European Food Research and Technology (2020) 246:2249–2257
https://doi.org/10.1007/s00217-020-03569-x

ORIGINAL PAPER

Structured fat–water–fiber systems as fat substitutes in shortbread formulation: modulation of dough characteristics following a multiscale approach

Fatma Boukid[1] · Agoura Diantom[2] · Roberto Corte[1] · Elena Curti[1] · Eleonora Carini[1] · Elena Vittadini[3]

Received: 19 December 2019 / Revised: 3 July 2020 / Accepted: 4 July 2020 / Published online: 26 July 2020

Abstract

The use of "fat substitutes" is trending upward in food industry to meet dietary recommendations and to respond to the increasing number of health-conscious consumers. In this frame, a multiscale approach was applied to study the effect of structured fat–water–fiber systems on shortbread dough quality. Several formulations ($n=9$) were designed based on three structured fat–water–fiber systems at different fat levels (15%, 20%, and 23%), three conventional fats (butter-B, palm oil-PO, and sunflower oil-SO), and three combinations of conventional fats with structural emulsions (at 20% fat and 20% water contents). The partial substitution of B and PO by the structured emulsions resulted in moister, less sticky, and softer doughs. Structured emulsion-based doughs exhibited higher structural stability (higher tan δ) than those made with conventional fats. ^{1}H nuclear magnetic resonance (NMR) revealed significant differences in water dynamics, where doughs containing structured fat–water–fiber systems (richer in unsaturated fats) were characterized by a more rigid population, while those rich in saturated ones (B and PO) had more mobile protons. Overall, this multilevel screening emphasized the usefulness of ^{1}H nuclear magnetic resonance in monitoring the molecular differences among the different formulation (which were less evident at mesoscopic and macroscopic levels), as confirmed by multivariate statistics.

Keywords Structured emulsions · Dough texture · Dynamic rheology · NMR · Multivariate statistics

Introduction

Shortbread is an important bakery items, which contain high fat amounts (20–50 g/100 g of flour) depending on the formulation. In the last decades, the overconsumption of fat has been associated with several chronic diseases, such as obesity, high blood cholesterol, coronary heart diseases, and diabetes [1]. People are becoming more attentive toward the quality and the quantity of consumed fats and particularly saturated fatty acids [2]. The daily intake of total fat content should not exceed 30% and 10% from saturated fats, and 20% from monounsaturated and polyunsaturated of the total energy intake [3]. Therefore, the food industry is focusing on the development of low-fat/low-calorie, high-fiber foods in response to public interest for these functional products [4].

In shortbread, reduction and/or substitution of fat without hampering the technological properties is a challenging task for food scientists, since fat plays a crucial role in rheological properties of dough and affects the final product quality. During dough mixing, fat interacts with flour particles to form a sort of coating that favors dough handling by acting as lubricant among flour constituents, limits protein and starch hydration, prevents gluten development, and results in a dough with desirable structure with no shrinking after lamination [5, 6]. Subsequently, the baked product will have a tender and crumbly texture, also called "short" texture [5–7]. Furthermore, fat allows the correct incorporation of air during dough formation and determines dough resistance to baking temperatures [7, 8]. Consequently, texture, mouth feel, lubricity, flavor, and overall perception of the

✉ Elena Vittadini
elena.vittadini@unicam.it

[1] Department of Food and Drug, University of Parma, Parco Area delle Scienze 47/a, 43124 Parma, Italy

[2] Present Address: Ecole Supérieure des Techniques Biologiques et Alimentaires, University of Lomé, BP 1515, Lomé, Togo

[3] Present Address: School of Biosciences and Veterinary Medicine, University of Camerino, Via Gentile III da Varano, 62032 Camerino, MC, Italy

final product are largely dependent on the fat component of the formulation [5, 6, 9].

Solid fats (at ambient temperature) are commonly used in the bakery industry, since they play a key role in the quality features of baked goods, providing a unique mechanical and sensory attributes to dough and final product [9–11]. These materials are mainly composed by triacylglycerols (TAG) with high content of saturated fatty acids (SFA) or transfatty acids (TFA), which are associated with negative effects on health [12]. Therefore, the use of vegetable oils to replace solid fats in biscuits is an alternative to make products with a healthier fatty acid profile [13]. However, doughs containing oils are softer and result in harder texture shortbread cookies [14, 15].

A successful design of high-quality fat-reduced products requires appropriate strategies to replace fat role or some of its attributes to avoid the undesirable technological effects in shortbread, including stiffening/softening of the dough, increasing in cohesive, adhesive and elastic dough properties, shrinkage of the final product, loss of color, and textural and sensory properties [16]. For these reasons, fat replacement in shortbread has been intensively studied, where a spectrum of substances were assayed, such as corn fiber, maltodextrin and lupine [17], inulin [18–20], polydextrose [2], resistant starch and polydextrose [6], cellulose [5], and inulin and hydroxypropyl methylcellulose [21]. Recently, a structured emulsion (50:50 water:oil biphasic systems) created by the action of a commercial fiber was proven efficient as a butter substitute in shortbread cookies [9]. In light of these previous knowledge, this research gives a wider overview about the usefulness of this structured emulsion on the product quality by thoroughly investigating dough properties in substitution of various fats commonly used in shortbread formulations. To this aim, a multilevel investigation on the effect of structured emulsions as "fat substitutes" on the quality characteristics of shortbread cookie dough, as compared to fats classically used in shortbread formulations (butter and palm oil), is carried out. The experimental design aimed at (i) comparing physico-chemical properties of dough made with single fats/oils (saturated: palm oil and butter; unsaturated: sunflower oil), structured emulsions, and fat/oil combinations, at constant water and fat contents, and (ii) evaluating the effects of the replacement of fat with structured emulsions at different levels (15%, 20%, and 23%). The properties of dough of these formulations were evaluated at macroscopic (moisture content, water activity, and texture), mesoscopic (dynamic rheology), and molecular (Proton Time Domain Nuclear Magnetic Resonance, TD-NMR) levels.

