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Linking the physicochemical properties of bulking agents to the sensory characteristics of fat-based suspensions

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ABSTRACT

Consumers are becoming more aware of the importance of healthy foods but do not want to bargain on their sweet sins. The pressure to reduce sucrose intake increases but poses major challenges at the technological level. Two main quality defects of fat-based suspensions are adherence and grittiness which are believed to be mainly related to the flow behaviour and particle size of these suspensions. This research wanted to investigate a possible link between the physicochemical properties (moisture content, particle density, particle morphology, particle size, solid state, solubility and viscosity) of several alternative bulking agents (*i.e.*isomalt ST, fructo-oligosaccharides, inulin and rice starch) and the mouthfeel of the corresponding sucrose-free fat-based suspensions. Sensory tests performed by a trained panel showed that the solid state and solubility of the bulking agents, and the viscosity of their solutions have a direct impact on grittiness and adherence. These insights will help the food industry in screening bulking agents for their applicability in sugar-free or sugar-reduced fat-based suspensions.

1. Introduction

Since 1980, worldwide obesity has increased to alarming levels. In 2014, 13% of the worldwide adult population was obese (WHO Media Centre, 2016). Obesity is associated with metabolic diseases, especially if a positive energy balance is combined with a sedentary life style. Brand-Miller and Barclay (2017) challenged the widespread belief that the intake of added sugars is uniquely linked to obesity and suggested a possible additional role of alcoholic drinks and energy-dense savory snacks. Nevertheless, governmental and regulatory agencies agree that added sugars should be targeted as a potential means of balancing energy intake with energy expenditure in order to curb obesity (Edwards et al., 2016). Meanwhile, worldwide health agencies are calling for further research on the sugar intake and health relationship. Given the pressure to reduce the sugar intake and consumers' awareness of the importance of healthy foods, food technologists face a major challenge to reduce the sugar level of their processed food products.

Sucrose, derived from sugar cane and beet, is the primary sweetener and has a highly accepted, clean taste showing early maximum sweetness intensity (Fujimaru et al., 2012). In fat-based suspensions, sucrose has additional functionalities, such as the provision of volume and other textural characteristics (Afoakwa et al., 2008). Chocolate, chocolate flavoured coatings, chocolate spreads, nut spreads and nut fillings exemplify fat-based suspensions comprising solid particles in a continuous lipid phase (Chevalley, 1975). Sucrose is often the main component of these dispersed solids, thus influencing important quality attributes such as taste and mouthfeel (Jeffery, 1993). It should be clear that through replacement of sucrose by alternative bulking agents, the sensory characteristics of high-quality confectionery products could be compromised.

Several studies focused on the effect of sucrose substitution on the physicochemical properties of fat-based suspensions. The technological feasibility of maltitol as a sucrose replacer in dark chocolate and chocolate spread was demonstrated by Sokmen and Gunes (2006) and Petkovic et al. (2012), respectively. Alternatively to sugar alcohols, low-digestible polysaccharides can be used to replace sucrose. Abbasi and Farzanmehr (2009) and Konar et al. (2014) observed a substantial impact of sucrose replacement by polydextrose on the chocolate flow parameters, affected by substitution level, fineness grade and process conditions. Bonarius et al. (2014) investigated the potential use of fibre-rich materials,

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in particular spent grain, pecan fibre, lemon peel and grape pomace, to replace sucrose in confectionery products. Since these fibre-rich materials had a specific influence on the flow behaviour of oily model systems, they stated that only a partial replacement of sucrose by fibre-rich materials in commercial applications can be successful. Aidoo et al. (2014) investigated the functionality of binary mixtures of bulking agents in sugar-free dark chocolate and found that an inulin/polydextrose 25/75 system had similar flow parameters than the sucrose-sweetened reference. None of the above-mentioned studies included sensory evaluations in their experimental set-up. It is true that the flow properties of fat-based suspensions directly affect the mouthfeel (Afoakwa et al., 2007). Hereby, highly viscous products have a pasty mouthfeel persisting in the mouth, *i.e.* highly adherent (Beckett, 2000). However, the overall sensory perception only becomes obvious when the fat-based suspension liquefies and the dispersed phase interacts with the saliva (Zumbé et al., 2001). Thus, the behaviour of the dispersed bulking agent(s) in a watery environment seems also crucial for product acceptance but is neglected in most studies.

There are scientific studies about consumer acceptance of sugar-free fat-based suspensions. Markey et al. (2015) evaluated the consumer acceptance of commercially available regular and maltitol-sweetened milk chocolate. Despite the fact that the overall liking score was similar, the liking scores for appearance, texture and flavour were higher for the regular chocolate than for the sugar-free counterpart. Several research teams approached sucrose substitution in chocolate by combinations of an intensive sweetener and bulking agents. Melo et al. (2009) performed consumer tests with regular and diabetic milk chocolate formulated with stevioside or sucralose and polydextrose/lactitol. Crucial attributes responsible for the lower acceptability of the sucrose-free milk chocolates were believed to be sweetness, bitterness, melting rate, grittiness and adherence. Shah et al. (2010) investigated sucrose-free milk chocolates, sweetened with steviol glycosides and containing polydextrose, different types of inulin and maltodextrin, in relation to their physicochemical and sensory properties. Although, substantial quality differences with the control sample were observed, the authors recommended inulin with a high degree of polymerisation for sucrose-free chocolate formulations. A similar approach led Farzanmehr and Abbasi (2009) to optimised mixtures of bulking agents (inulin/polydextrose 75/25 and 50/50) for sucralose-sweetened milk chocolate. In the abovementioned sensory studies, the physicochemical properties of the bulking agents were discarded.

To our knowledge, there are no studies about the relationship between the most important physicochemical properties of bulking agents and the sensory attributes of corresponding sugar-free fat-based suspensions. This paper targets to map textural implications following sucrose replacement in fat-based suspensions by alternative bulking agents, *i.e.* isomalt ST, fructo-oligosaccharides (FOS), two types of inulin and rice starch. Firstly, the bulking agents were characterised for moisture content, particle density, particle morphology, particle size, solid state, solubility and viscosity. Next, fat-based suspensions formulated with these bulking agents were subjected to physiochemical characterisation, *i.e.* moisture level, particle size, degree of particle agglomeration and flow properties, and sensory analysis. The latter focused on grittiness and adherence, two main quality defects in the product type of investigation.

