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Universal strategy for sugar replacement in foods?

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ABSTRACT

In this paper we present a sugar replacements strategy, derived from physical theory, which we think is applicable for a wide range of food categories. The strategy is based on the hypothesis that reformulated foods must mimic the values of two physical characteristic numbers, related to the plasticizing and hygroscopic properties of sugar, to reproduce the texture of a sugar-rich product. We will show the validity of the strategy for reformulated biscuits, using experimental determination of physical properties of dough and baked biscuits, as well as sensorial evaluation by a trained panel. Our investigations shown that the majority of these physical and sensorial attributes correlate with the two physical characteristic numbers. Furthermore, multiple reformulations can be defined which are close to the reference product (in terms of the two physical characteristic numbers), which are indeed scored similar by the trained sensory panel. Hence, our strategy also leaves room for further optimization of the reformulated food towards dietary fiber content, laxative properties or sweetness.

1. Introduction

To counter the prevalence of obesity food science and industry are striving for reducing the caloric content of food products, or lowering the amount of added/free sugars, for example via replacing sugars by low-energy carbohydrates and/or high intensity sweeteners (Struck et al., 2014; Yeung et al., 2017; McCain et al., 2018; Hutchings et al., 2019; Sahin et al., 2019; Velázquez et al., 2021; Deliza et al., 2021; McKenzie & Lee, 2022). Often, this is part of an overall strategy to improve the whole nutritional profile of foods.

However, at the same time the sugar replacement method should retain the texture, taste and shelf life of the original food product, which all are significantly influenced by sugars (Pareyt & Delcour, 2008; Wilderjans et al., 2013). Hence, the development of a sound sugar replacement strategy is hampered by an incomplete knowledge of the multiple functionalities of sugars in foods. In sweet bakery foods the biggest challenge of sugar replacement is getting the right texture (van der Sman & Renzetti, 2019; Van der Sman & Renzetti, 2020).

Most current strategies concerning sugar reduction replace sucrose by one single other ingredient via trial-and-error approach, with little use of knowledge about sugars physical properties (Kweon et al., 2009; Laguna, Primo-Martín, et al., 2013,b; Struck et al., 2014; Rojo-Poveda et al., 2020; Javanmardi et al., 2020; Marzec et al., 2021; Roze et al., 2021; Di Cairano et al., 2021).

Our recent reviews on sucrose functionality in biscuits and cakes (van der Sman & Renzetti, 2019; Van der Sman & Renzetti, 2020) show that the physical properties of sucrose modulate many phenomena important for the texture of food: for example the rheology, the phase transitions of biopolymers, and the water distribution over different phases.

We have shown that most of the functionalities of sugar present in biscuit can be linked to the physical behaviour of sugars acting either as plasticizer or as humectant (van der Sman & Renzetti, 2019). Here, we pose that these functionalities can be quantified by only two characteristic numbers: $\varphi_{w,eff}$ and χ_{eff}, which quantify a) the plasticizing via the volumetric density of hydrogen bonds, and b) the hygroscopicity via the volume-averaged Flory-Huggins interaction parameter. This proposition is based on theories our group has developed in the last decade on how to quantify the plasticizing and hygroscopic properties of sugars and their replacers in complex food matrices (van Der Sman, 2013; van der Sman, 2013, 2016, 2017, 2019; Van Der Sman, 2018; Van der Sman, 2019; Van der Sman & Mauer, 2019; van der Sman et al., 2020; Renzetti et al., 2020, 2021).

In various papers we have shown that $\varphi_{w,eff}$ controls the solutions viscosity (Van der Sman & Mauer, 2019), the mixture glass transition temperature T_g (van Der Sman, 2013), as well as the melting transitions of biopolymers (van der Sman, 2016). The latter property is shown for starch gelatinization (Van der Sman & Mauer, 2019; Renzetti et al.,

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2021) and for denaturation of proteins, like gelatin, soy, egg white (Renzetti et al., 2020). The fact that $\varphi_{w,eff}$ controls both the glass transition and the melting temperature is not obvious, but it has been put forward earlier (Bell & Hageman, 1996; Coppola et al., 2012). An explanation of this fact at the molecular level is not yet available, but we accept it as an empirical fact.

A list of sugar functionality in biscuits, as discussed in our review (van der Sman & Renzetti, 2019), is summarized in Table 1, where we have indicated whether it is governed by either $\varphi_{w,eff}$ and χ_{eff}. Some of the functionalities of sucrose are related to its crystallinity (as indicated by the mnemonic *Xtal*), but these functionalities are only relevant in the mixing method, which includes a creaming step where sucrose crystals are mixed with fat first. At industrial scale often the all-in-one mixing method is applied (van der Sman & Renzetti, 2019), where the crystalline properties are less relevant. This is fortunate, as the crystalline and dissolution properties are very specific for each sugar and sugar replacer (Van der Sman, 2017; van der Sman et al., 2020). Hence, based on our recent findings, we have formulated the following sugar replacement strategy, holding for sweet bakery products:

> A sugar-reduced bakery product will have a similar texture if one can exactly match the values of $\varphi_{w,eff}$ and χ_{eff} from the original product.

While applying this strategy it should be understood that similar dough preparation, processing and baking conditions should be applied to all formulations. Otherwise, processing variations will also lead to texture variations, for example due to differences in sugar dissolution, gluten hydration/development, and moisture loss during baking.

Other sensorial traits like color and sweetness of the baked biscuits one can tune relatively independently with small amounts of ingredients, like high-intensity sweeteners or reducing sugars like fructose, which are expected not to influence much the textural properties (van der Sman & Renzetti, 2019; Van der Sman & Renzetti, 2020). Alternatively, food industry can tackle the sweetness target, via industry-wide gradual reduction of sweetness levels in foods (Deliza et al., 2021; Hutchings et al., 2019). For clarity, this study targets the sugar replacement for textural functionality only.