Materials and methods

Ingredients

Soft wheat flour [ashes ≤ 0.55% (dry basis, d.b.), proteins ≥ 9% (d.b.), moisture ≤ 14.5% (wet basis, w.b.); *W*: 134 10^{-4} J; 0.49 P/L] were kindly provided by a local producer (Molino Agugiaro & Figna, Collecchio, PR, Italy). Sugar, fat, leavening agent (Lievito Bertolini, sodium bicarbonate, and disodium diphosphate), and egg yolk were obtained from a local supermarket.

Structured emulsions (SE) were produced by mixing commercial fiber (HI-FIBREWF, HI-FOOD, Parma, Italy) with sunflower oil (From local supermarket) in a bowl chopper (Polyfunctional Qbo 8-3, Roboqbo, Bologna, Italy) at 25 °C for 1 min at 1000 Hz, then adding water and mixing for 4 min at 2000 Hz to obtain a uniform and shiny mass. The obtained structured emulsions were stored at refrigerated temperature until use.

Dough formulation

As shown in Table 1, dough shortbread formulations were developed using SE at different levels (SE1: 15%; SE2: 20%; SE3: 23%) to substitute fat. Shortbread formulations with combinations of SE and traditional fats were also designed to have equal amounts of both fat (20 g fat/100 g dough)

Table 1 Short bread formulations expressed in g

	SE1	SE2	SE3	B	PO	SO	SE2 + B	SE2 + PO	SE2 + SO
Flour	100	100	100	100	100	100	100	100	100
Sugar	40	40	40	40	40	40	40	40	40
Eggs	20	20	20	20	20	20	20	20	20
Leavening agent	3.2	3.2	3.2	3.2	3.2	3.2	3.2	3.2	3.2
Water	15	20	23	20	20	20	20	20	20
Structured emulsion	32	42	48	–	–	–	21	21	21
Butter	–	–	–	20	–	–	10	–	–
Palm oil	–	–	–	–	20	–	–	10	–
Sunflower oil	–	–	–	–	–	20	–	–	10
Lecithin	–	–	–	–	–	0.3	–	–	–

and water (21 g water/100 g dough). Fat phase included SE (SE2), anhydrous butter (B), anhydrous butter palm oil (PO), sunflower oil (SO), and combination of SE and other sources of fats [butter (SE2 + B) and palm oil (SE2 + PO) and sunflower oil (SE2 + SO)] (Table 1).

For dough preparation, fat phase and sugar were creamed (3 min, speed 3) at room temperature in a mixer (XBE10S, Electrolux, Senlis, France). Then, water and eggs were added with continuous mixing for 30 s at a speed of 3. Finally, flour was added and mixed for 2 min at a speed of 3 to obtain a dough with a proper structure (1.5 kg dough per each formulation). Two productions were made for each formulation.

The resulting doughs were easy to handle except for that made with 100% sunflower, where the dough was too soft and not workable. Therefore, lecithin was required to enable an interaction between oil and water, and to help maintaining a stable emulsion between these two unmixable liquids and allowing for shortbread production.

Dough characterization

Macroscopic analysis

Water activity was measured at 25 °C with an Aqualab 4 TE (Decagon Devices, Inc. WA, USA) [22]. For each production, triplicate analysis was carried out.

Moisture content (MC %) was measured by drying at 105 °C to constant weight with a forced air oven (ISCO NSV 9035, ISCO, Milan, Italy) [22]. For each production, triplicate analysis was carried out.

Texture analysis A single compression test was conducted using a TA.TXplus texture analyser (490 N load cell, Stable Micro Systems) with a 35 mm spherical probe (1″ Spherical Probe: P/1sp). Before the analysis, the dough was gently inserted into cylinders of 50 mm diameter flattened to an approximate height of 5 cm and wrapped with parafilm (later removed before analysis) to avoid drying of superficial layer of dough. Dough cylinders were allowed to rest and placed in a thermostat in a cell at 25 °C for 3 h prior to analysis. Dough cylinders were then compressed (test speed = 2 mm/sec; distance: 10 mm; trigger force = 0.049 N) and the maximum force at 20% strain was taken as hardness (*N*), while the area of the negative peak (*N/s*) as stickiness. For each production, five cylinders of dough were tested for a total of ten replicates of each formulation.

Mesoscopic analysis

Viscoelastic properties of the dough were measured using a rheometer ARES (TA Instruments, New Castle, USA) equipped with parallel plates (PRL–ARNS 8ATH; 50 mm diameter covered with 100 grit medium sand paper to avoid slippage effects) and commanded by an Orchestrator software (Rheometric Scientific Ltd, USA).

Once the gap is taken to test gap, sample was trimmed and vaseline oil was applied to the edges of the samples, which are not in contact with the plate surfaces. After sample loading, sample went through a resting time until axial force reached about 0 N prior to start experiment to allow for temperature equilibration and dough relaxation.

Strain sweep test (strain range 0.01–10%; frequency, 5 Hz) was conducted to identify the linear viscoelastic region. Frequency sweep test (frequency range 0.1–10 Hz; strain, 0.035%) was carried out to measure storage modulus (G′, MPa), loss modulus (G″, MPa), and tan δ (G″/G′). For each production, five replicates were carried out for a total of ten samples of each formulation. All values were maintained, and no discrimination was performed to avoid biased results. Rheological curves were obtained using Microsoft Office Excel Software (Microsoft Corporation, USA).

Molecular analysis: ^{1}H molecular mobility (time domain proton nuclear magnetic resonance, TD-NMR)

^{1}H molecular mobility was investigated with a low-resolution spectrometer (TD-NMR, 20 MHz, the miniSpec, Bruker Biospin, Milano, Italy). Dough samples were inserted in specimen tube until reaching 10 mm of height and sealed with Parafilm® to avoid evaporation of water from the sample during the NMR test. Two samples were analyzed for each formulation of each production. Free induction decay (FID), ^{1}H T_2, and ^{1}H T_1 relaxation times experiments were executed at 25.0 ± 0.1 °C.

