacing 50% of sunflower oil with the commercial fat replacer was substantially different from the other reduced-fat emulsions. It had significantly ($p < 0.05$) the highest consistency coefficient, hysteresis, zero-shear viscosity, firmness, and adhesiveness compared to all the experimental samples.

4 Discussion

PCA was applied to correlate tribological and rheological data with in-mouth sensory attributes (Figure 3), and Pearson's correlation coefficients from PCA were used to evaluate the significance of the relationships between the data (Table 7). The PCA biplot explained 81.40% of the variation between the data. The first component (PC1) explained 55.13% of the total variance and, the second (PC2) accounted for the remaining 26.26%. PC1 associated reduced-fat emulsions formulated with starch/monoglyceride with the full-fat mayonnaise and these were separated from the emulsions formulated with starch/stearic acid. The reduced-fat emulsions formulated with starch/monoglyceride were associated with the following attributes: creaminess, smoothness, mouth-coating, zero-shear viscosity, thickness, firmness, tanginess, and astringency. The reduced-fat emulsions formulated with starch/stearic acid were, on the other hand, associated with melting and easy-to-swallow attributes.

Table 7. Pearson correlation coefficients between tribology, rheology, and in-mouth sensory properties of reduced-fat mayonnaise-type emulsions

	Texture and flow properties						Tribology		Sensorial texture (in mouth)								
Variables	Firmness [N]	Adhesiveness [N]	Zero shear viscosity [Pa s]	*n*	*K* [Pa sn]	Hysteresis [Pa s]	Friction coefficient (μ) @ 5	Friction coefficient (μ) @ 100	Thickness	Smoothness	Oiliness	Creaminess	Mouth-coating	Melting	Ease-to-Swallow	Astringent	Tangy
Firmness [N]																	
Adhesiveness [N]	−0.93																
Zero shear viscosity [Pa s]	0.68	−0.74															
n	−0.89	0.82	−0.55														
K [Pa sn]	0.50	−0.54	0.81	−0.60													
Hysteresis [Pa s]	0.68	−0.69	0.52	−0.46	0.02												
Friction coefficient (μ) @ 5	−0.26	0.17	−0.11	0.15	0.27	−0.76											
Friction coefficient (μ) @ 100	0.29	−0.21	−0.35	−0.41	−0.28	0.03	0.17										
Thickness	0.75	−0.85	0.94	−0.58	0.68	0.69	−0.28	−0.31									
Smoothness	0.39	−0.49	0.82	−0.36	0.89	0.13	0.09	−0.62	0.77								
Oiliness	0.02	−0.06	0.66	0.05	0.72	−0.13	0.18	−0.82	0.50	0.85							
Creaminess	0.22	−0.37	0.80	−0.05	0.57	0.39	−0.20	−0.79	0.80	0.82	0.80						

	Texture and flow properties						Tribology		Sensorial texture (in mouth)							
Mouth-coating	0.53	−0.57	0.94	−0.49	0.93	0.20	0.16	−0.39	0.80	0.88	0.80	0.73				
Melting	−0.60	0.75	−0.94	0.46	−0.68	−0.55	0.03	0.26	−0.92	−0.70	−0.53	−0.78	−0.85			
Ease-to-Swallow	−0.91	0.95	−0.66	0.88	−0.53	−0.57	0.02	−0.43	−0.71	−0.35	0.05	−0.18	−0.54	0.70		
Astringent	0.80	−0.76	0.29	−0.85	0.14	0.68	−0.40	0.63	0.42	−0.05	−0.48	−0.16	0.09	−0.30	−0.82	
Tangy	0.65	−0.80	0.77	−0.42	0.61	0.38	0.20	−0.15	0.82	0.69	0.44	0.61	0.71	−0.82	−0.70	0.23

Values indicate correlation coefficients significant at ($p < 0.05$); Green showing high positive correlation and red showing high negative correlation.