Hence, in this paper we will test this sugar replacement strategy for the case of biscuits. A range of reformulated biscuits have been produced, varying in values of χ_{eff} and $\varphi_{w,eff}$, but several having values near that of our reference biscuit - which follows the AACC method 10–53.01. We test whether the textural properties are indeed governed by these two parameters using measurements of relevant physical properties, as well as using sensory evaluation of the perceived textural properties with a trained panel. Above all, we test whether reformulations with values of χ_{eff} and $\varphi_{w,eff}$ most close to the reference value, indeed produce biscuits with similar texture.

Table 1
Sugar functionality for biscuit texture, with associated governing parameter.

Function during mixing	Parameter
Sheared sugar crystals lead to abrasion of fat crystals, promoting air bubble stability	Xtal
Stabilization of air bubbles via steric hindrance	Xtal
Slow dissolution allows partial hydration of gluten	χ_{eff}
Dissolved sugar competes with gluten, arabinoxylans, and damaged starch for water	χ_{eff}
Together with fat it prevents full development of hydrated gluten network	χ_{eff}
Dissolved sugar provides viscosity to the dough	$\varphi_{w,eff}$
Sugar syrup provides cohesion to the dough	$\varphi_{w,eff}$
Function during baking	
Delays its own dissolution	Xtal,$\varphi_{w,eff}$
Decrease of viscosity of dough by further dissolution	Xtal,$\varphi_{w,eff}$
Promotes spreading (via decrease of viscosity and quantity of solvent)	$\varphi_{w,eff}$
Retards bubble growth via increase viscosity if moisture evaporates	$\varphi_{w,eff}$
Regulates a_w, rate of evaporation, steam pressure	χ_{eff}
Delay gluten denaturation at higher temperatures	$\varphi_{w,eff}$
Prevents starch gelatinization	$\varphi_{w,eff}$
Promote surface cracking via recrystallization	Xtal
Sets biscuit into glassy state; renders crispiness	$\varphi_{w,eff}$

2. Theory

In this section we disclose how the two parameters characterizing the hygroscopicity and plasticizing effects, χ_{eff} and $\varphi_{w,eff}$, can be calculated. Hygroscopic properties of sugar replacers like carbohydrates and amino-acids can be described by the Flory-Huggins theory (He et al., 2006; van der Sman, 2013; Van der Sman, 2017; Van der Sman & Mauer, 2019). The study of complex food matrices showed that the Flory-Huggins interaction between solutes is small (Silva et al., 2021; van der Sman, 2013; Van der Sman, 2017), and thus the hygroscopic properties of mixtures are governed by the volume-averaged interaction parameter of all solutes with water (van der Sman, 2013). Hence, it follows:

$$\chi_{eff} = \frac{\sum_i \chi_i \varphi_{s,i}}{\sum_i \varphi_{s,i}} \tag{1}$$

Thus, χ_{eff} is the volume averaged interaction parameter of all sugar replacers (i.e. plasticizers, excluding water) present in the food formulation, with $\varphi_{s,i}$ the volume fraction of each sugar (replacer).

$\varphi_{w,eff}$ can be calculated if the glass transition temperature of the dry sugar replacer, $T_{g,s}$, is known. Following the original theories by Nakanishi and Nozaki (Nakanishi et al., 2011b,a) with correction by Pawlus and coworkers (Pawlus et al., 2012), we have shown that $T_{g,s}$ of sugar (replacers) correlates to the number of hydroxyl groups per molecule, that are available for intermolecular hydrogen bonds, $N_{OH,s}$. Their relation is (van Der Sman, 2013):

$$\frac{1}{2} - \frac{1}{N_{OH,s}} = \frac{1}{2}\frac{T_{g,s} - T_{g,w}}{T_{g,\infty} - T_{g,w}} \tag{2}$$

with $T_{g,w} = 139$ K the glass transition of water, and $T_{g,\infty} = 475$ K is the limiting glass transition temperature of a large glucose-oligosaccharide like a maltopolymer or starch.

$\varphi_{w,eff}$ accounts for the volumetric density of these available intermolecular hydrogen bonds, provided by all plasticizers in a mixture, which also includes water. Water has two available hydroxyl groups ($N_{OH,w} = 2$, which is the limit of Eq. (2)). $\varphi_{w,eff}$ is computed as follows (van der Sman, 2016; Van der Sman & Mauer, 2019):

$$\varphi_{w,eff} = \varphi_w + \sum_i \varphi_{s,i} \frac{N_{OH,s,i}}{N_{OH,w}} \frac{\nu_w}{\nu_s} \tag{3}$$

φ_w is the regular volume fraction of water, and $\varphi_{s,i}$ are the volume fractions of all plasticizers present in the formulation.

As inspired by Djabourov and coworkers (Coppola et al., 2008, 2012), we have shown that this volumetric density of (intermolecular) hydrogen bonds controls the structuring pathway of bakery products in the supplemented state diagram for heating and cooling (Renzetti et al., 2020; van der Sman & Renzetti, 2019; Van der Sman & Renzetti, 2020).

Hence, the two characterizing numbers of the hygroscopic and plasticizing behaviour of sugars and their replacers are founded by well-established physical theories. Over the last years we have established the values of these two numbers for a wide variety of sugar replacers (van Der Sman, 2013; van der Sman, 2016; Van der Sman & Mauer, 2019; Renzetti et al., 2020, 2021). With the theory and the database of $\varphi_{w,eff}$ and χ_{eff} in place, we are now in the position to formulate the sugar replacement strategy and test it on bakery products, like a biscuit.