^{1}H FIDs were acquired using a single 90° pulse, followed by a dwell time of 7 µs, a recycle delay of 0.5 s (> 5T_1), a 0.5 ms acquisition window, and 900 data points. The curves were fitted with a two components model (exponential and Gaussian; [23]; Sigmaplot, v6, Systat Software Inc., USA):

$$F(t) : y0 + A \times \exp\left(-t/T_A\right) + B \times \exp\left[-\left(t/T_B\right)^2\right], \quad (1)$$

where *y*0 is the FID decay offset, *A* and *B* are the intensities of each relaxation component, and T_A and T_B are the apparent relaxation times.

^{1}H T_1 (longitudinal relaxation times) were determined by the inversion recovery pulse sequence with a recycle delay of 0.5 s, and an interpulse ranging from 0.1 ms to 1000 ms. ^{1}H T_2 relaxation time was measured with a Carr–Purcell–Meiboom–Gill (CPMG) pulse sequence with a recycle delay of 0.5 s (≥ 5 ^{1}H T_1), and an interpulse spacing of 0.04 ms and 26,000 data points. ^{1}H T_2 curves were analyzed as quasi-continuous distributions of relaxation times using a UPENWin software (Alma Mater Studiorum, Bologna, Italy). Default values for all UPEN settings parameters were used with the exception of the LoXtrap parameter that was set to 1 to avoid

the extrapolation of relaxation times shorter than the first experimental point. ^{1}H T_2 CPMG relaxation decays were fitted with a discrete exponential model (Sigmaplot, v.6, Systat Software Inc., USA).

Statistical analysis

Analysis of variance (one-way ANOVA) was conducted to evaluate the effect of fat type/amount on each parameter at a significance level of $\alpha = 0.05$. Significant differences among the mean values were calculated using Duncan's test ($p \leq 0.05$). Correlation coefficients (r) were computed using Pearson's coefficient ($p \leq 0.05$). Principal component analysis (PCA) was conducted on average values due to unequal number of determinations among properties. PCA was performed based on correlation matrix. The relevant features were discriminated based on the load scores ($> \pm 0.6$). All experimental data were analyzed using the SPSS version 25.0 (SPSS Inc., Chicago, IL, USA).

Results and discussion

Macroscopic properties

Table 2 summarizes the macroscopic properties of shortbread dough. As expected, water content was higher in SE3 followed by SE2 and SE1. No relevant differences were observed among the other samples, which were designed to contain the same amounts of water. Water activity was the highest in SE3 (~ 0.899), followed by SE2 (~ 0.886) and SE1 (~ 0.863), due to the increasing water content associated with the larger amount of SE present in the formulation. B and PO showed similar water activity to SE2, but slightly lower than SO. For the combinations, no significant differences were found between SE2 + PO and SE2 + B, which were slightly lower than SE + SO.

Regarding dough texture, stickiness was significantly different among dough formulations. No trend related to fat amount in the SE was observed in SE1, SE2, and SE3, with SE2 having the lowest value. Considering the same water and fat contents, SO had the highest stickiness, and PO, B, and SE2 + SO showed intermediate values, while SE2 + B and SE2 + PO had the lowest stickiness. These results might suggest a possible synergy between oil–water–fiber systems and saturated fats (palm oil and butter) that resulted in a lower dough stickiness.

As for hardness, the increase of fat amount in emulsions-based formulations (SE1, SE2, and SE3) resulted in softer dough due to the shortening effect of lipids and, likely, the higher amount of water (SE1 < SE2 < SE3) [9]. When compared to the control (B), SE2 had similar hardness, emphasizing that structured emulsion was able to generate a proper dough structure, with a comparable performance to butter. Noteworthy, SO were softer than B and PO probably due to the presence of unsaturated fats (liquid at room temperature), which modulated the structure of the dough [24].

Mesoscopic properties

The viscoelastic properties of a dough are crucial features for its machinability and shortbread quality [5, 20, 21]. Representative curves (versus frequency) obtained for different shortbread doughs in terms of elastic (G′, Fig. 1a) and viscous (G″, Fig. 1b) moduli are presented in Fig. 1. G′ was significantly higher than G" at all frequencies, indicating a predominant elastic behavior of shortbread doughs. This behavior was confirmed in all samples, suggesting that the use of structured emulsions as fat substitutes did not affect the prevalence of elastic component versus viscous component.

To highlight the effect of fat replacement on dough viscoelastic properties, Table 3 shows elastic (G′) and viscous (G″) moduli at the same frequency (1 Hz). These data revealed that dough viscoelastic properties were significantly

Table 2 Macroscopic properties of shortbread dough

Formulation	Moisture content (%)	Water activity (dimensionless)	Hardness (*N*)	Stickiness (*N/s*)
SE1	21.71 ± 0.08d	0.863 ± 0.004e	5.52 ± 0.35a	0.604 ± 0.049c
SE2	22.96 ± 0.07c	0.886 ± 0.001cd	4.09 ± 0.22cd	0.498 ± 0.065bc
SE3	24.13 ± 0.15a	0.899 ± 0.001a	2.67 ± 0.11e	0.624 ± 0.075cd
SE2 + B	23.55 ± 0.16b	0.889 ± 0.002bc	3.76 ± 0.19d	0.428 ± 0.04ab
SE2 + PO	23.54 ± 0.17b	0.889 ± 0.002bc	2.55 ± 0.13e	0.340 ± 0.039a
SE2 + SO	23.49 ± 0.14b	0.893 ± 0.001b	1.69 ± 0.05f	0.833 ± 0.141ef
B	23.62 ± 0.09b	0.884 ± 0.002d	4.39 ± 0.17c	0.762 ± 0.075de
PO	23.62 ± 0.12b	0.885 ± 0.001d	4.83 ± 0.31b	0.645 ± 0.074cd
SO	23.45 ± 0.05b	0.888 ± 0.002cd	2.28 ± 0.08e	0.977 ± 0.083f

Different letters in the same column indicate significant differences among samples ($p \leq 0.05$)