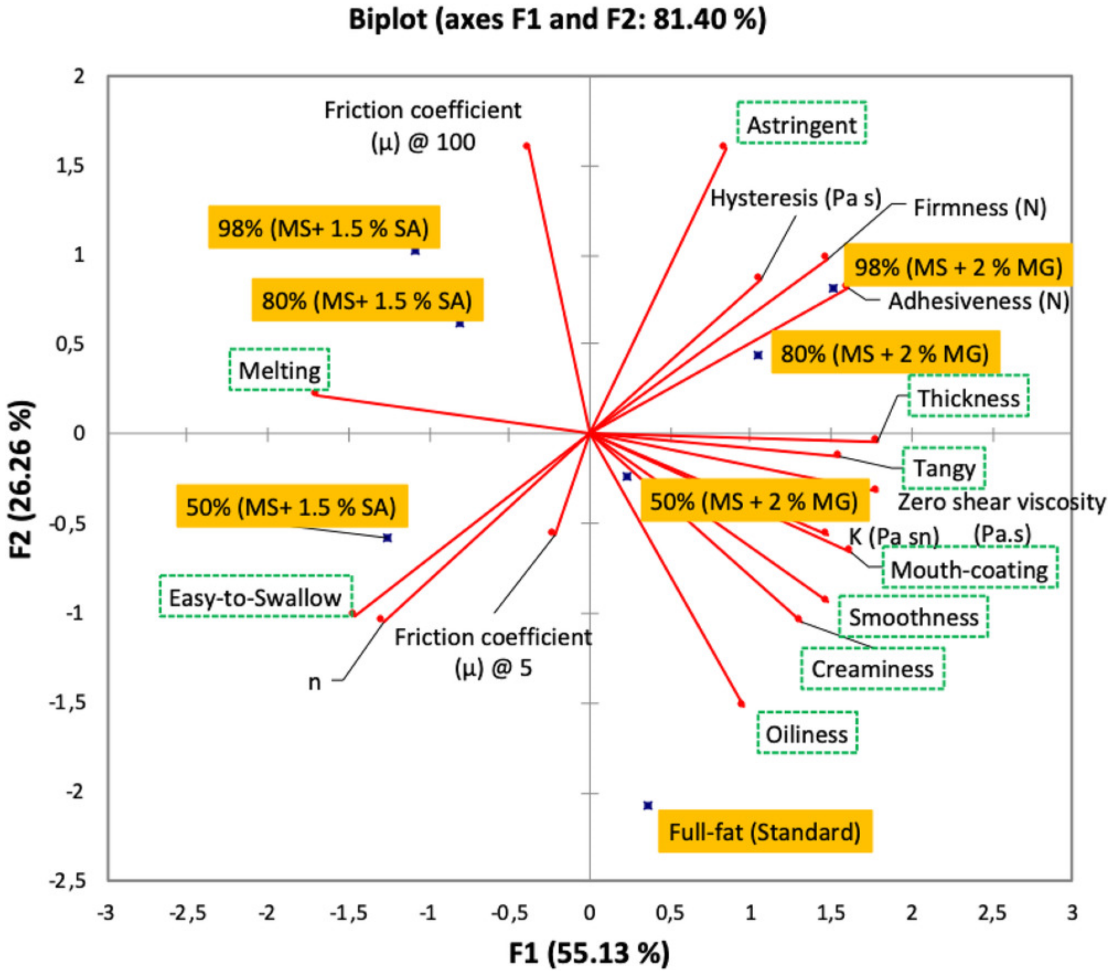

Figure 3. PCA biplot showing the relationship between tribology, rheology, and in-mouth sensory properties (see dashed green boxes) of the different reduced-fat mayonnaise-type emulsions with maize starch modified with stearic acid (SA) and monoglycerides (MG).

Oiliness and creaminess ratings (Table 4) by the sensory panel can be related to the tribology results (Figure 1A,B) in the elastohydrodynamic regime, where higher oiliness and creaminess was perceived for samples with lower friction coefficient. The reduced-fat emulsions prepared with starch/stearic acid were rated less oily and creamy than the corresponding emulsions prepared with starch/monoglyceride and full-fat emulsion. The PCA biplot (Figure 3) and Pearson correlation coefficient matrix (Table 7) showed that the friction coefficient in the elastohydrodynamic regime correlated negatively with oily mouthfeel (−0.82) and creaminess (−0.79), while a weaker negative correlation was also found for smoothness (−0.62). Similar correlations of the measured friction with the perceptions of oiliness and creaminess were found by Dresselhuis et al.[22] suggested that both tribology and sensory results originated in the different tendencies of the studied emulsions to coalesce. No significant correlation was found for sensory attributes and friction in the mixed regime (assessed at 5 mm s^{-1}).