3. Materials and methods

3.1. Materials

Soft wheat flour (Croplan 594 W, a soft red winter wheat (13.3% water, 7.6% protein, and 0.396% ash), skimmed milk powder (lactose

51%, protein 37%, fat 1%, ashes 5.7%), shortening (Crisco all-purpose (Smuckers, Orville, OH)), high-fructose corn syrup (F42), and baking powders were supplied by Mondelēz (Reading, UK). Extra fine granuled sucrose (SUC) was sourced from (Domino, Yonkers, NY, U.S.A.). Erythritol (ERY), maltitol (MALT) and xylitol (XYL) were obtained from Roquette (Lestrem, France). The oligofructose Frutalose OFP (FOS) was obtained from Sensus (Roosendaal, the Netherlands), the polydextrose (PDX) Litesse Ultra from DuPont (Wilmington, Delaware, US) and mylose (MDX) from Tereos Syral (Aalst, Belgium), which is a maltodextrin with a DE value between 20 and 42.

3.2. Model biscuit preparation

Model biscuits were prepared according to the AACC International Method 10–53.01 for wire-cut cookies. The formulation of this reference model system is given in Table 2. We note that the mixture of sodium bicarbonate and ammonium bicarbonate is acting as a baking powder, with reactions as described in (van der Sman, 2021).

The used equipment was a Hobart N50 mixer with paddle beater (Hobart, Woerden, The Netherlands), a 60 mm cookie cutter and a WP-haton type Rototherm REC 1020 baking oven (Werner & Pfleiderer Lebensmitteltechnik GmbH, Dinkelsbühl, Germany). Baking was performed with 18 biscuit doughs at 170 °C for 12 min. Each batch of dough produced 9 biscuits, thus requires two rounds of baking to produce the 18 biscuits. The amount of water in flour can be variable, which is measured via a drying oven method. The amount of added deionized water is adjusted to compensate for the water in flour. The nominal amount of water in flour is about 13%.

3.3. Sugar replacers

In Table 3 we have listed the used sugar replacers together with the known characteristic physical parameters, as derived in our previous papers (van Der Sman, 2013; Van der Sman, 2017; Van der Sman & Mauer, 2019; Renzetti et al., 2021, 2020). $T_{g,s}$ is the glass transition of the dry material, $N_{OH,s}$ is the effective number of hydroxyl groups per molecule (as computed in (van Der Sman, 2013) from $T_{g,s}$ values), $M_{w,s}$ is the molar weight, ρ_s is the mass density, χ is the Flory-Huggins interaction parameter, T_m is the melting temperature, and ΔH_m is the melting enthalpy at T_m. Oligofructose, mylose and polydextrose are assumed not to crystallize.

The oligofructose Frutalose OFP had an average degree of polymerization (DP) of 3.5, with properties listed in (Renzetti et al., 2021). The polydextrose Litesse Ultra, with average DP = 12, was assumed too large for sugar replacement. Hence, it was always mixed it with Mylose on a 50%/50% weight basis in all reformulations. For mylose, with an average DP of 3.0, we assumed that its properties are similar as maltotriose (Van der Sman & Mauer, 2019).

3.4. Reformulations

In Tables 4 and 5 we have listed the investigated reformulations. The numbers indicate the mass percentages used in the total recipe. The standard AACC recipe (REF) has 18% of sucrose and 15% of added water, as shown in Table 2. In the reformulations we have replaced part (about half) of the sucrose with the sugar replacers listed in Table 3. In some formulations, as indicated in Table 5, we have changed the ratio between all sugars (replacers) and water. We note that water acts as a plasticizer, and hence via varying added water we can change $\varphi_{w,eff}$ without modifying $\chi_{w,eff}$. The bottom row of Tables 4 and 5 indicates the symbols we have used in the figures to reference the individual formulations, following the nomenclature of Matlab or Python Matplotlib plotting routines.

With regard to reformulation we note that the applicability of some sugar replacers (like ERY) can be limited by their solubility (see Supplementary Material). But, sucrose was already practically dissolved after mixing of the dough. Moreover, at the elevated temperatures during baking it holds for all sugars and replacers that the used concentrations were below the solubility limits, as can be computed via the Flory-Huggins theory (Van der Sman, 2017).

In Tables 4 and 5 we have given the computed $\varphi_{w,eff}$ and χ_{eff}. The range of their values, covered by all reformulations, is displayed in Fig. 1. As one can observe these values are quite distributed around the value of the reference, but with several formulations close to the reference (indicated with yellow symbols, MALT, XYL/FOS and ERY/PDX). The symbols used in Fig. 1, representing the different reformulations, will be used again in the plots of the regression analysis (as presented in the Supplementary Material).

Table 2
Reference recipe (AACC International Method 10–53.01).

Ingredient	Weigth (g)
Sucrose	94.5
Skimmed milk powder	2.3
NaCl	2.8
Sodium bicarbonate	2.3
All-purpose shortening	90.0
High-fructose corn syrup (HFCS) 42%	3.4
Ammonium bicarbonate	1.10
Deionized water; g = (225 – g flour) + 49.5	Variable
Flour (13%)	225

Table 3
List of used sugar replacers together with known physical properties.

Polyhydroxy	$T_{g,s}$ (K)	$N_{OH,s}$	$M_{w,s}$	ρ_s	χ	T_m	ΔH_m
	(K)	[]	g/mol	kg/m^3	[]	(K)	kJ/mol
sucrose (SUC)	336	4.48	342	1550	0.50	463	56.0
erythritol (ERY)	228	2.75	122	1450	0.27	394	42.4
maltitol (MALT)	320	4.34	344	1550	0.51	425	52.9
xylitol (XYL)	249	2.97	152	1520	0.32	367	38.6
Mylose (MDX)	389	7.81	504	1550	0.60	–	–
Oligofructose (FOS)	328	5.16	626	1550	0.65	–	–
polydextrose (PDX)	353	5.51	2160	1550	0.80	–	–

3.5. Rheological measurements

Rheological measurements of dough were carried out using a Physica MCR 301 rheometer (Anton Paar GmbH, Stuttgart, Germany) in oscillation mode. The rheometer was equipped with parallel plate geometry (25 mm diameter) with sanded surface probe to prevent slippage. The temperature, set at 21^oC, was regulated by a circulating water bath and Peltier heating system. Before analysis, a 2.5–3 g of dough was rolled into a ball by hand and placed between the plates. After loading, the excess dough was trimmed and the sample's perimeter was covered with a thin layer of paraffin oil to prevent dehydration. Next, the sample was left to rest for 5 min.