2. Materials and methods

2.1. Bulking agents

2.1.1. Types

Refined sucrose was supplied by Barry Callebaut (Wieze, Belgium). Isomalt ST (grade PF), Remy DR, Orafti P95, Orafti HSI and Orafti HP were provided by Beneo (Oreye, Belgium). Remy DR is a native rice starch. Orafti P95 consists of fructo-oligosaccharides (94% on dry basis), with a degree of polymerisation (DP) in the range 2–8, and glucose, fructose and sucrose (6% on dry basis). Orafti HSI (inulin 1) and Orafti HP (inulin 2) can be categorised as inulins. Orafti HP does not contain remaining sugars and has an average DP $\geq$ 23. Orafti HSI has an intermediate average DP compared to Orafti P95 and Orafti HP, and contains 4% glucose, fructose and sucrose.

2.1.2. Physicochemical characterisation

The moisture content (g/100 g) was determined by Karl Fischer titration. Measurements were done in triplicate using 1 g of sample and a 719S Titrino device (Metrohm, Switzerland), the appropriate Hydranal solvent for hydrophilic substances and Hydranal titrant (Sigma-Aldrich, Bornem, Belgium). Calibration of the titrant was done using water.

The particle density ρ (g/cm^3) was measured using pycnometry. Hereto, distilled water was used to determine the volume of the pycnometer. Next, the pycnometer was filled with hexane to measure the density of this solvent. Finally, the pycnometer was filled with 10 g of bulking agent and hexane. Assuming that the bulking agent was insoluble in hexane, sample's density could be calculated. Prior to weighing, the pycnometer holding the different samples was incubated for 30 min at 25 °C.

The particle size distribution (PSD) was measured in triplicate using laser light diffraction (Malvern Mastersizer S, Malvern Instruments Ltd., Worcestershire, UK) equipped with a 300 RF lens to measure particles in the range of 0.05–900 μm. From the recorded PSD, the characteristic parameters D_{43} (μm), *i.e.* volume-weighed mean diameter, and $D_{v,0.9}$ (μm), *i.e.* 90th percentile of the cumulative, volume-weighed size distribution, were derived.

The solid state was determined using the Q1000 differential scanning calorimeter (TA Instruments, New Castle, Delaware, USA) equipped with a refrigerated cooling system. Nitrogen was used as purge gas. The cell constant was set with indium (TA Instruments). In addition to indium, azobenzene (Sigma-Aldrich, Bornem, Belgium) and undecane (Acros Organics, Geel, Belgium) were used for temperature calibration. Samples (3.0–6.0 mg) were sealed in hermetic aluminium cups (TA Instruments) and an empty pan was used as a reference. The applied time-temperature protocol was: equilibration at 20.0 °C and holding for 10 min followed by heating at a rate of 5.0 °C/min to 200.0 °C. Melting ($T_{m,\,onset}$, $T_{m,\,max}$, $T_{m,\,offset}$ and ΔH_m) and glass transition parameters ($T_{g,\,onset}$, $T_{g,\,mid}$ and $T_{g,\,offset}$) were determined using Universal Analysis 2000 (TA Instruments). Measurements were done in triplicate.

The bulking agents were visualised using a Jeol JSM 7100F scanning electron microscope (JEOL Ltd, Tokyo, Japan). They were mounted on aluminum stubs, vitrified in a nitrogen slush and transferred under vacuum conditions into the cryo-preparation chamber (PP3010T Cryo-SEM Preparation System, Quorum Technologies, UK) conditioned at −140 °C. Subsequently, the sample was sublimated for 5–10 min at −70 °C to remove frost artefacts, sputter-coated with platinum using argon gas, transferred to the SEM stage at −140 °C and electron beam targeted at 3 keV.

Solubility (g/100 g) at 25 °C was estimated by solubilisation in distilled water under stirring using a magnetic stirrer. Hereby, bulking agent was dosed in steps of 5 g/100 g until solubilisation was not possible anymore. In addition, solubilised state was verified the day after preparation.

Viscosity measurements of solutions (20 g/100 g) of bulking agents in distilled water at 37.0 °C were performed using an AR2000ex rheometer (TA Instruments, New Castle, USA) equipped with conical concentric cylinders. After a temperature equilibration

of 15 min, the shear rate was increased from 0.01 s^{-1} to 100 s^{-1} with 10 measuring points per decade, and the shear stress was recorded. Newtonian behaviour was verified and viscosity η (mPa.s) was calculated as the slope of the shear stress – shear rate data. Triplicate measurements were performed.

2.2. Fat-based suspensions

2.2.1. Productions

The reference fat-based suspension consisted of 46.0% refined sucrose, 33.4% palm-/palm kernel-based fat (Palkena S 292, Fuji Oil Europe, Ghent, Belgium), 20.0% skim milk powder (Barry Callebaut) and 0.6% soy lecithin (Barry Callebaut). Sucrose replacement by alternative bulking agents was done on volume basis.

The suspensions (1 kg) were prepared at Cacaolab bvba (Evergem, Belgium). Prior to mixing, the fat was melted at 45 °C. A Hobart mixer (Hobart, Troy, Ohio, US) was used for mixing the bulking agent, skim milk powder and a part of the molten fat (fat content of pre-mix: 27.0%) for 10 min at low speed. Simultaneously, the bowl was continuously heated with a heat gun. Next, the particle size of the mixture was reduced using a 3-roll refiner (Exakt 80S Apparatebau GmbH & Co. KG, Norderstedt, Germany) with gap setting 2:1 at 35 °C and 400 rpm. The refined product was liquefied at 45 °C through addition of the remaining fat and lecithin in the Stephan mixing device UMC 5 (Stephan Food Service Equipment GmbH, Hameln, Germany) for 10 min at 1500 rpm. Part (300 g) of the compound was stored in sealed plastic containers at ambient temperature prior to physicochemical characterisation. The remaining part (700 g) was poured in plastic moulds for shaping in discs having a diameter of 35 mm and a height of 5 mm. The discs were shock-cooled at 5 °C for 30 min and demoulded. They were matured at 18 °C for two weeks prior to sensory analysis.