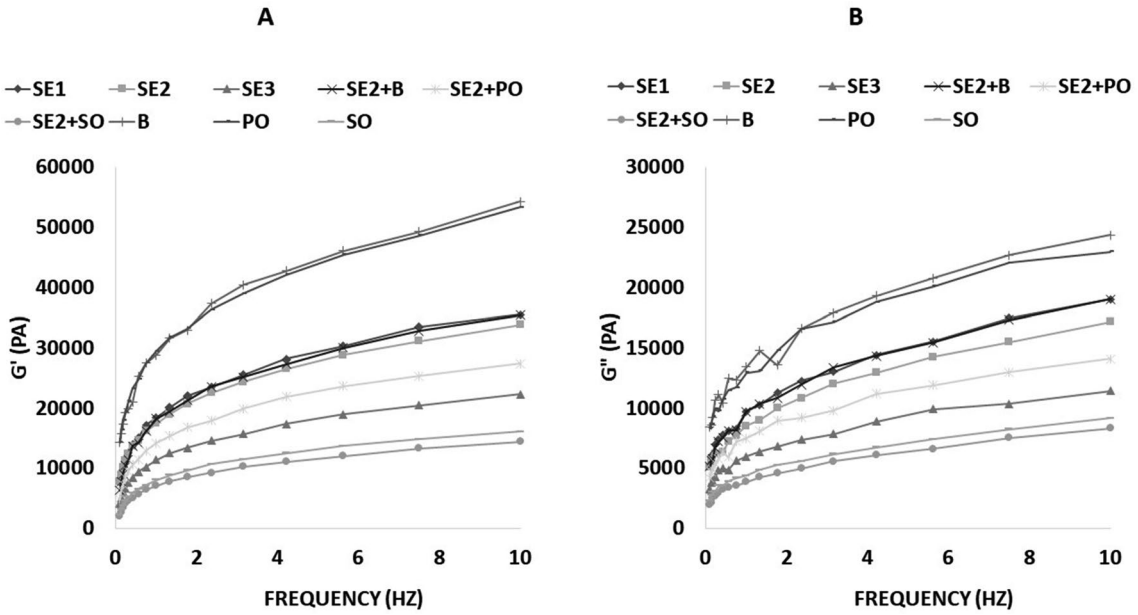

Fig. 1 G′, elastic modulus (**a**) and G″, viscous modulus (**b**) as a function of frequency (from 0.01 to 10 Hz). Abbreviations: dough formulations: SE1, SE2, SE3, B, SO, PO, SE2 + SO, SE2 + PO, and SE2 + B

Table 3 Mesoscopic properties of shortbread dough

Formulation	G′	G″	Tanδ
SE1	18,400 ± 825b	9680 ± 378b	0.53 ± 0.04ab
SE2	17,400 ± 605b	8730 ± 505bc	0.50 ± 0.03abc
SE3	11,300 ± 67.5d	5950 ± 227d	0.53 ± 0.02ab
SE2 + B	18,200 ± 931b	9690 ± 557b	0.53 ± 0.04ab
SE2 + PO	14,100 ± 372c	7470 ± 248c	0.53 ± 0.01ab
SE2 + SO	7060 ± 306e	3840 ± 226e	0.54 ± 0.01a
B	28,800 ± 1490a	13,400 ± 1120a	0.47 ± 0.02bc
PO	29,400 ± 1640a	12,900 ± 883a	0.44 ± 0.01c
SO	8040 ± 420e	4380 ± 162e	0.55 ± 0.03a

Different letters in the same column indicate significant differences among samples ($p \leq 0.05$)

affected by fat type and amount. Doughs containing saturated fats (B and PO) showed the highest G′ and G″ values, but when fiber-induced oil-in-water biphasic systems was incorporated in the formulation (SE2 + PO and SE2 + B), G′ was drastically lowered. These results are consistent with what previously found in dough made with oil–water–cellulose emulsions [5] and resistant starch and polydextrose [6].

Such a result can be attributed to the increasing amounts of unsaturated fats [25, 26]. SE2 + SO had the lowest values of G′ and G″ due to the lubrification effect of unsaturated fatty acids, and consequent dough smoothening [5]. Noteworthy, doughs, SE2 and SE2 + B, had similar G′. Tanδ (the ratio between the two modules) ranged from 0.44 to 0.55, with an average value of 0.50.

By comparing the three structured emulsion-based dough, no trend was observed in tan δ as SE1 and SE3 had the same value (0.53), while SE2 was lower with a value of 0.50. For partially substituted-based doughs, no significant difference was found between SE2 + PO and SE2 + B in tan δ (0.53). Likewise, their traditional counterparts have comparable tan δ as PO: 0.44; B: 0.47. Therefore, the partial substitution of B and PO by SE2 induced similar impact on the viscoelasticity of the dough, with a decrease in elasticity, as expected by substitution of saturated with unsaturated (more flexible) fats. The presence of saturated fats in B and PO similarly impacted the structure that was characterized by a harder network than the unsaturated counterparts [5]. SO showed the highest tan δ value (0.55) among the traditional fats, and no significant was found when SO was partially substituted by SE2. These findings might be attributed to the effect of unsaturated fatty acids on dough softening. Overall, the incorporation of structured emulsions (alone or in combination with traditional fats) induced relevant changes in dough viscoelastic properties, which are significantly related to dough softening [r (hardness, tan δ) = − 0.774] and shortbreads cookies height reduction [r (height, tan δ) = − 0.785], but no correlation was found with hardness of shortbreads cookies (unpublished data).

Molecular properties

Molecular mobility of shortbread doughs was carried out using low-resolution ^{1}H NMR, which is a valuable analytical technique that can provide important insights into water

properties and dynamics of cereals-based products (model systems, bread, and pasta) [27–32]. However, water properties and dynamics of shortbread dough are less documented [33, 34]. In this study, ^{1}H rotational mobility was studied for the fastest-relaxing component (FID) and the slowest-relaxing proton components (characterized throughout T_1 and T_2 relaxation time distributions).