Following this preparation procedure, amplitude and frequency sweeps were performed in duplicate for each sample. Amplitude sweeps were performed in the strain range 0.001–10 at a constant frequency of 1 Hz. Within the linear viscoelastic regime of the wet dough we have determined G' (Glve), and the tan(δ) (tdlve).

The thermo-mechanical behaviour of the different dough was determined by dynamical mechanical thermal analysis (DMTA). A DHR2 rheometer (TA Instruments, Crawley, UK), equipped with the Environmental Test Chamber (ETC) and serrated parallel plates (25 mm) were used for this purpose. Temperature sweeps at a constant deformation within the linear viscoelastic region (0.005) and at a constant frequency (1 Hz) were carried out by increasing the temperature from 21

Table 4
Composition (% w/w) and physical characteristics of reformulations with constant water. Reformulations are indicated with acronym with capital letters. Asterisks indicate formulations selected for sensory analysis.

compound (%)	REF*	ERY	XYL*	MALT*	FOS	PDX*	ERY/PDX*	XYL/FOS	MALT/FOS*	ERY/MALT*	XYL/MALT	MALT/PDX	FOS/PDX*
Sucrose	18	9	9	9	9	9	9	9	9	9	9	9	9
Water	15	15	15	15	15	15	15	15	15	15	15	15	15
Erythritol		9					4.5			4.5			
Xylitol			9					4.5			4.5		
Maltitol				9					4.5	4.5	4.5	4.5	
Oligofructose					9			4.5	4.5				4.5
PDX/MDX						9	4.5					4.5	4.5
χ_{eff}	.500	.381	.409	.500	.575	.593	.470	.492	.538	.440	.454	.540	.583
$\varphi_{w,eff}$	.281	.290	.288	.281	.274	.277	.284	.281	.278	.286	.284	.279	.276
Symbol	ro	mo	bo	yo	co	bs	ys	y*	gX	cs	gs	g*	c*

Table 5
Composition (% w/w) and physical characteristics of reformulations with varying water. Reformulations are indicated with acronym with capital letters. Asterisks indicate formulations selected for sensory analysis.

compound (%)	XYL-1	MALT-1	FOS-1 *	XYL+1 *	MALT+1 *	PDX+1 *
Sucrose	9	9	8	9	8	9
Water	14	14	14	16	16	16
Xylitol	10			8		
Maltitol		10			9	
Oligofructose			11			
PDX/MDX						8
χ_{eff}	.404	.500	.587	.414	.500	.586
$\varphi_{w,eff}$	0.278	0.270	0.262	0.298	0.292	0.288
Symbol	b*	gd	bd	bv	gv	bX

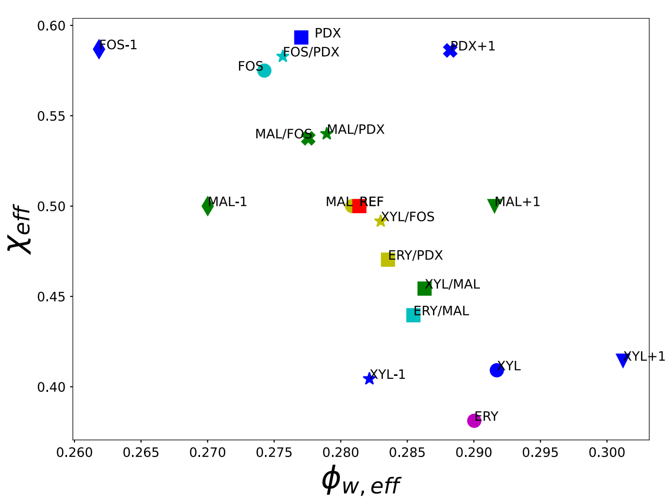

Fig. 1. The values of $\varphi_{w,eff}$ and χ_{eff} of the various tested reformulations. The colors of the symbols indicate the distance from the reference biscuit (REF) with only sucrose. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

to 120°C at a constant rate of 5 °C/min. Before analysis, a 2.5–3 g of dough was rolled into a ball by hand and placed between the plates. From the analysis of DMTA data, the setting temperature *TonG* of biscuit structure setting was determined as the inflection point following an increase in G'. The G' and the $\tan(\delta)$ at setting temperature were also recorded, denoted as *Gonst* and *tdonst* respectively. We note that the DMTA measurements are intended to investigate how the amount of plasticizer (as expressed in terms of $\varphi_{w,eff}$) will affect rheology. It is not intended to simulate baking conditions.

However, knowledge over what is happening during baking can be obtained via plotting *TonG* as function of $\varphi_{w,eff}$ in the supplemented state diagram of biscuit baking, which can indicate the relevance of the phase transitions of biopolymers during baking. The latter requires also data on how the state of the dough (in terms of temperature and moisture content) changes during baking, as a consequence of the heating and evaporation.

3.6. Instrumental evaluation biscuit quality

Biscuit dimensions were determined as the height (bheight), diameter in the rolling direction (blength) and the diameter in the direction perpendicular to the rolling direction (bwidth).

The moisture content (bmc) of the different biscuits was determined according to AACC method (44–15.02, 2001). Their water activity (baw) was determined using a Decagon Aqua Lab meter (Pullman, WA, USA) calibrated with a standard lithium chloride solution ($a_w = 0.5$).