All samples showed good lubrication in simulated condition modeled by soft surfaces with high surface roughness. The differences in the tribological properties for different samples were relatively small but still significant. Low friction and similarity of all collected Stribeck curves (Figure 1A,B) are consistent with a view that contact areas were lubricated by the oil phase for all tested emulsions. The oil that is initially present in the form of emulsified droplets forms a thin film in the contact area after surface-induced droplet coalescence takes place at the polydimethylsiloxane surfaces. This phenomenon was previously observed in a similar set up by Dresselhuis et al.,[23] where such coalescence occurred for emulsions with as low as 1% oil content. Since the lowest oil content in our samples is 1.4% (for samples with 98% level of fat replacement), the presence of oil in the contact area is a reasonable assumption. This assumption is further supported by the presence of the "hump" in the curves collected for the reduced-fat emulsions. At entrainment speeds of about 200 mm s^{-1}, that is, well within the full film (elastohydrodynamic) regime, the friction coefficient starts to decreases with speed, which is rather peculiar for full film lubrication during the measurement. Assuming the presence of oil film in the contact area, the "hump" observation could be explained within the contact starvation theory that is well accepted in the field of emulsion-based machine lubricants.[24] For metal rolling contacts, it was shown that oil is preferentially entrained into contact area at low speeds; however, some water gets entrained along with the oil after reaching critical (high enough) entrainment speed. The presence of water in the contact area then leads to decrease of full film thickness and decrease in friction coefficient for emulsions in comparison to neat oil lubricants.[25] The phenomena should be more pronounced for emulsions with lower oil content, which is consistent with our data when it is present only for the fat-replaced samples with oil content 35% and lower and absent for full-fat emulsion with 70% oil.

Possible differences in lubrication behavior of reduced-fat emulsions formulated with starch/stearic acid and reduced-fat emulsion with starch/monoglyceride may arise from differences in their stability, that is, in the ability of emulsions components to promote or prevent oil droplet coalescence. Our hypothesis is that the higher friction, which was observed for all reduced-fat emulsions with starch/stearic acid and for 98% reduced-fat starch/monoglyceride sample is related to an incomplete/patchy coverage of the polydimethylsiloxane substrates by oil films due to higher stability of oil droplets in the corresponding emulsion matrixes. In the study of Dresselhuis et al.[22] on the relation of in-mouth coalescence of emulsions to the perception of fat, it was shown that the emulsions that were more sensitive to coalescence gave rise to lower friction. In this context, the friction results indicate that fat-reduction by modified starch with amylose–stearic acid complexes leads to more efficient stabilization of the remaining oil-droplets against coalescence than fat-reduction by modified starch with amylose-monoglyceride complexes; the latter gives rise to higher friction only at the highest fat-replacement level tested. In future, studies of oil droplet stabilization by resistant starch particles containing amylose–lipid complexes could elucidate the mechanism behind the detected differences between starch/stearic and starch/monoglyceride samples.

The high descriptive sensory ratings for thickness, smoothness, melting, and easy-to-swallow for all the reduced-fat emulsions can be explained by the rheological properties of the fat replacer containing amylose–lipid complexes that can associate into regular crystalline structures and occur as nanoparticles of less than 100 nm.[26, 27] These sensory attributes are highly cross-correlated, for example, the perceived thickness of samples was positively correlated to smoothness (0.77), creaminess (0.80), mouth-coating (0.80), and negatively correlated with melting (−0.92) (Figure 3 and Table 7). Chen and Stokes,[28] Jervis et al.,[29] and Upadhyay et al.[30] also observed similar research outcomes. They reported the relationship between the creaminess and the amount of fat present in a product in association with attributes like viscosity, taste, aroma, smoothness, and thickness.

Rheological and textural properties play a prominent role in sensory perception. Zero shear viscosity, which was positively cross-correlated with consistency coefficient, K shows a significantly high correlation with sensory attributes such as thickness (0.94), smoothness (0.82), creaminess (0.80), and mouth-coating (0.94). Zero shear viscosity, on the other hand, correlated highly negative with the rate of melting (−0.94). The textural property of firmness, which was negatively cross-correlated with adhesiveness, possibly affected sensory perception of thickness (0.75), while a significant negative correlations were found with easy to swallow attribute (−0.91) and positive correlation with the perception of astringency (0.80).

Higher thickness ratings for the reduced-fat emulsions formulated with starch/monoglyceride can be supported by the high zero shear viscosity and firmness values measured instrumentally (Table 6). The high zero-shear viscosity and firmness values recorded for the mayonnaise with 50% commercial fat replacer could be explained by the high concentration of polymers. This possibly caused an increase in molecular entanglement in the commercial fat replacer. Concentrated polymer solutions with constant entanglement density show an increasing zero shear viscosity with increasing concentration.[31] Lower firmness of the reduced-fat mayonnaise-type emulsions formulated with the lipid-modified maize starches used in our study compared with the mayonnaise formulated with the commercial fat replacer (Table 6) can be attributed to the reduced rate of retrogradation due to the presence of the amylose–lipid complexes in the "clean-label" fat replacers.[17]

The zero shear viscosity is substantially affected by the structure and association properties of the amylose–lipid complexes present in the fat-replacers. The viscosity of shear-thinning fluids as shear rate approaches zero, is referred to as it is zero shear viscosity,[31, 32] and indicates its physical stability. The zero shear viscosity phenomenon occurs when a constant density of entanglements is reached between releasing entanglements caused by shearing and their reformation caused by Brownian motions at low shear rates.[31, 32] The higher zero shear viscosity and firmness values imply that reduced-fat emulsions formulated using starch/monoglyceride are more stable at a set position just before shear is applied. Teklehaimanot et al.[15] produced low-calorie reduced-fat mayonnaise-type emulsions with lipid-modified starches that had a lower viscosity at different shear rates, especially those at low shear rates compared with the unmodified starches when they used starch modified with stearic acid-containing amylose–lipid complexes, as a fat-replacer in their formulation. They attributed the observation to the properties of the starch-lipid complex used as a fat replacer. Formation of amylose–lipid complexes result in softer non-gelling pastes[17] by preventing the re-association of amylose molecules from forming junction zones[33, 34] hence retarding the gelling process.