2.2.2. Physicochemical characterisation

Dry matter content (g/100 g) determination of the fat-based suspension was based on weight loss after drying in a hot air oven at 105 °C. Measurements were done in triplicate.

The particle size distribution of the suspended particles in the continuous fat phase was measured using the Malvern Mastersizer S, as described in Section 2.1.2. Here, three subsamples of 0.5 g fat-based suspension were mixed with 10 ml of isopropanol (VWR, Leuven, Belgium), put in an oven at 45 °C for 1 h and measured five times. Simultaneously, three subsamples of each fat-based suspensions in isopropanol were subjected to a sonication treatment using an ultrasound water bath (Elmasonic S, Elma Schmidbauer GmbH, Singen, Germany) at 45 °C and 80 Hz for 15 min.

The suspended particles of the fat-based system were visualised using scanning electron microscopy, as outlined in Section 2.1.2. The dry suspended phase was obtained after solubilisation of the fat phase in isopropanol followed by filtration using a Büchner funnel, filter paper and vacuum flask connected to a vacuum source.

The flow curves of the fat-based suspensions at 40 °C were recorded in triplicate using the official ICA46-method and the AR2000ex rheometer (TA Instruments), equipped as described in Section 2.1.2. The flow data were fitted to the Casson model from which the Casson yield stress σ_{CA} (Pa) and Casson viscosity η_{CA} (Pa.s) were deduced. In addition, the apparent viscosity η^* at 50 s^{-1} was included in the data set.

2.2.3. Sensory analysis

Textural attributes, *i.e.* grittiness and adherence, were scored using a panel trained by UGent Sensolab and a 15-point hedonic scale, a score 0 indicating lowest intensity and 15 highest intensity. The panellists applied a fixed tasting protocol, consisting of biting the disc, rolling it between tongue and palate until melted and swallowing. The products were presented on a white plastic plate and labelled with a randomly selected three-digit code. Between textural evaluations, mineral water and plain crackers were used as palate cleansers.

2.2.4. Statistical analysis

The linear correlation between measured parameters was quantified by the Pearson correlation coefficient. Statistical analysis was performed using SPSS 22.0 software (SPSS Inc., Chicago, IL). Particle size distribution, flow and sensory parameters of fat-based suspensions were subjected to variance analysis (ANOVA) at 5% significance level. Testing for homogeneity of variances was performed using the Levene Test. When the conditions for homogeneity of variances were fulfilled, Tukey test was used to determine differences among the samples. In case variances were not homogeneous, Games–Howell testing was performed.

3. Results and discussion

3.1. Bulking agents

Important physicochemical parameters of sucrose, isomalt ST, FOS, inulin 1, inulin 2 and rice starch are shown in Table 1. Sucrose likely owes its high suitability as bulking agent in fat-based suspensions to a low moisture content, crystalline nature, high particle density, high solubility and low viscosity. All sampled alternative bulking agents contained more moisture. For isomalt ST (2.5 g/100 g), this can be attributed to its chemical composition, being a proportional mixture of anhydrous α-D-glucopyranosyl-1-6-sorbitol and α-D-glucopyranosyl-1-6-mannitol dihydrate (Borde and Cesàro, 2001). Its solid state can be deduced from the thermal behaviour showing two endothermic, first-order transitions (Fig. 1A), *i.e.* a small peak (26 J/g) in the range 90–102 °C due to the loss of water of crystallisation (Borde and Cesàro, 2001) and a large peak (79 J/g) at 105–151 °C reflecting the crystalline to liquid phase transition. Conversely, crystalline sucrose shows a very large (136 J/g) and narrow (T_m: 184–197 °C) melting peak. Its low moisture content is because sucrose only interacts with water molecules at the crystal surfaces through hydrogen bonds inhibiting moisture uptake (Bell and Labuza, 2000). The amorphous nature of FOS (3.7 g/100 g), inulin 1 (3.4 g/100 g) and inulin 2 (3.1 g/100 g) is responsible for their elevated moisture levels. In contrast to crystalline substances, the hydroxyl groups of amorphous materials are more exposed to the environment making them more hygroscopic (Rahman and Labuza, 1999). Upon heating in the differential scanning calorimeter, a glass transition is observed as a second-order transition, *i.e.* shift in base line due to a glassy to rubbery state transition (Liu et al., 2006), as shown in Fig. 1B for FOS (T_g: 54–62 °C), inulin 1 (T_g: 54–64 °C) and inulin 2 (T_g: 88–98 °C). Below the glass transition temperature range, these ingredients are in the glassy solid state, characterised by a liquid-like structure with an extremely high viscosity. Thus, in comparison to the crystalline phase, the molecular arrangement within the glassy state is rather disordered (Liu et al., 2006). Glassy materials are kinetically in a non-equilibrium state and can approach a more stable state, when stored below the glass transition temperature. This phenomenon is called physical ageing or enthalpic relaxation and is due to the local molecular motion of (parts of) molecules (Kim et al., 2003). Enthalpic relaxation can be deduced from an overshoot of the baseline during the glass transition (Wungtanagorn and Schmidt, 2001) and was observed for FOS and to a lower extent for inulin 1. Inulin 2 has a higher T_g range than FOS and inulin 1 due to its higher molecular weight (Le Meste et al., 2002). Based on the compositional data provided, one could have expected a somewhat larger difference in T_g range between FOS and inulin 1 but the

Table 1
Physicochemical characteristics of powdery bulk sweeteners and their solutions.