Textural properties were determined using a three point bending probe and support (A/3 PB). The experimental conditions were: test speed 1 mm/s; distance between supports of 20 mm; probe travel distance 6 mm. The biscuits were fractured with the bottom side upwards and the biscuit length parallel to the supports. The analysis was performed in 8-fold for each biscuit type. With use of the calculated area of the biscuit, the fracture stress (Stress) is computed from the force at fracture.

3.7. Sensory evaluation of biscuits

The sensory properties of the biscuits were evaluated by a trained test panel from Mondelēz using the Descriptive Sensory Profiling (DSP) method. The sensory panel consisted of 10 trained, experienced subjects selected based on their ability to recognize and identify odors and basic tastes, verbal creativity and texture sensitivity. The panel is established at Mondelēz and regularly trained for sensory evaluation of biscuits based on a list of relevant scored attributes as given in Table 7. The trained panel has self defined the meaning of the attributes in previous training sessions, while evaluating regular Mondelēz biscuits and the AACC 10–53 standard. With these definitions the panel can perform comparisons of different biscuits regarding, aroma, appearance, tactile texture, and mouthfeel. Before the final scoring of reformulated biscuits, training sessions were performed prior to measurements to familiarize with samples and (re)confirm the list of attributes.

Subjects rated all attributes of each sample on a non-structured 100 point line scale going from "very weak/very little" to "very strong/very much". Panellists assessed all samples in triplicates over three different sessions. Samples were assessed in randomized order. The assessments took place in a temperature-controlled room under white light and in standard sensory booths, designed in accordance with ISO 8589 (ISO, 2007). The subjects were instructed to rinse their mouths with distilled water between samples to avoid the carry-over effect.

Table 6
Linear regression results on physical parameters.

Symbol	Description	r^2	b_φ	b_χ	p_φ	p_χ	p_{tot}
Dough parameters							
Glve	G' wet dough	0.40	−0.60	−0.40	0.015	0.010	0.024
tdlve	tan(δ) wet dough	0.71	0.16	0.84	0.919	0.000	0.000
TonG	Setting temperature	0.63	−0.70	0.30	0.003	0.418	0.000
Gonst	G' setting dough	0.53	−0.36	−0.64	0.087	0.001	0.002
tdonst	tan(δ) setting dough	0.61	−0.50	0.50	0.075	0.046	0.000
Biscuit parameters							
baw	water activity	0.89	0.44	−0.56	0.000	0.000	0.000
bmc	moisture content	0.92	0.48	−0.52	0.000	0.000	0.000
bheigth	width	0.88	−0.40	0.60	0.006	0.000	0.000
blength	length	0.87	−0.44	0.56	0.006	0.000	0.000
bwidth	height	0.83	0.30	−0.70	0.010	0.000	0.000
Stress	Fracture stress	0.80	−0.50	0.50	0.007	0.004	0.000

A familiarisation training session also took place prior to the data collection. The biscuits were also tested on a wider variety of aromas, appearance, and tactile texture. However, these attributes showed very low correlation with χ_{eff} or $\varphi_{w,eff}$, and thus they are not listed. The number of tested formulations for the trained panel was limited to 12. The reformulations involved in the DSP test are indicated in Tables 4 and 5 via an asterisk behind the acronym. In our selection we have tried to have formulations having values of χ_{eff} or $\varphi_{w,eff}$, distributed as uniformly as possible over the available range, while still keeping several formulations that are close to the reference.

3.8. Linear regression analysis

We have performed regression analysis using a multivariate linear model:

$$\hat{y} = b_0 + b_1\tilde{\varphi}_{w,eff} + b_2\tilde{\chi}_{eff} \tag{4}$$

$\hat{y}$ is the estimated values of the physical or sensorial attribute y of the dough or biscuit. $\tilde{\varphi}_{w,eff}$ and $\tilde{\chi}_{eff}$ are reduced variables defined as:

$$\begin{aligned} \tilde{\varphi}_{w,eff} &= \frac{\varphi_{w,eff} - \varphi^{min}_{w,eff}}{\varphi^{max}_{w,eff} - \varphi^{min}_{w,eff}} \\ \tilde{\chi}_{eff} &= \frac{\chi_{eff} - \chi^{min}_{eff}}{\chi^{max}_{eff} - \chi^{min}_{eff}} \end{aligned} \tag{5}$$

The superscripts ^{min} and ^{max} indicate the minimal and maximal values of the variables in the set of biscuit reformulations. To distinguish the different contributions of the two characteristic numbers we compute the following coefficients:

$$\begin{aligned} b_\varphi &= \frac{b_1}{\|b_1\| + \|b_2\|} \\ b_\chi &= \frac{b_2}{\|b_1\| + \|b_2\|} \end{aligned} \tag{6}$$

Furthermore, we have computed the regression coefficient r^2 of the multivariate linear regression.

The results of the regression analysis will be summarized in polar plots, which are in a way similar to PCA plots. For each of the analysed attributes we will draw a line from the centre, with the length equal to the regression coefficient r^2, and the angle determined by the ratio b_φ/b_χ. Or equivalently, the endpoint of the line is determined by the co-ordinates $b_\varphi r^2$ and $b_\chi r^2$. The lines will indicate the strength of the correlation of the attribute with $\varphi_{w,eff}$ and χ_{eff}. The sign of b_φ or b_χ will indicate whether the correlation is either positive or negative with $\varphi_{w,eff}$ or χ_{eff}. The angle of the line with the horizontal axis indicates the relative contribution of either $\varphi_{w,eff}$ or χ_{eff} to the linear correlation.