Considering ^{1}H FID, two proton populations were obtained, population A (pop A) and B (pop B). The pop B described the same protons observed in the first ^{1}H T_2 population and, therefore, was not considered in the discussion. The most rigid protons, pop A, and its corresponding relaxation time (T_A) results are illustrated in Table 4, where pop A abundance progressively decreased with increasing SE content (SE1 ~ 82%, SE2 ~ 81%, SE3 ~ 79%). At constant fat/water contents, pop A of dough containing fat/oil together with structured emulsions was lower than that of mono-components (PO, SO, and B). Remarkably, SE2 + SO have the lowest pop A compared to those with saturated fats (SE2 + B and SE2 + PO). Pop A, which represents the most rigid protons of the systems, was closely related to dough hardness, as confirmed by the correlation coefficient ($r = 0.782$; $p < 0.05$). Pop A can be assigned to protons of starch and gluten, which are not in contact with water, while pop B contain probably the same protons and are assigned to protons of amorphous starch and of gluten in little contact with water [36]. T_A slightly varied as a function of fat contents (SE1: ~ 0.012 ms; SE2: ~ 0.012 ms; SE3: ~ 0.013 ms), whereas no significant differences were found between single oil/fats and their combinations with structured emulsions-based doughs (T_A: ~ 0.013 ms).

As shown in Table 4, 1H T_2 populations [pop C, D, E, and F, from the more rigid (*C*) to the more mobile protons (*F*)] and their corresponding relaxation time (T_{2C}, T_{2D}, T_{2E}, and T_{2F}, respectively) were significantly influenced by the content/type of fats added to dough formulation. Pop C was previously attributed to protons of amorphous starch and CH protons of proteins, while pop D contains protons of sucrose and pop E contains exchanging protons of water, starch, proteins, sucrose, and egg yolk lipids. Regarding the most mobile protons (pop F), no particular trend was found as a function of fat contents [35], but were also associated with protons of non-polar phase of lipids [36].

Pop C was the highest in SE1 (~ 23%), followed by SE2 (~ 22%), and SE3 (~ 20%), and hence, this trend can be related to the decrease in water content (SE1 < SE2 < SE3). Considering formulations with the same fat/water contents, B and PO did not show significant difference in pop C, but they significantly increased when combined with SE2 (SE2 + B and SE2 + PO). Pop D showed the same trend as pop C, contrary to pop E. Both T_{2D} and T_{2E} were inversely related to dough hardness ($r = \sim -0.77$, $p < 0.05$; $r = \sim -0.74$, $p < 0.05$, respectively). Relaxation time T_{2F} was significantly higher in SE2 + SO (~ 179 ms), followed by SE2 + PO (~ 175 ms), SE2 + B (~ 163 ms), SE2 (~ 159 ms), SP (~ 156 ms), PO (~ 144 ms), and B (~ 135 ms). With

Table 4: ^{1}H FID, ^{1}H T_1, and ^{1}H T_2 proton populations and corresponding relaxation times

	SE1	SE2	SE3	SE2 + B	SE2 + PO	SE2 + SO	B	PO	SO
^{1}H FID									
Population A (%)	82 ± 0.3a	81.4 ± 0.34b	79.5 ± 0.25e	80.4 ± 0.13d	80.4 ± 0.13d	79.8 ± 0.36e	82.6 ± 0.24a	81.3 ± 0.33bc	80.9 ± 0.63c
T_A (ms)	0.012 ± 0.001b	0.012 ± 0.001b	0.013 ± 0.001a	0.013 ± 0.001a	0.013 ± 0.001a	0.013 ± 0.001a	0.013 ± 0.001a	0.013 ± 0.001ab	0.013 ± 0.001b
^{1}H T_1									
T_1 (ms)	58 ± 1c	68 ± 2b	73 ± 2a	68 ± 1b	68 ± 2b	69 ± 1b	68 ± 1b	71 ± 1a	67 ± 1b
^{1}H T_2									
Population C (%)	23 ± 0.82a	22 ± 0.63bc	20 ± 0.67e	23 ± 0.84ab	23 ± 0.43a	22 ± 0.58bc	20 ± 0.41de	20 ± 1.08de	20 ± 0.65 cd
Population D (%)	42 ± 0.65a	41 ± 0.70b	41 ± 0.90b	39 ± 1.42d	39 ± 0.71 cd	40 ± 1.02bc	36 ± 1.22e	36 ± 1.94e	40 ± 1.09bcd
Population E (%)	21 ± 0.04d	24 ± 0.02c	26 ± 0.05bc	26 ± 0.05b	25 ± 0.04bc	24 ± 0.05c	32 ± 0.04a	31 ± 0.05a	25 ± 0.06bc
Population F (%)	14 ± 1.07bc	13 ± 0.89 cd	14 ± 1.38ab	13 ± 2.03de	13 ± 1.21de	14 ± 1.21ab	12 ± 1.20e	12 ± 2.27e	14 ± 1.09a
T_{2C} (ms)	1.31 ± 0.44bc	1.34 ± 0.21bc	1.66 ± 0.40a	1.3 ± 0.35 cd	1.34 ± 0.43bc	1.38 ± 0.95b	1.10 ± 0.46e	1.21 ± 0.95d	1.30 ± 0.95c
T_{2D} (ms)	9 ± 1.13c	10 ± 0.13b	11 ± 0.23a	10 ± 0.46b	11 ± 0.40a	11 ± 0.43a	8 ± 0.43d	10 ± 0.35bc	10 ± 0.24bc
T_{2E} (ms)	33 ± 0 de	36 ± 0.11bc	39 ± 0.33a	35 ± 0.53bcd	37 ± 0.27b	38 ± 0.36a	29 ± 0.31f	32 ± 0.69e	34 ± 0.38cde
T_{2F} (ms)	182 ± 17a	159 ± 3 cd	172 ± 6abc	163 ± 10bcd	175 ± 7ab	179 ± 9a	135 ± 5de	144 ± 8 ef	156 ± 4f

Different letters in the same row indicate significant differences among samples ($p \leq 0.05$)

regards to Pop F (T_{2F} ~ 130–180 ms), SE2 + SO and SO were characterized by the highest amount of pop F due to high amounts of unsaturated lipids (higher mobility). G′ and G″ were inversely related to Pop D ($r = \sim -0.7$; $p < 0.05$) and Pop F ($r = \sim -0.84$; $p < 0.01$), but positively related to Pop E ($r = \sim 0.75$; $p < 0.05$). Hence, the molecular level of this investigation was closely related to both macroscopic (hardness and moisture content) and mesoscopic (G′ and G″) levels, suggesting that TD-NMR can be a valuable analytical instrument to understand the physico-chemical changes in fat-reduced shortbread dough.