Parameter	Sucrose	Isomalt ST	FOS	Inulin 1	Inulin 2	Rice starch
Water content (g/100 g)	0.2 ± 0.1	2.5 ± 0.1	3.7 ± 0.1	3.4 ± 0.1	3.1 ± 0.1	12.3 ± 0.1
$T_{m,\ onset1}$ (°C)	184.1 ± 0.2	89.9 ± 0.4	n.d.	n.d.	n.d.	x
$T_{m,\ max1}$ (°C)	191.3 ± 0.3	97.4 ± 0.1	n.d.	n.d.	n.d.	x
$T_{m,\ offset1}$ (°C)	196.9 ± 0.9	101.5 ± 0.5	n.d.	n.d.	n.d.	x
ΔH_{m1} (J/g)	136 ± 3	26 ± 0	n.d.	n.d.	n.d.	x
$T_{m,\ onset2}$ (°C)	n.d.	105.4 ± 0.4	n.d.	n.d.	n.d.	x
$T_{m,\ max2}$ (°C)	n.d.	136.6 ± 0.0	n.d.	n.d.	n.d.	x
$T_{m,\ offset2}$ (°C)	n.d.	150.7 ± 0.0	n.d.	n.d.	n.d.	x
ΔH_{m2} (J/g)	n.d.	79 ± 0	n.d.	n.d.	n.d.	x
$T_{g,\ onset}$ (°C)	n.d.	n.d.	54.3 ± 0.7	54.0 ± 0.5	88.2 ± 0.5	x
$T_{g,\ mid}$ (°C)	n.d.	n.d.	58.5 ± 0.4	60.8 ± 0.2	94.0 ± 0.4	x
$T_{g,\ offset}$ (°C)	n.d.	n.d.	61.6 ± 0.4	64.4 ± 0.2	98.0 ± 0.6	x
ρ (g/cm^3)	1.56 ± 0.01	1.53 ± 0.01	1.45 ± 0.01	1.47 ± 0.00	1.42 ± 0.01	1.38 ± 0.01
D_{43} (μm)	51 ± 1	45 ± 5	73 ± 2	89 ± 1	98 ± 7	55 ± 6
$D_{v,0.9}$ (μm)	114 ± 2	104 ± 9	144 ± 3	163 ± 2	189 ± 13	105 ± 2
Solubility (g/100 g)	65–70	20–25	75–80	30–35	n.s.	n.s.
η (mPa.s) at 37 °C	1.43 ± 0.03	1.52 ± 0.02	1.57 ± 0.02	1.65 ± 0.01	x	x

x ± y: average ± standard deviation; n.d.: not detected; x: not determined; n.s.: not soluble.

difference in the amount of moisture, acting as plasticizer (Liu et al., 2006), and the presence of remaining sugars (Roos, 1995) probably counteracted the effect of the difference in molecular weight. Rice starch showed the highest moisture level (12.3 g/100 g). In starch granules, water is mainly associated with their amorphous regions (Zeleznak and Hoseney, 1987). In fact, starch is a polymer comprising crystalline and amorphous amylopectin regions, interspersed with amorphous amylose chains. In Fig. 1, the broad and ill-defined glass transition of the latter starts beyond 100 °C. Melting of the crystalline amylopectin can be expected to occur at higher temperatures than the glassy-rubbery state transition, as was observed by Singh et al. (2000).

In regard to processing of sugar-free fat-based suspensions, Zumbé et al. (2001) stated that the conching temperature of chocolate should be kept below that at which moisture inherently present is released, hereby, avoiding undesirable textural defects. For isomalt ST, this means below the dehydration temperature range, and for FOS, inulin 1, inulin 2 and rice starch below the glass transition temperature range. Therefore, the processing temperature of the sugar-free fat-based suspensions was chosen to be maximum 45 °C in our study.

The bulking agents were visualised using scanning electron microscopy (Fig. 2) from which they could be clustered in three groups according to their solid state. First, the crystalline materials sucrose and isomalt ST can be described as angular shaped solids with a rather broad particle size distribution (Fig. 2A and B). The latter is attributed to a refining pre-treatment (mentioned on the technical data sheet). The average particle sizes of sucrose and isomalt ST were similar, namely around 50 μm (Table 1). Second, the amorphous bulking agents FOS, inulin 1 and inulin 2 were more or less spherical (Fig. 2C, D and 2E). It can be noticed that the average particle size (73 μm, 89 μm and 98 μm) and the deviation from perfect sphericity differed according to the following sequence: FOS < inulin 1 < inulin 2. This observation can be attributed to their differences in DP, whereby a higher DP will induce a higher viscosity (Teraoka, 2002). In the production process, higher viscosities lead to larger spray droplets and longer drying times (Kleinebudde et al., 2000; Rao, 2014) during which partial coalescence can occur. Finally, semi-crystalline native rice starch granules are visualised (Fig. 2F) as predominantly polygonal with a granular size range of 2–10 μm as described in literature (Kittipongpatana et al., 2007; Wani et al., 2012). The higher recorded values for average size can be attributed to a high degree of agglomeration.

Sucrose and isomalt ST showed the highest particle density (Table 1). Compared to amorphous solids, these crystalline substances fill the space more efficiently because of their higher degree of structural order (Chancey, 2008). The lower particle density of semi-crystalline rice starch compared to amorphous FOS, inulin 1 and inulin 2 is very likely due to its substantially higher moisture level.

Sokmen and Gunes (2006) suggested that sucrose replacement on volume basis may more accurately reflect the impact of alternative bulking agents on the flow properties of fat-based suspensions than replacement on mass basis. In the latter case, bulking agents having different particle densities will directly affect the particle volume fraction, an important factor determining the flow parameters of suspensions. It should be stressed that also other parameters, *e.g.* particle shape and size distribution, surface roughness, wetting properties of the suspended particles in the liquid fat phase, and emulsifier-particle and particle-particle interactions, affect suspension rheology (Servais et al., 2002). Nevertheless, it was chosen to perform a sucrose substitution by isomalt ST, FOS, inulin 1 and 2, and rice starch on volume basis via the particle densities in Table 1.