4. Results and discussion

The central hypothesis of our study was that dough rheology, physical properties of biscuits and their texture-related sensory attributes are controlled by $\varphi_{w,eff}$ and χ_{eff}. As discussed in the methods section, we tested this hypothesis via linear regression analysis of physical and sensorial properties with $\varphi_{w,eff}$ and χ_{eff}, which we summarized in polar plots (Figs. 2 and 3). The numerical values of correlation coefficients, b_φ, b_χ, and the p-values are listed in Tables 6 and 7

In the Supplementary Material we have shown a multitude of parity plots, comparing the measured values of the physical or sensorial attributes with the estimated values according to the linear regression. The colors of the used symbols indicate the "distance" of the reformulation compared to the reference biscuit (in terms of $\varphi_{w,eff}$ and χ_{eff}). With these parity plots the validity of the sugar replacement strategy will be tested.

The results of the regression analysis are shown in the polar plots of Figs. 2 and 3. In the polar plots the length of each measured parameter indicate the strength of the correlation (the r^2 coefficient) while the angle indicate the relative contribution of $\varphi_{w,eff}$ and χ_{eff} based on the projection of the parameter's vector on the x-y axis. From Figs. 2 and 3 we observe that indeed many attributes relevant for the dough properties and the texture of the biscuit are well correlated with linear combinations of $\varphi_{w,eff}$ and χ_{eff}. Almost all physical attributes are largely

Table 7
Linear regression results on sensorial parameters.

Symbol	Description	r^2	b_φ	b_χ	p_φ	p_χ	p_{tot}
Aroma							
	Stale Aroma	0.59	−0.32	−0.68	0.537	0.025	0.019
Appearance							
	Convexity Surface	0.56	0.41	−0.59	0.140	0.338	0.011
Tactile texture							
	Oily Touch	0.70	−0.16	−0.84	0.833	0.016	0.005
Mouthfeel							
MF_HRD	Initial Bite Hardness	0.84	−0.05	0.95	0.984	0.002	0.000
MF_VOL	Volume of Sound	0.89	−0.04	0.96	0.956	0.000	0.000
MF_CMB	Crumbliness	0.10	–	–	–	–	–
MF_CHW	Effort to Chew	0.86	−0.20	0.80	0.501	0.002	0.000
MF_AIR	Airiness	0.70	0.29	−0.71	0.559	0.061	0.007
MF_DRY	Dryness	0.37	–	–	–	–	–
MF_PST	Pastiness of Mass	0.63	−0.16	−0.84	0.546	0.014	0.009
MF_STK	Stickiness to Teeth	0.44	–	–	–	–	–
MF_DSR	Dissolution Rate	0.83	0.11	−0.89	0.877	0.003	0.000
MF_GRN	Graininess	0.57	0.28	0.72	0.781	0.036	0.018
MF_MCT	Mouth Coating	0.03	–	–	–	–	–

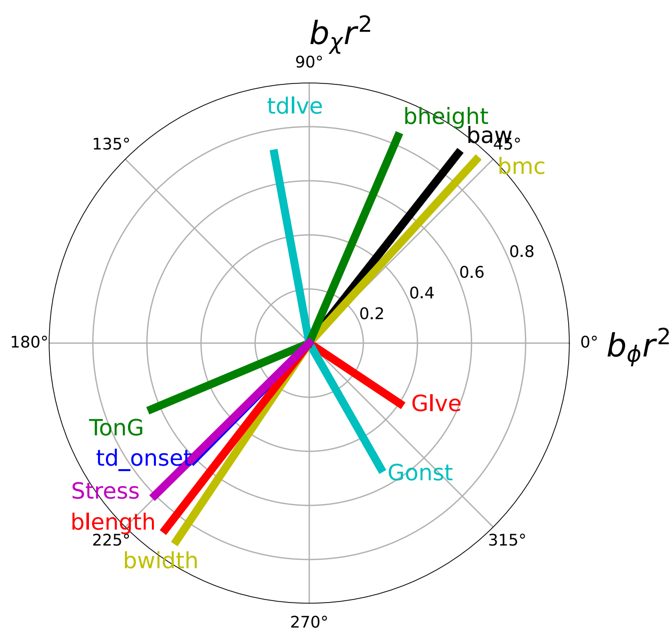

Fig. 2. Polar plot summarizing the results of the regression analysis for the physical attributes of dough and biscuits. The used abbreviations of the physical attributes are: Glve = G' wet dough, tdlve = tan(δ) wet dough, TonG = Setting temperature, Gonst = G' setting dough, tdonst = tan(δ) setting dough, baw = Biscuit water activity, bmc = Biscuit moisture content, bheigth = Biscuit width, blength = Biscuit length, bwidth = Biscuit height, Stress = Fracture stress. Table 6 lists the numerical results of the regression analysis.

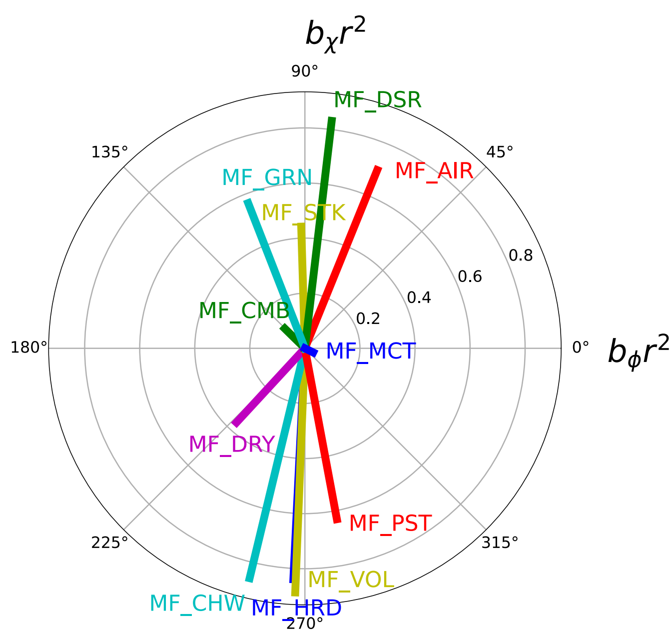

Fig. 3. Polar plot summarizing the results of the regression analysis for the sensorial attributes of the biscuits, related to mouthfeel. The used abbreviations of the attributes are: MF_HRD = Initial Bite Hardness, MF_VOL = Volume of Sound, MF_CMB = Crumbliness, MF_CHW = Effort to Chew, MF_AIR = Airiness, MF_DRY = Dryness, MF_PST = Pastiness of Mass, MF_STK = Stickiness to Teeth, MF_DSR = Dissolution Rate, MF_GRN = Graininess, MF_MCT = Mouth Coating. Table 7 lists the numerical results of the regression analysis.

