LWT - Food Science and Technology 133 (2020) 109888

Contents lists available at ScienceDirect

LWT

journal homepage: www.elsevier.com/locate/lwt

Inulin-based emulsion filled gel as fat replacer in shortbread cookies: Effects during storage

Maria Paciulli [a], Paola Littardi [a], Eleonora Carini [a], Vito Michele Paradiso [c,**], Maria Castellino [b], Emma Chiavaro [a,*]

[a] Department of Food and Drug, University of Parma, Parco Area delle Scienze 47/A, 43124, Parma, Italy
[b] Department of Soil, Plant and Food Science, University of Bari Aldo Moro, Via Amendola 165/A, 70126, Bari, Italy
[c] Department of Biological and Environmental Sciences and Technologies, Ecotekne Center - S.P. 6, 73047, Monteroni - Lecce, Italy

ARTICLE INFO

Keywords:
Bakery products
Fat substitute
Storage
Thermal properties
^{1}H-NMR

ABSTRACT

Shortbread cookies with 0, 20, 40 and 50% fat replacement were obtained using an emulsion filled gel (EFG) based on inulin and extra virgin olive oil and studied during 60 storage days. Increasing the amount of EFG in shortbread cookies, higher volumes and harder texture were observed, in relation to the higher water availability and the lower fat content. Thermal analysis conducted by means of DSC and ^{1}H-NMR confirmed the key role of lipids and water status on the mechanical properties of shortbread cookies. Darker colour and toasted notes were registered for increasing levels of EFG, in relation to the presence of inulin involved in the Maillard reaction. During storage, all the cookies resulted very stable regarding dimensions, colour and rancidity perception; only slight texture changes were observed, probably related to the moisture redistribution among the cookies components. EFG can be proposed as valuable ingredient to replace fats in shortbread cookies, allowing the use of the health claim “reduced saturated fat content” already from 40% butter substitution.

1. Introduction

Dietary saturated fats (SF) and their effects on health are a topic of long debate. Recent systematic reviews and meta-analyses have called into question the effect, traditionally attributed to dietary SF, to increase the risk of insurgence of cardiovascular disease (CVD) and coronary heart disease (CHD) (Chowdhury et al., 2014; de Souza et al., 2015). As a matter of fact, current dietary guidelines suggest the reduction of SF (which should provide no more than 10–11% of total daily energy) and their replacement with unsaturated fats (UF) (Natalicchio et al., 2018; World Health Organization, 2015). Research is studying solutions for the replacement of SF with UF to obtain healthier foods, and reduce the risk of CVD, CHD and other diseases (Chiu, Williams, & Krauss, 2017; Parks, Yki-Järvinen, & Hawkins, 2017).

Edible oil structuring is a possible approach to replace SF with UF, improving the nutritional quality of foods without jeopardizing structure, texture, and mouthfeel of the products (Patel & Dewettinck, 2016). The conversion of liquid oils, rich of unsaturated fatty acids, into self-standing structured solids can be achieved in several ways, including the direct dispersion of structuring agents, indirect methods using water continuous emulsions as templates, oil sorption techniques and the development of structured biphasic systems (Patel & Dewettinck, 2016). Emulsion filled gels (EFG) are structured biphasic systems, consisting of oil droplets dispersed and physically entrapped in an aqueous phase gelled using hydrocolloids (Patel & Dewettinck, 2016). Several applications have been proposed for EFG as fat/saturated fat replacers, in fresh sausages (Pintado, Herrero, Jiménez-Colmenero, Pasqualin Cavalheiro, & Ruiz-Capillas, 2018), salad dressings (Mantzouridou, Spanou, & Kiosseoglou, 2012) and bakery products (Curti et al., 2018). The nutritional improvement achieved by replacing saturated fats with unsaturated ones can be further increased by formulating EFG with high-value ingredients, sources of fibre, minerals, antioxidants or vitamins. As examples, chia flour, oat bran, inulin, extra virgin olive oil, rice bran oil, β-carotene have been considered in the formulation of EFG (Nourbehesht, Shekarchizadeh, & Soltanizadeh, 2018; Pintado et al., 2016, 2018).