Solubility of bulking agents in water depends on the ability of water molecules to form hydrogen bonds with the bulking agents (Davis, 1995). The determined solubility ranges (Table 1) of sucrose, isomalt ST, FOS and inulin 1 correspond to literature (O'Donnell and Kearsley, 2012). Inulin 2 and rice starch were only slightly (<5 g/100 g) or not soluble in water. For comparison reasons, 20 g/100 g solutions were prepared and subjected to viscosity measurements. The viscosity of the solutions at body temperature, reflecting mouthfeel, increased in following order; sucrose < isomalt ST < FOS < inulin 1. The difference in the viscosity of FOS and inulin 1 solutions could be attributed to a distinct DP (Teraoka, 2002). In this study, it was of interest to see if the solubility and viscosity of the bulking agents and their solutions, respectively, would affect the mouthfeel of the corresponding fat-based suspensions.

3.2. Fat-based suspensions

The following part focuses on important physicochemical parameters of the fat-based suspensions formulated with the bulking agents under investigation. Relevant properties with regard to mouthfeel are moisture content, particle size and flow parameters (Table 2). High moisture levels might increase the thickness of fat-based suspensions (Beckett, 2011), hereby, inducing an adherent

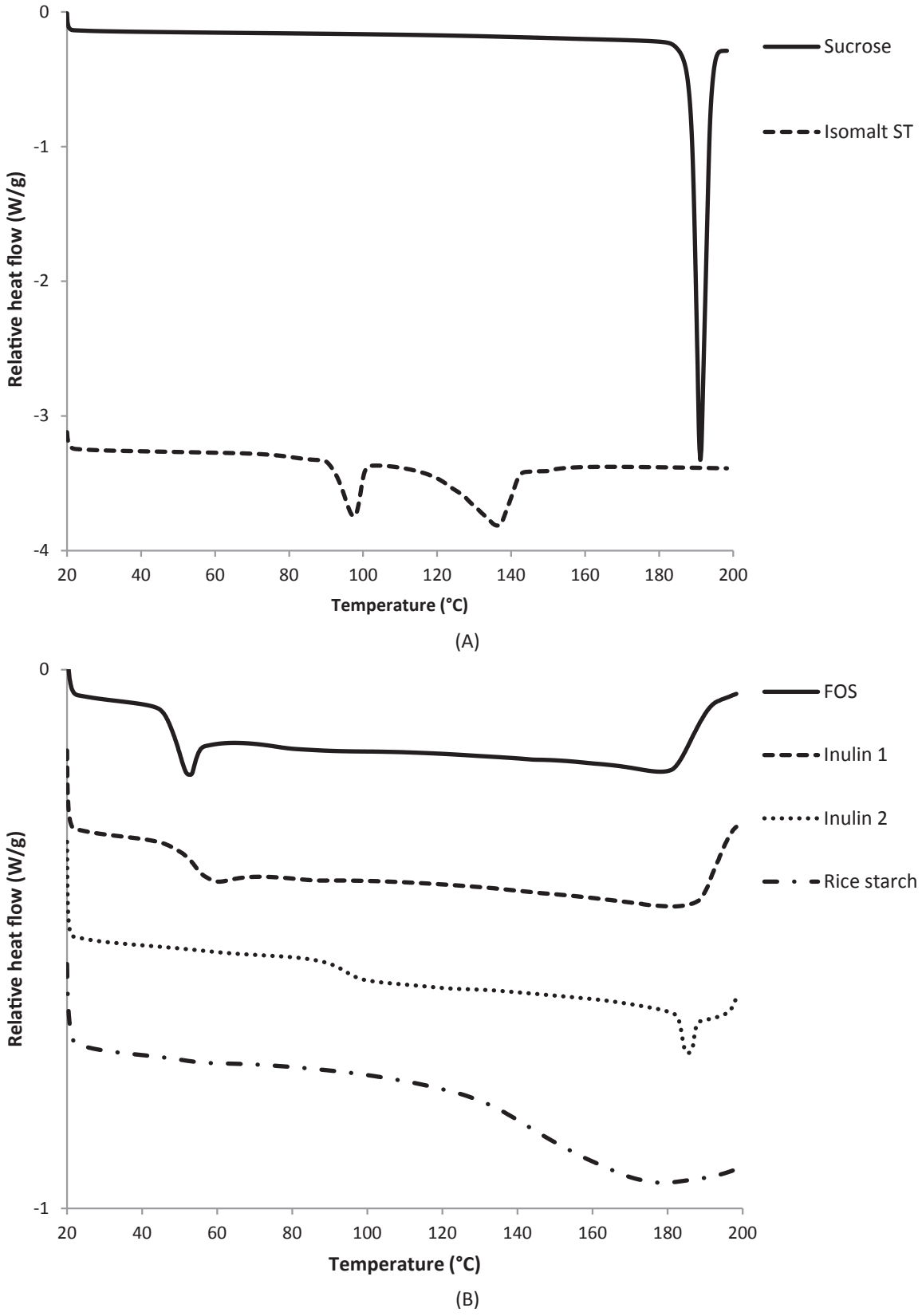

Fig. 1. Thermal behaviour of (A) sucrose and isomalt ST, and (B) FOS, inulin 1, inulin 2 and rice starch.