dominated by both $\varphi_{w,eff}$ and χ_{eff}, albeit that parameters related to dough rheology are more dominated by χ_{eff}. The sensorial attributes are more dominated by χ_{eff} than by $\varphi_{w,eff}$.

These findings are for a great deal in agreement with the hypotheses stated in Table 1 regarding the influence of $\varphi_{w,eff}$ and χ_{eff} on sugar functionality. The relative dominance of χ_{eff} on parameters related to dough rheology expresses the importance of the partial gluten hydration for the dough rheology. In our recent review we have discussed in more detail the importance of limited gluten hydration for limited network formation (van der Sman & Renzetti, 2019).

The other parameters, related to physical parameters after baking, are determined by the moment of thermosetting of the dough, the viscoelastic properties of the setting dough and the moisture sorption properties. Hence, both $\varphi_{w,eff}$ and χ_{eff} are expected to correlate with these parameters.

The importance of gluten thermosetting is also evident from the supplementary state diagram of biscuits, as presented in Fig. 4. As it can be assumed that water is equally partitioned between gluten and starch, based on their comparable sorption isotherms (Roman-Gutierrez et al., 2002; Van der Sman, 2017; Wang et al., 2004), phase transitions for both gluten and starch can be drawn in the same state diagram. The phase transitions are predictions following the Flory-Huggins theory (Cornet et al., 2020; Van der Sman & Mauer, 2019; Van der Sman & Meinders, 2011; van der Sman & Renzetti, 2019), but with glass transition data for gluten from (Bier et al., 2014; Cornet et al., 2020). The thermosetting temperature of gluten and gliadin is from (Toufeili et al., 2002; Kokini et al., 1994; Madeka & Kokini, 1994, 1996; Icoz & Kokini, 2008) (where we have also included data on thermosetting of zein, assuming similar behaviour as gluten). In the state diagram we have included the change of the state of the biscuit during baking, as measured by (Chevallier et al., 2002) and (Fahloul et al., 1994), which is indicated as the processing pathway. The supplemented state diagram is given as function of $\varphi_{w,eff}$, as both glass transition and biopolymer melting are governed by this parameter (van der Sman, 2016).

We have included our *TonG* data in the supplementary state diagram, indicating that the setting of the dough is governed by gluten thermosetting. During biscuit baking starch will not gelatinize (Pareyt et al., 2009), which is also indicated by the processing path, which remains below the onset of starch gelatinization. Hence, we can conclude that the texture formation during baking is strongly governed by the thermosetting of gluten. This finding is also in agreement with (Pareyt et al., 2010), showing that redox agents modulate gluten aggregation during biscuit baking, which in turn control biscuit dimensions and internal structure.

The DSP panel results show that the mouthfeel parameters are largely determined by χ_{eff}, indicating that for sensorial properties the

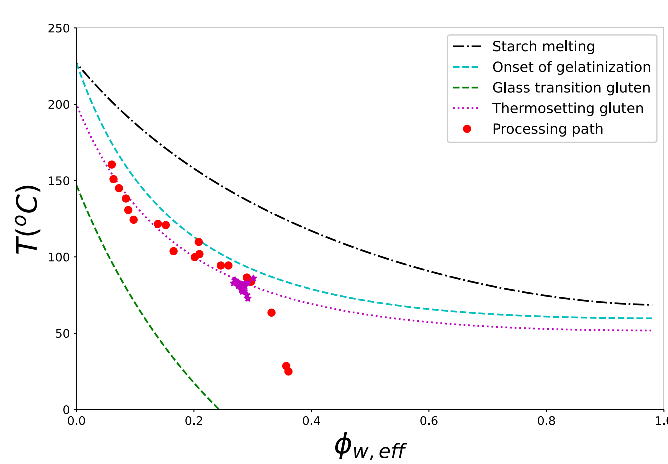

Fig. 4. Supplemented state diagram for biscuits, indicating starch melting and onset of gelatinization, gluten thermosetting, and glass transition of gluten. Furthermore, the processing pathway indicates how the state of the biscuit changes during baking. Data for the phase transitions, and processing pathway is from literature, as discussed in the text. The magenta symbols (stars) indicate the values of *TonG*, as measured in this study.

dough rheology, and thus gluten network development is rather important. A stronger gluten network (due to a lower χ_{eff}) implies a stronger hardness, sound emission, effort to chew, and less air pore developments, and less crumbly texture. This is in line with the (signs) of the correlations shown in Fig. 3.

For the sensorial attributes, other than mouthfeel, Table 7 indicates that $\varphi_{w,eff}$ and χ_{eff} only contribute to biscuit appearance in terms of convexity of surface, while they contribute little to flavour and aroma. Many flavours and aromas are scored, as normally done by Mondelez panel, but only a slight correlation with stale aroma has been observed. As shown explicitly in Fig. 7 we find little correlation between $\varphi_{w,eff}$ and χ_{eff} and sweetness (regression analysis shows $r^2 = 0.11$), though some formulations scored reasonably close to the reference.