Regarding ^{1}H T_1 distributions, only one population was found in all samples, which were significantly influenced by the content/type of fats added to dough formulation. The corresponding relaxation time (T_1) increased significantly with increasing amounts of structured emulsions in the

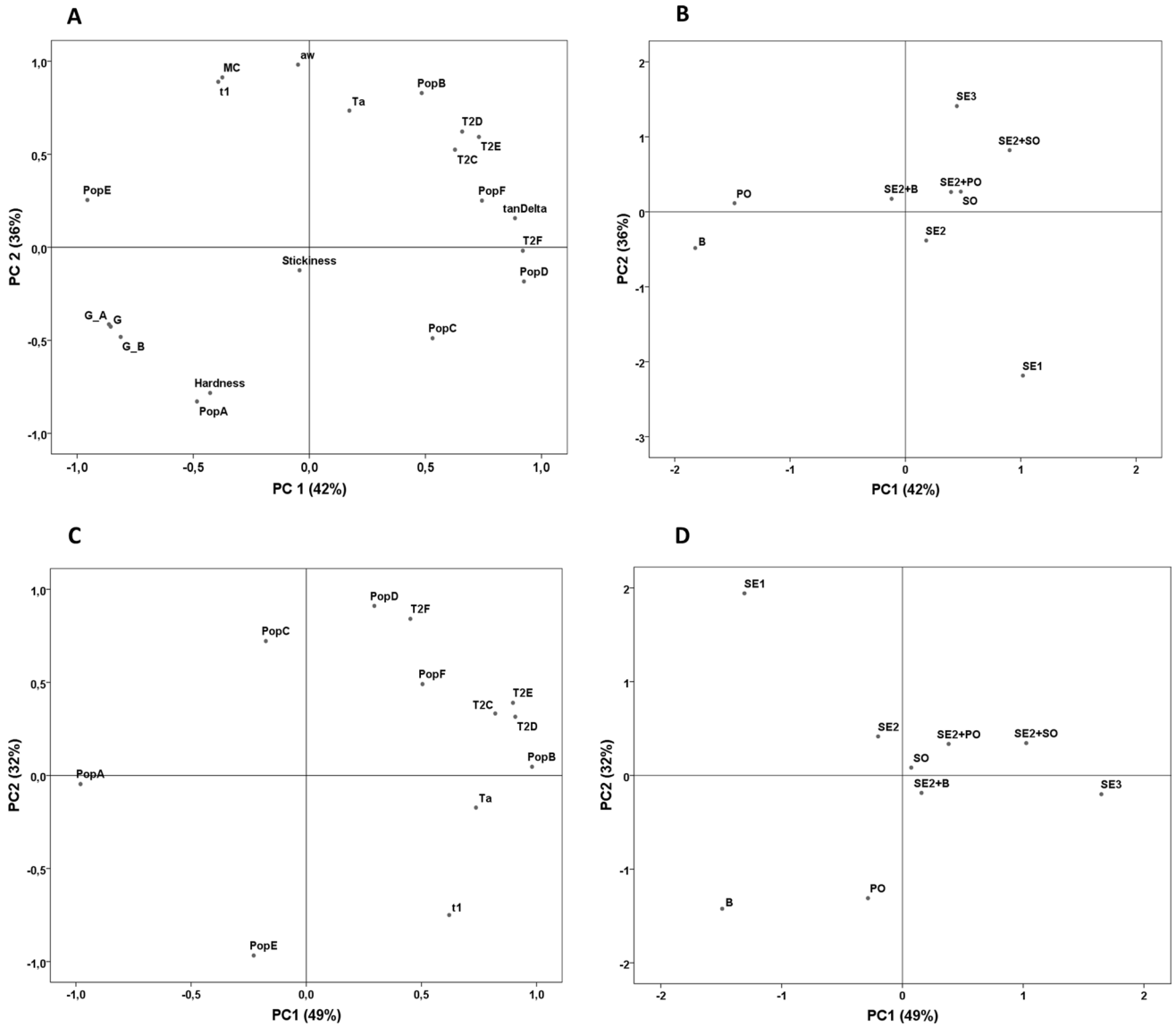

Fig. 2 Principal component analysis (PCA) results obtained for the two principal components, showing (left) the projection of the variables on the factor plane, and (right) the projection of the cases on the factor plane (dough formulations). **a** Biplot of the two first principal components based on the all studied parameters, **b** Rotated principal scores of dough formulations projected into the first two principal components, **c** Biplot of the two first principal components based on NMR parameters, **d** Rotated principal scores of dough formulations projected into the first two principal components. *dough formulations* SE1, SE2, SE3, B, SO, PO, SE2 + SO, SE2 + PO, and SE2 + B), *MC* moisture content, *aw* water activity, *Stickiness* stickiness, *Hardness* hardness, *G′* G_A, *G″* G_B, *G** G, *tan Delta* tan delta, *Population A* proton abundance population A, *Ta* relaxation time of population A, *t1* relaxation time, *T2C* T_2 relaxation time population C, *T2D* T_2 relaxation time population D, *T2E* T_2 relaxation time population E, *T2F* T_2 relaxation time population F, *Population C* proton abundance population C, *Population D* proton abundance population D, *Population E* proton abundance population E, *Population F* proton abundance population F

formulation (SE1, ~58 ms; SE2 ~68 ms; SE3 ~72 ms), which can be correlated to the increase of moisture content as confirmed by the correlation coefficient [$r \sim 0.94$, $p < 0.01$]. However, no significant differences were between SE2 and all the doughs containing single or combined fats/oil-structured probably due to constant water/fat contents used in their formulations, possibly because T_1 was able to show only a mediated signal of different molecular environments.