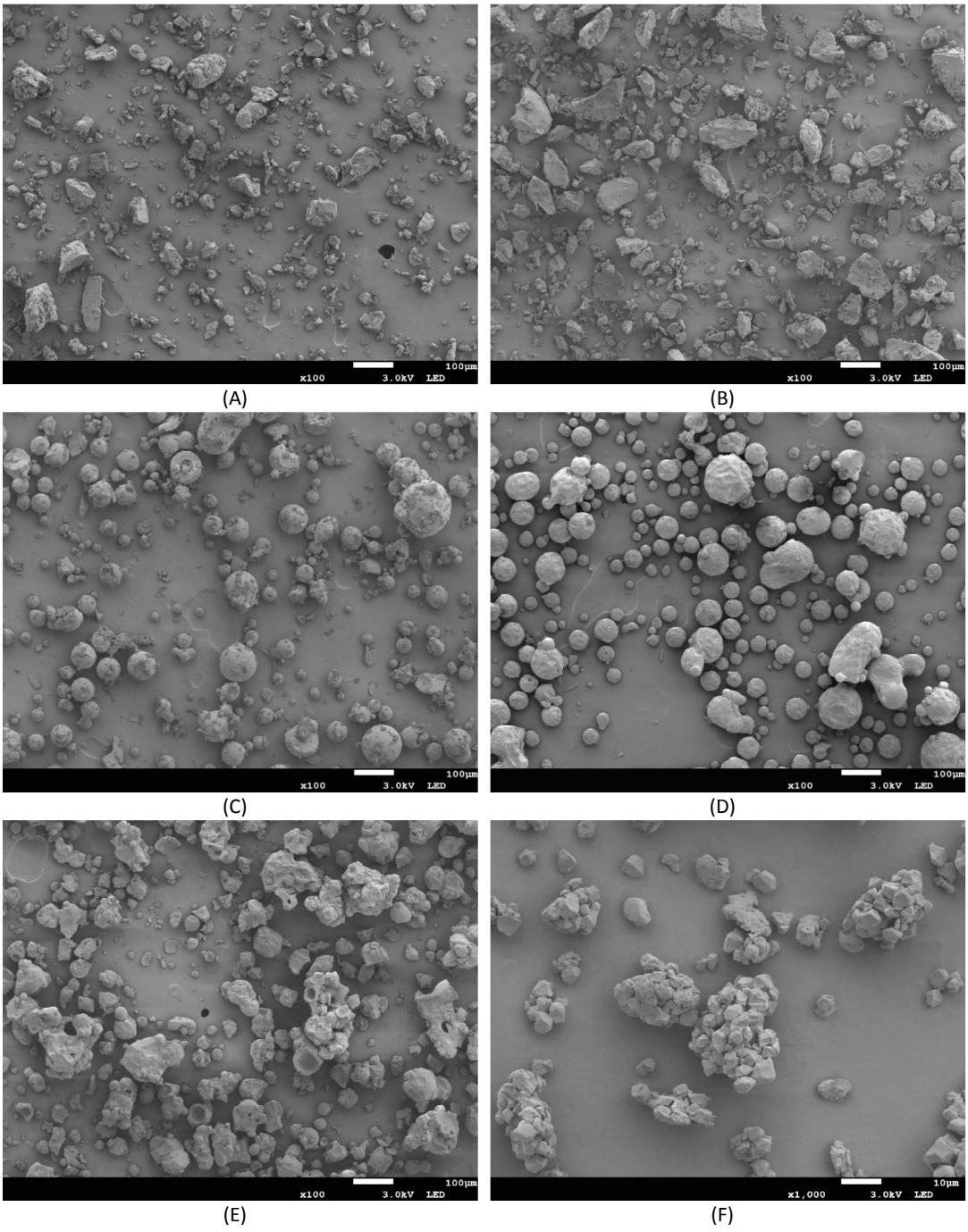

Fig. 2. Particle morphology of (A) sucrose, (B) isomalt ST, (C) FOS, (D) inulin 1, (E) inulin 2 and (F) rice starch.

Table 2
Physicochemical characteristics of fat-based suspensions formulated with different bulk sweeteners.

Parameter	Sucrose	Isomalt ST	FOS	Inulin 1	Inulin 2	Rice starch
Water content (g/100 g)	0.8 ± 0.0^{A}	1.3 ± 0.1^{B}	2.0 ± 0.1^{c}	2.2 ± 0.1^{c}	2.1 ± 0.0^{c}	4.4 ± 0.1^{D}
D_{43} (µm)	14.4 ± 0.3^{A}	33.3 ± 0.5^{C}	16.0 ± 1.4^{AB}	18.6 ± 0.4^{B}	19.2 ± 0.7^{B}	18.1 ± 0.5^{B}
$D_{v,0.9}$ (µm)	33.1 ± 0.7^{A}	68.2 ± 1.0^{C}	33.8 ± 2.4^{A}	40.1 ± 0.6^{B}	39.5 ± 1.2^{B}	38.2 ± 1.0^{B}
$\Delta D_{43\text{-sonication}}$ (%)	−8	−33	−12	−7	−27	−33
$\Delta D_{v,0.9\text{-sonication}}$ (%)	−5	−24	−8	−5	−23	−30
σ_{CA} (Pa)	3.6 ± 1.7^{C}	14.7 ± 1.9^{D}	0.3 ± 0.0^{B}	0.2 ± 0.1^{AB}	0.1 ± 0.0^{A}	0.0 ± 0.0^{A}
η_{CA} (Pa.s)	1.21 ± 0.06^{A}	2.32 ± 0.11^{C}	1.29 ± 0.02^{A}	1.28 ± 0.01^{A}	1.56 ± 0.02^{B}	6.37 ± 0.07^{D}
η^* at 50 s^{-1} (Pa.s)	1.86 ± 0.18^{B}	3.92 ± 0.23^{C}	1.50 ± 0.02^{A}	1.46 ± 0.01^{A}	1.66 ± 0.01^{B}	6.63 ± 0.07^{D}

^{A,B,C,D}: different superscripts indicate significant differences between fat-based suspensions per parameter ($p < 0.05$).

(sticky) mouthfeel. The sucrose-sweetened fat-based suspension had the lowest moisture content followed by the one with isomalt ST, those formulated with FOS, inulin 1 and inulin 2, and ultimately the one with rice starch. The moisture levels of bulking agents correlated highly ($R = 0.98$) with those of the respective fat-based suspensions.