The rheological differences between the emulsions containing starch/stearic acid and starch/monoglyceride are related to different ability of various lipids to complex with amylose.[35] Long-chain saturated fatty acids such as stearic acid, form amylose–lipid complexes more readily compared to the monoglyceride.[36] Thus available uncomplexed amylose, in the case of the monoglyceride, can associate with each other to form junction zones and thus higher amount of molecular entanglement[34] leading to higher viscosity. Polymers with more junction zones are more rigid and rebuild their structure less easily when disturbed by shearing force,[33] which would explain the higher zero shear viscosity between the two emulsions; starch/monoglyceride in comparison to starch/stearic acid.

All tested emulsions had clear shear thinning characteristics. Shear-thinning occurs when there is a breakdown of structural units in food during shearing and particles orientate along the shear direction, minimizing resistance to flow.[31, 34] For thixotropic shear-thinning fluids, when the concentration or molecular weight increases, flow behavior index (n) decreases.[37] This trend was observed with all the reduced-fat emulsions formulated with the different fat replacers as their concentrations increased along with increasing fat-replacement level (Table 6). Mun et al.[10] and Teklehaimanot et al.[15] also reported a decrease in flow behavior index for all the reduced-fat mayonnaise samples when they substituted fat with modified starch-based fat replacers. Shear-thinning is often exhibited by emulsions that contain particles (e.g., droplets, crystals, or biopolymers) that are aggregated by weak forces.[38] During the shearing process, the aggregated particles present in the emulsion deform and disrupt and eventually reduce the emulsion's resistance to flow, resulting in a reduction in its apparent viscosity over time.

Starch–lipid complexes have the ability to increase viscosity during pasting.[8, 17] Hence, the addition of the lipid-modified maize starch complexes as fat-replacers, consequently, increased the viscosity of the continuous phase of the reduced-fat emulsions. This leads to an increase in the energy needed to rebuild its network structure, hence their high hysteresis area. Tárrega et al.[39] suggested a possible relationship between the viscosity and hysteresis of a thixotropic fluid, that is, the higher the viscosity, the larger the hysteresis. The viscous nature of the reduced-fat emulsions containing starch/monoglyceride probably contributed to the higher hysteresis values.

In our rheological and tribological experiments, we have not tested our samples in the presence of saliva that affects food emulsions during oral processing. Food emulsions are prone to flocculation and destabilization as a result of mixing with saliva and oral shearing.[40-42] The droplet destabilization process, which affects lubrication, is dependent on their charge status in an emulsion and is critical for their oral stability.[41] A non-flocculated emulsion would normally be perceived as smooth and creamy, but a flocculated emulsion would often be sensed as rough and dry with probably increased thickness sensation[43] and severe flocculation could lead to coalescence of oil droplets which could be perceived as a greasy or oily emulsion.[44] In our study, all the samples were perceived as highly smooth and creamy (Table 6), possibly indicating the absence of flocculated droplets.

Presence of saliva might affect the perception of food also through the action of salivary enzymes. Lubrication of starch-based fat-reduced food exposed to salivary amylase was studied by de Wijk and Prinz,[45] showing the break-down of food matrix and fat-release. The corresponding effect is not expected in our sample since the preparation process of the amylose–lipid complexes used in our study results in the formation of highly crystalline and digestion resistant starches.[8, 17]

5 Conclusions

The fat replacers can successfully be used to replace sunflower oil to produce reduced-fat mayonnaise-type emulsions with good lubricity without significantly compromising important in-mouth textural sensory perceptions of smoothness, creaminess, melting, and mouth-coating. The thickness and mouth-coating ratings as sensory properties can be correlated with measured rheological properties. Perceptions of creaminess and oiliness have been correlated with tribological properties and attributed to the ability of the modified starches containing amylose–lipid complexes to stabilize (remaining) oil droplets in the food emulsions matrix against coalescence. Thus, sensory properties are a combination of the rheology and tribology properties of the reduced-fat mayonnaise type emulsion.

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Conflict of Interest

The authors declare no conflict of interest.

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