Hence, one can steer the texture of biscuits with $\varphi_{w,eff}$ and χ_{eff} quite independently of their taste. But, insufficiencies in sweetness perception must be compensated for with non-plasticizing compounds like high-intensity sweeteners or with naturally occurring sweetness enhancing aromas, such as vanilla (Bertelsen et al., 2021; Romeo-Arroyo et al., 2022). A combination of these two strategies may be most effective, without altering texture. Alternatively, one could combine our strategy with others like multi-modal effects or inhomogeneous distribution of sweeteners (Di Monaco et al., 2018), or food industry can opt for gradual decrease of sweetness in their product, such that consumers accustom slowly to it (Deliza et al., 2021; Hutchings et al., 2019).

The above findings confirm the hypothesis that texture of biscuits is controlled by both characteristic numbers, $\varphi_{w,eff}$ and χ_{eff}. From the parity plots, figures A.5 and A.6 one can observe that reformulations with values of $\varphi_{w,eff}$ and χ_{eff} close to that of the reference (as indicated by yellow symbols) indeed perform very similar to the reference (which is indicated by the red symbol).

Our results show that there can be multiple solutions to the sugar replacement problem in biscuit, leaving room for optimization of other quality aspects of the biscuit. For, example our research shows that sucrose can be completely replaced by maltitol, but it is often not preferred by food industry because of its strong laxative properties (Ruskone--Fourmestraux et al., 2003). Yet, our study shows that mixtures of sugar replacers can perform similar to maltitol: e.g. a mixture of a large oligosaccharide (like a fructooligosaccharide or polydextrose) and a small polyol like xylitol or erythritol (Jurgens et al., 2016, p. 286). Especially, erythritol has a good gastrointestinal tolerance (Storey et al., 2007), but its solubility is quite limited compared to other polyols (Van der Sman, 2017) (see also Supplementary Material). Problems with solubility of erythritol can be resolved via mixing it with other small polyols. The use of complex mixtures of sugar replacers has the advantage of 1) fine tuning of $\varphi_{w,eff}$ and χ_{eff}, 2) control of possible problems regarding solubility or laxative properties, and 3) optimization of the food properties like sweetness, or dietary fiber content (via maximizing the content of fructooligosaccharides).

Our evaluation of the physical and sensorial properties of the biscuits shows the validity of our sugar replacement strategy. We think that this strategy is valid for many other food categories like other sweet bakery products, confectionery products, deserts or even beverages. For cakes we have shown that the sugar functionality is very similar to those in biscuits (Van der Sman & Renzetti, 2020): the plasticizing and hygroscopic properties control the rheology, phase transitions of biopolymers, and moisture distribution (Van der Sman & Mauer, 2019; Renzetti et al., 2020; 2021). In contrast to biscuits, the swelling of starch is also crucial for the cake texture, which appears to be controlled via $N_{OH,s}/\nu_s$ and thus not directly via $\varphi_{w,eff}$ or χ_{eff} (Renzetti et al., 2021). Earlier, we have observed similar effects of sugars on swelling of dextran microgels, which are viewed as model systems of gelatinizing (crosslinked) starch granules (Van Der Sman, 2018). But, this third characteristic number can also be computed with the same theory. In fact, we have recently demonstrated that by controlling $\varphi_{w,eff}$ or χ_{eff} and $N_{OH,s}/\nu_s$ one can steer the cake volume, the crumb texture (e.g. hardness, cohesiveness) and water activity (Renzetti & van der Sman, 2022).

Sugars have similar functionality as plasticizer and humectant in confectionery products like fruit gums, where biopolymers like gelatin or starch determine the texture (Hartel et al., 2018). In some confectionery products sugar functionality also depends on the crystallinity of sucrose. Hence, their sugar replacement can be challenging as the solubility of sugar replacers does not show universal behaviour (Van der Sman, 2017). In beverages sugars provide sweetness and thickness via their viscosity enhancing properties. The sugar replacement strategy can still be applied, as the viscosity is controlled via $\varphi_{w,eff}$, or equivalently T_g/T (Van der Sman & Mauer, 2019). Surprisingly, we have also observed this scaling of rheology with T_g/T for concentrated biopolymers like maltodextrins, starch, and plant proteins (Siemons et al., 2022; Van der Sman et al., 2022; van der Sman et al., 2022).

5. Conclusion

Based on our results we conclude that the sugar functionality and biscuit texture is indeed largely determined by the two characteristic numbers: $\varphi_{w,eff}$ or χ_{eff}. Furthermore, reformulated biscuits with similar values of $\varphi_{w,eff}$ or χ_{eff} as the reference biscuit, have physical and sensorial textural properties that are most close to that of the reference. Hence, both findings validate our sugar replacement strategy. Furthermore, we have shown that multiple solutions exist for the sugar replacement problem, allowing room for optimization of the biscuit for other quality or nutritional traits.

We pose that the sugar replacement strategy has a universal character, and that it can apply to other food categories like sweet bakery products, confectionery and beverages. In a companion paper (Renzetti & van der Sman, 2022), we have shown that for the case of cakes $\varphi_{w,eff}$ or χ_{eff} are also important contribution to physical measures of the cake texture. However, a third parameter $N_{OH,s}/\nu_s$, also accounts for enhanced swelling of starch, and thereby also for the cohesiveness of the cake. Yet, this third parameter is derived from the same theories, we have based the sugar replacement strategy for biscuits.

Author statement

RGM van der Sman – Conceptualization; Methodology/Theory; Writing - review &editing. A Jurgens – (Physical) Data curation. A. Smith – (Sensorial) Data curation. S. Renzetti – Supervision experimental work; Experimental methodology; Review.

Declaration of competing interest

Authors declare there is no conflict of interest, otherwise that one of the co-authors is employed by Mondelez, who co-funded the research. That fact did not influence the outcomes of this research.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodhyd.2022.107966.

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