In fat-based suspensions such as chocolate, a maximum particle size of 35 μm is targeted to prevent a gritty mouthfeel (Servais et al., 2002). The 90th percentile of the volume-weighed distribution ($D_{v,0.9}$) of the suspensions with sucrose (33 μm) and FOS (34 μm) fitted this specification, in contrast to the ones with isomalt ST (68 μm), inulin 1 (40 μm), inulin 2 (40 μm) and rice starch (38 μm). Crystalline materials, such as sucrose and isomalt ST, are brittle and fragment easily in response to the applied stresses during roll-refining (Ziegler and Hogg, 2009). Although amorphous materials are considered brittle below their glass transition temperature range as well, they might be more difficult to grind because of their structural features (Blanshard, 1995). Such an effect of the solid state was observed by Aguilar and Ziegler (1995) when comparing particle sizes of refined chocolate masses

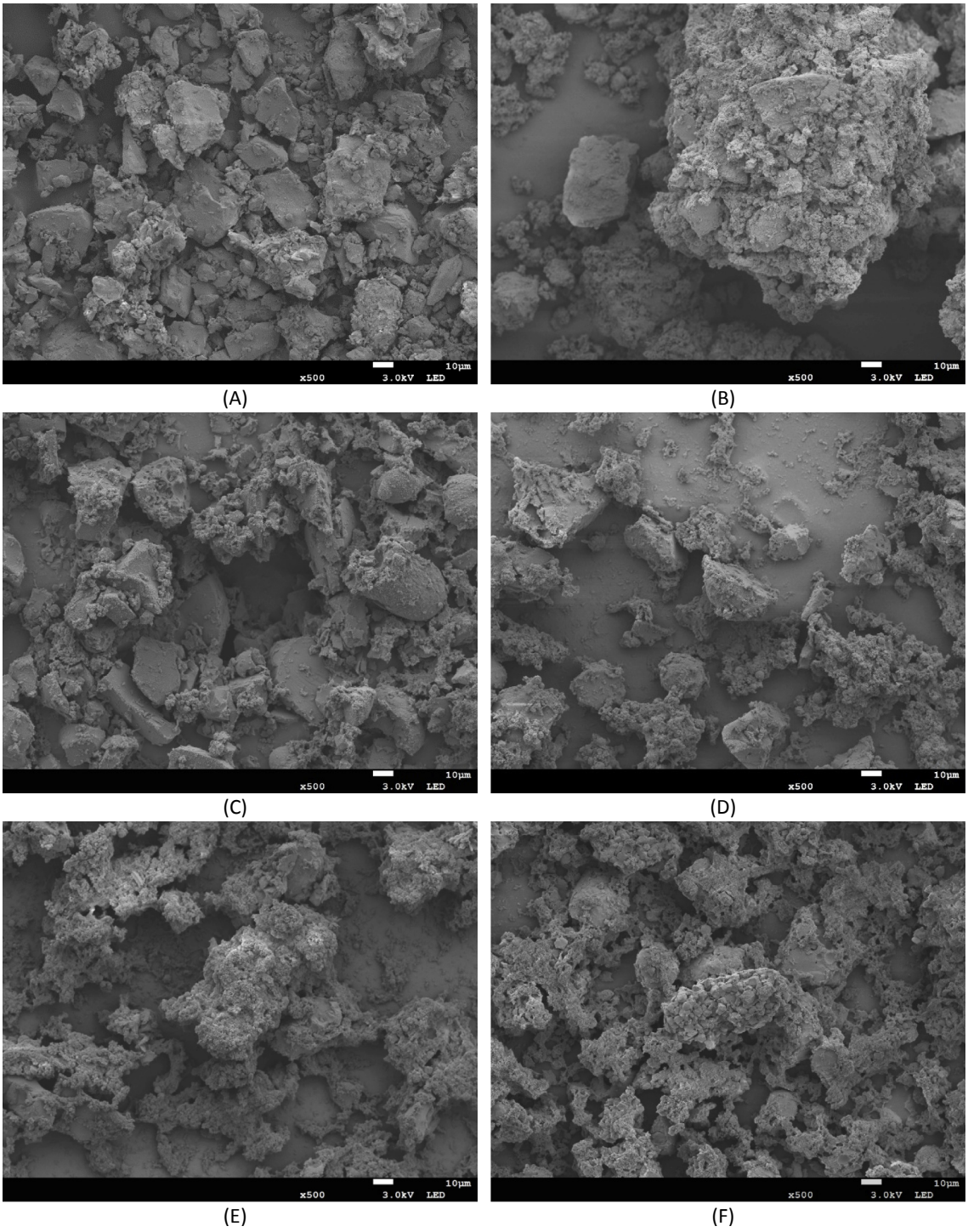

Fig. 3. Solid phase of the fat-based suspensions formulated with (A) sucrose, (B) isomalt ST, (C) FOS, (D) inulin 1, (E) inulin 2 and (F) rice starch.

comprising crystalline and amorphous lactose. FOS seemed to perform better than inulin 1 and inulin 2, which might be due to its lower fineness and/or high degree of enthalpic relaxation, *i.e.* an indication of a more stable state. The PSD parameters D_{43} and $D_{v,0.9}$ of the bulking agents correlated poorly ($R = -0.35$ and $R = -0.34$, respectively) with those of the fat-based suspensions, although the refining settings were similar. However, when discarding isomalt ST (crystalline hydrated sugar alcohol) and rice starch (semi-crystalline polymer) high correlation coefficients ($R = 0.98$ and $R = 0.86$, respectively) were observed. This clearly shows that also the molecular structure and microstructural properties have to be taken into account. Moreover, a high degree of agglomeration was observed in fat-based suspensions formulated with isomalt ST, inulin 2 and rice starch. This observation was based on the decrease in PSD parameters following a sonication treatment and was supported by SEM images (Fig. 3). A very pronounced agglomeration of the solids of the suspensions formulated with isomalt ST (Fig. 3B), inulin 2 (Fig. 3E) and rice starch (Fig. 3F) was seen, in contrast to those with sucrose (Fig. 3A), FOS (Fig. 3C) and inulin 1 (Fig. 3D). In case of isomalt ST, roll-refining might have induced dehydration (Borde and Cesàre, 2001) or amorphisation of the crystals (Elamin et al., 1994; Kruger, 2011; Ndindayino et al., 2002) resulting in agglomeration due to sticky patches. The fact that the degree of agglomeration in fat-based suspension sweetened with crystalline sucrose is limited indicates that the first explanation is the more plausible one. Sucrose also becomes partially amorphous during roll-refining (Ziegler and Hogg, 2009) but does not contain water of crystallisation. Thus, it is expected that isomalt LM, *i.e.* dried version of isomalt LM, would behave similar to sucrose during refining. In fact, isomalt LM has already been claimed to ensure outstanding chocolate quality (O'Donnell and Kearsley, 2012).