Multivariate statistics

Multivariate analysis was an attempt to put the pieces of the puzzle together, evaluating the usefulness of this multilevel approach to characterize fat-reduced dough formulations. Considering all the attributes (macroscopic, mesoscopic, and molecular), PCA explained 78% using the first principal components (PC1 and PC2) (Fig. 2a). PC1 (42%) was explained as a function of mesoscopic (G′, G″, and tan δ) and molecular parameters (T_{2E}, T_{2F}, T_{2D}, T_{2C} and population C, D, E, and F). PC2 (36%) was described as function of macroscopic (a_w, MC, hardness, and stickiness) and molecular parameters (T_A, T_1, and Pop A and B). Figure 2b illustrated the projection of dough formulations on the factorial space generated by PC1 and PC2. As result, three groups can be clearly separated: (i) SE1, (ii) B and PO, and (iii) the remaining formulations (SE2, SE3, SO, SE2 + SO, SE2 + PO, and SE2 + B).

Given the important contribution of NMR parameters in dough quality description, a second PCA was performed with NMR parameters only. The first two PCs explained 81% of the total variability, with PC1 accounting for 49% of total variability (T_{1A}, T_{1B}, T_{2E}, T_{2D}, T_{2C}, population A, B, and F), and PC2 for 32% (T_1, T_{2F}, T_{2C}, population C, D, and E) (Fig. 2c). As illustrated in Fig. 2d, five groups were identified [(i) SE1; (ii) SE3; (iii) B; (iv) PO; (v) SE2, SO, SE2 + SO, SE2 + PO, and SE2 + B]. Such result underlined that TD-NMR was more able to discriminate the different formulations over macroscopic and mesoscopic approaches.

Conclusion

From the above results, it can be concluded that the use of structured emulsions as fat substitutes allowed for acceptable dough technological properties, particularly doughs made with B and SE2 showed comparable hardness. The use of SE2 in shortbread dough resulted in reducing stickiness, a partial substitution of butter and palm oil with structured emulsion resulted in less sticky dough and moistened doughs. Furthermore, viscoelastic rheological properties confirmed that the addition of structured emulsions did not impact the prevalence of elastic component versus viscous component, but it decreased both components compared to the conventional fats (butter and palm oil). Low-resolution ^{1}H NMR underlined relevant changes in water properties and dynamics of the dough. Particularly, the substitution with SE in combination with saturated fats increased the rigid population (population C), while doughs with only SE were characterized by greater proton mobility (population F). Notably, multivariate statistics underlined the relevant role of NMR parameters (molecular level) in the description of dough quality features (mesoscopic and macroscopic levels). Future works are required to cover the quality of the formulated functional shortbread.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Compliance with ethics requirements This article does not contain any studies with human or animal subjects.

References

1. Melanson EL, Astrup A, Donahoo WT (2009) The relationship between dietary fat and fatty acid intake and body weight, diabetes, and the metabolic syndrome. Ann Nutr Metab 55:229–243. https://doi.org/10.1159/000229004
2. Aggarwal D, Sabikhi L, Sathish Kumar MH (2016) Formulation of reduced-calorie biscuits using artificial sweeteners and fat replacer with dairy–multigrain approach. NFS J 2:1–7. https://doi.org/10.1016/j.nfs.2015.10.001
3. Dietary Guidelines for Americans (2010) The 2010 Dietary Guidelines policy document features the final recommendations developed and released by the federal government. https://health.gov/dietaryguidelines/2010/. Accessed 3 Jan 2019
4. Biguzzi C, Lange C, Schlich P (2015) Effect of sensory exposure on liking for fat- or sugar-reduced biscuits. Appetite 95:317–323. https://doi.org/10.1016/J.APPET.2015.07.001
5. Tarancón P, Hernández MJ, Salvador A, Sanz T (2015) Relevance of creep and oscillatory tests for understanding how cellulose emulsions function as fat replacers in biscuits. LWT Food Sci Technol 62:640–646. https://doi.org/10.1016/j.lwt.2014.06.029
6. Moriano ME, Cappa C, Alamprese C (2018) Reduced-fat soft-dough biscuits: multivariate effects of polydextrose and resistant starch on dough rheology and biscuit quality. J Cereal Sci 81:171–178. https://doi.org/10.1016/J.JCS.2018.04.010
7. Pareyt B, Delcour JA (2008) The role of wheat flour constituents, sugar, and fat in low moisture cereal based products: a review on sugar-snap cookies. Crit Rev Food Sci Nutr 48:824–839. https://doi.org/10.1080/10408390701719223
8. Tarancón P, Salvador A, Sanz T (2013) Sunflower oil–water–cellulose ether emulsions as trans-fatty acid-free fat replacers in biscuits: texture and acceptability study. Food Bioprocess Technol 6:2389–2398. https://doi.org/10.1007/s11947-012-0878-6
9. Curti E, Federici E, Diantom A et al (2018) Structured emulsions as butter substitutes: effects on physicochemical and sensory attributes of shortbread cookies. J Sci Food Agric 98:3836–3842. https://doi.org/10.1002/jsfa.8899

10. Oh IK, Amoah C, Lim J et al (2017) Assessing the effectiveness of wax-based sunflower oil oleogels in cakes as a shortening replacer. LWT 86:430–437. https://doi.org/10.1016/j.lwt.2017.08.021
11. Škrbić B, Cvejanov J (2011) The enrichment of wheat cookies with high-oleic sunflower seed and hull-less barley flour: impact on nutritional composition, content of heavy elements and physical properties. Food Chem 124:1416–1422. https://doi.org/10.1016/j.foodchem.2010.07.101
12. Palla C, Giacomozzi A, Genovese DB, Carrín ME (2017) Multi–objective optimization of high oleic sunflower oil and monoglycerides oleogels: searching for rheological and textural properties similar to margarine. Food Struct 12:1–14. https://doi.org/10.1016/j.foostr.2017.02.005
13. Tarancón P, Salvador A, Sanz T et al (2015) Use of healthier fats in biscuits (olive and sunflower oil): changing sensory features and their relation with consumers' liking. Food Res Int 69:91–96. https://doi.org/10.1016/j.foodres.2014.12.013
14. Jacob J, Leelavathi K (2007) Effect of fat-type on cookie dough and cookie quality. J Food Eng 79:299–305. https://doi.org/10.1016/j.jfoodeng.2006.01.058
15. Mert B, Demirkesen I (2016) Evaluation of highly unsaturated oleogels as shortening replacer in a short dough product. LWT Food Sci Technol 68:477–484. https://doi.org/10.1016/J.LWT.2015.12.063
16. Chung C, Smith G, Degner B, McClements DJ (2016) Reduced fat food emulsions: physicochemical, sensory, and biological aspects. Crit Rev Food Sci Nutr 56:650–685. https://doi.org/10.1080/10408398.2013.792236
17. Forker A, Zahn S, Rohm H (2012) A combination of fat replacers enables the production of fat-reduced shortdough biscuits with high-sensory quality. Food Bioprocess Technol 5:2497–2505. https://doi.org/10.1007/s11947-011-0536-4
18. Błońska A, Marzec A, Błaszczyk A (2014) Instrumental evaluation of acoustic and mechanical texture properties of short-dough biscuits with different content of fat and inulin. J Texture Stud 45:226–234. https://doi.org/10.1111/jtxs.12068
19. Krystyjan M, Gumul D, Ziobro R, Sikora M (2015) The effect of inulin as a fat replacement on dough and biscuit properties. J Food Qual 38:305–315. https://doi.org/10.1111/jfq.12148
20. Rodríguez-García J, Laguna L, Puig A et al (2013) Effect of fat replacement by inulin on textural and structural properties of short dough biscuits. Food Bioprocess Technol 6:2739–2750. https://doi.org/10.1007/s11947-012-0919-1
21. Laguna L, Primo-Martín C, Varela P et al (2014) HPMC and inulin as fat replacers in biscuits: Sensory and instrumental evaluation. LWT Food Sci Technol 56:494–501. https://doi.org/10.1016/j.lwt.2013.12.025
22. AACC International (2001) AACC International approved methods of analysis. AACC International, Saint Paul
23. Le Grand F, Cambert M, Mariette F (2007) NMR signal analysis to characterize solid, aqueous, and lipid phases in baked cakes. J Agric Food Chem 55:10947–10952. https://doi.org/10.1021/jf071735r
24. Hoque MZ, Hossain KM, Akter F (2009) The effect of lecithin-a non-absorbing emulsifying agent on cookie production. Pak J Nutr 8:1074–1077
25. Agyare KK, Xiong YL, Addo K, Akoh CC (2006) Dynamic rheological and thermal properties of soft wheat flour dough containing structured lipid. J Food Sci 69:297–302. https://doi.org/10.1111/j.1365-2621.2004.tb13633.x
26. Agyare KK, Addo K, Xiong YL, Akoh CC (2005) Effect of structured lipid on alveograph characteristics, baking and textural qualities of soft wheat flour. J Cereal Sci 42:309–316. https://doi.org/10.1016/J.JCS.2005.03.008
27. Carini E, Curti E, Littardi P et al (2013) Water dynamics of ready to eat shelf stable pasta meals during storage. Innov Food Sci Emerg Technol 17:163–168. https://doi.org/10.1016/j.ifset.2012.09.010
28. Carini E, Curti E, Fattori F et al (2017) Staling of gluten-free breads: physic–chemical properties and ^{1}H NMR mobility. Eur Food Res Technol 243:867–877. https://doi.org/10.1007/s00217-016-2801-2
29. Diantom A, Curti E, Carini E et al (2019) A multi-scale approach for pasta quality features assessment. LWT 101:285–292. https://doi.org/10.1016/J.LWT.2018.11.004
30. Littardi P, Diantom A, Carini E et al (2018) A multi-scale characterisation of the durum wheat pasta cooking process. Int J Food Sci Technol. https://doi.org/10.1111/ijfs.14057
31. Hemdane S, Jacobs PJ, Bosmans GM, Verspreet J, Delcour JA, Courtin CM (2017) Study on the effects of wheat bran incorporation on water mobility and biopolymer behavior during bread making and storage using time-domain 1H NMR relaxometry. Food Chem 236:76–86
32. Rondeau-Mouro C, Cambert M, Kovrlija R, Musse M, Lucas T, Mariette F (2015) Temperature-associated proton dynamics in wheat starch-based model systems and wheat flour dough evaluated by NMR. Food Bioprocess Technol 8(4):777–790
33. Assifaoui A, Champion D, Chiotelli E, Verel A (2006) Characterization of water mobility in biscuit dough using a low-field 1H NMR technique. Carbohydr Polym 64:197–204. https://doi.org/10.1016/j.carbpol.2005.11.020
34. Serial MR, Blanco Canalis MS, Carpinella M et al (2016) Influence of the incorporation of fibers in biscuit dough on proton mobility characterized by time domain NMR. Food Chem 192:950–957. https://doi.org/10.1016/j.foodchem.2015.07.101
35. Luyts A, Wilderjans E, Waterschoot J et al (2013) Low resolution 1H NMR assignment of proton populations in pound cake and its polymeric ingredients. Food Chem 139:120–128. https://doi.org/10.1016/j.foodchem.2013.01.062
36. Todt H, Guthausen G, Burk W et al (2006) Water/moisture and fat analysis by time-domain NMR. Food Chem 96:436–440. https://doi.org/10.1016/j.foodchem.2005.04.032