Fat-based suspensions show shear-thinning behaviour and their flow behaviour is often described by the Casson yield value σ_{CA} and plastic viscosity η_{CA} (Beckett, 2011; Servais et al., 2003). The flow parameters of liquid suspensions are highly impacted by the particle size of the solid phase. Hereby, larger particle sizes correspond to lower yield stresses and plastic viscosity (Afoakwa, 2011). Regarding σ_{CA}, an opposing trend ($R = 0.89$ and $R = 0.91$ for D_{43} and $D_{v,0.9}$, respectively) was observed, while for η_{CA} no correlation ($R < 0.1$) with the particle size parameters was found, indicating that other factors play an important role. Here, moisture was considered as an important factor, as was stated by Afoakwa et al. (2007). Compared to the sucrose-sweetened fat-based suspensions (3.6 Pa), the systems comprising FOS, inulin 1, inulin 2 and rice starch had a significantly lower σ_{CA} (0.0–0.3 Pa). Conversely, the isomalt ST sample (14.7 Pa) showed a substantially higher σ_{CA} than the reference suspension. This mismatched trend between moisture level and σ_{CA} is likely linked to the structural organisation of the water molecules in the suspension matrix. Following refining, the moisture in the suspensions formulated with FOS, inulin 1, inulin 2 and rice starch is largely associated with the amorphous parts of the bulking agents, while in the one with isomalt ST, possibly a release of moisture in the fat-continuous matrix occurred (Borde and Cesàre, 2001). This difference in how moisture is present in the suspension might also explain the somewhat higher η_{CA} in case of the isomalt ST sweetened suspension (2.3 Pa s compared to 1.2–1.6 Pa s). The very high η_{CA} (6.4 Pa s) of the rice starch sample might be explained at least partly by a highly inefficient packing of the particles (Servais et al., 2002). SEM imaging showed that the small rice starch granules were not or hardly refined during the grinding step (Fig. 3F). Additionally, poor wetting of the starch granules by the lipid phase will induce high viscosities values.

The apparent viscosity η^* at 50 s^{-1} corresponds better to mouthfeel than the abovementioned Casson parameters. In fact, shear rates applied during chewing range from 10 to 100 s^{-1} (Vanderdeelen & Van Der Meeren, 1998). Assuming no interaction with saliva, one could expect the FOS and inulin 1 suspensions (~1.5 Pa s) to be perceived the least adherent, more particularly somewhat less adherent than the suspensions formulated with sucrose and inulin 2 (1.7–1.9 Pa s) but a lot less and extremely less adherent than the one containing isomalt ST (3.9 Pa s) and rice starch (6.6 Pa s), respectively.

Table 3
Grittiness and adherence of fat-based suspensions formulated with different bulk sweeteners.

Parameter	Sucrose	Isomalt ST	FOS	Inulin 1	Inulin 2	Rice starch
Grittiness	0^{A}	3^{B}	7^{CD}	6^{C}	8^{D}	10^{E}
Adherence	0^{A}	2^{B}	8^{C}	12^{D}	15^{E}	15^{E}

Common defects in fat-based suspensions following sugar replacement by low-calorie sweeteners are grittiness and adherence (O'Donnell and Kearsley, 2012). These two attributes were evaluated through sensory analysis (Table 3). The sucrose-sweetened suspension was perceived as the least gritty and least adherent one with a good runner-up being the one formulated with isomalt ST. The suspensions comprising FOS, inulin 1, inulin 2 and rice starch suffered from a moderate to high degree of grittiness and adherence. Furthermore, the texture of the inulin 2 and rice starch suspensions could be considered as the worst. The sensory attributes of the fat-based suspensions correlated poorly with their particle size distribution or flow parameters ($|R| = 0.16–0.68$). This can be explained by the fact that also the interaction between saliva and the suspended particles have on impact on the mouthfeel. Hereby, the least soluble bulking agents (inulin 2 and rice starch) resulted in a high degree of grittiness and adherence. Once the lipid phase has molten, lumps of suspended particles are formed, due to the lack of solubility, which are responsible for grittiness and apparently also adherence. In this experimental set-up, grittiness and adherence correlated strongly ($R = 0.93$). The moisture content of the fat-based suspension, influenced by the solid state of the corresponding bulking agents, correlated well with grittiness ($R = 0.87$). Omitting insoluble inulin 2 and rice starch from the data set in order to challenge a possible link between the viscosity of solutions of bulking agents and adherence of corresponding fat-based suspensions, a strong positive correlation was found ($R = 0.96$). Conversely, adherence and apparent viscosity of the suspensions correlated poorly ($R = -0.55$).

4. Conclusions

This aim of this paper was to investigate which physicochemical properties of sucrose, isomalt ST, FOS, inulin and rice starch affect the textural characteristics of the corresponding fat-based suspensions. Although it is assumed that the flow behaviour and particle size are important for the mouthfeel of fat-based suspensions, this study showed that the solid state and solubility of bulking agents, and the viscosity of their solutions are significantly affecting grittiness and adherence of alternatively sweetened fat-based suspensions. In fact, low-soluble bulking agents, *i.e.* inulin with a high DP and rice starch, induced a high degree of grittiness and adherence in the end products. For sucrose, isomalt ST, FOS and inulin with a relative low DP, the viscosity of their solutions and the adherence of the corresponding fat-based suspensions were positively correlated. The moisture level of bulking agents influenced by their solid state seemed to have a direct impact on grittiness of the suspensions. These relationships might even allow a more efficient

screening of newly developed bulking agents in sucrose-free or sucrose-reduced fat-based suspensions.

Conflicts of interest

None.

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