In our previous research, an EFG based on inulin and extra virgin olive oil was developed and used as total or partial butter replacer in

---

* Corresponding author.
** Corresponding author.
E-mail addresses: vito.paradiso@unisalento.it (V.M. Paradiso), emma.chiavaro@unipr.it (E. Chiavaro).

https://doi.org/10.1016/j.lwt.2020.109888
Received 5 May 2020; Received in revised form 11 July 2020; Accepted 13 July 2020

shortbread cookies (Giarnetti, Paradiso, Caponio, Summo, & Pasqualone, 2015; Paradiso et al., 2015). The obtained EFG combined the phenolic content, oxidative stability and desirable fatty acid composition of extra virgin olive oil with the prebiotic properties of inulin as soluble dietary fibre (Cerretani et al., 2012; Shoaib et al., 2016). From the results of this study was visible that cookies obtained by replacing 50% butter with EFG showed an improved nutritional profile; moreover, they resulted comparable to control cookies as regards to microstructure and fracture properties, being also well accepted by consumers. Scarce information is available in literature regarding the effect of fat replacement with EFG on physicochemical and sensory changes occurring during storage of bakery products. The present paper, with the aim of filling this gap, reports the evolution of the sensory and physical characteristics of shortbread cookies with different levels (0, 20, 40 and 50%) of butter replacement with an EFG based on inulin and extra virgin olive oil during a storage period of 60 days.

2. Materials and methods

2.1. Samples preparation

The ingredients were those reported by (Giarnetti et al., 2015). The formulation and preparation of the EFG followed the indications of (Paradiso et al., 2015). Briefly, Extra Virgin Olive Oil (EVOO) (37 g/100g), inulin (19 g/100g), soy lecithin (2 g/100g), and water (42 g/100g) were mixed and omogenized for 5 min with high power ultrasound using a 200 W transducer (Sonopuls HD 3200, Bandelin Electronic, Berlin, Germany) with 6 mm diameter tapered tip (KE 76, Bandelin Electronic, Berlin, Germany).

Four shortbread cookie typologies were prepared using the raw materials and the method reported by Giarnetti et al. (2015) and Paradiso et al. (2015) by using the same quantity of all the ingredients and different butter/EFG proportions: 100:0 (EFG0), 80:20 (EFG20), 60:40 (EFG40), and 50:50 (EFG50) (Table 1). The four cookie treatments were arranged in the baking trays according to Latin-square design, then cookies were cooled down and stored in polyethylene sealed bags for 60 days at 40% RH and 25 °C in a Constant Climate Chamber (HPP/749, Memmert, Germany). Cookies were analysed at sampling intervals of 0, 7, 15, 30, 60 days.

2.2. Fat analysis

Fat was extracted and determined by Soxhlet apparatus using diethyl ether as solvent (AOAC, 2000 Official Method 920.39). Fatty acid composition was determined according to (Caponio, Summo, Paradiso, Pasqualone, & Gomes, 2009). Samples were analysed in triplicate.

2.3. Physico-chemical characterization during storage

- Moisture content and water activity

Moisture content (MC, g/100g) was determined on crumbled cookies according to the AACC standard method, 44–15.02 (AACC, 2000). Water activity (a_w) was determined on the crumbled cookies by AcquaLab (Decagon Devices Inc, Pullman, Washington, USA) at room temperature (25 °C). For each formulation and storage time, triplicate analysis was carried out.

Table 1
Cookies formulations.

		EFG 0	EFG 20	EFG 40	EFG 50
Flour	*g*	300	300	300	300
Butter	*g*	130	104	78	65
EFG	*g*	0	26	52	65
Sugar	*g*	100	100	100	100
Eggs	*n*	2	2	2	2
Leavening agent	*g*	8	8	8	8
Salt	*g*	2	2	2	2

- Dimensions and volume

Base diameter and height of the cookies were measured using a calliper. Volume (cm^3/g) was determined according to the Rapeseeds Displacement method (AACC method 10–05.01). The analysis were conducted on four replicate samples for each formulation and storage time.

- Texture

A TA-TX2i Texture Analyzer equipped with a 25 kg load cell (Stable Micro Systems, Goldamin, UK) and a 3 mm aluminium cylinder probe (P/3) was used to evaluate hardness (Newton, N) of cookies, by means of penetration test. The test was conducted at room temperature setting the following parameters: strain: 50%; test speed: 2 mm/s. Ten replicates were measured for each formulation and storage time.

- Colour

Colour determination was performed using the Minolta Colorimeter (CM 2600d, Minolta Co., Osaka, Japan) equipped with a standard illuminant D65 at room temperature (25 °C). The Spectramagic Software (version 3.6) was used to measure the CIE-Lab colour parameters L^*, a^* and b^* (lightness, redness and yellowness indexes, respectively), using 10° position of the standard observer (Commission Internationale de l'Eclairage, 1978). The colour difference (ΔE) between fresh and stored cookies as well as between EFG0 and the other cookies formulations was calculated as $\Delta E = \sqrt{(L^*_R - L^*_E)^2 + (a^*_R - a^*_E)^2 + (b^*_R - b^*_E)^2}$, where R refers to the reference cookie and E to the experimental cookies. Eleven measurements were performed for each condition.

2.4. Thermal analysis

Thermal analysis was conducted by Differential Scanning Calorimetry (DSC Q100, TA Instruments, New Castle, DE, USA) calibrated with indium (melting point: 156.6 °C, melting enthalpy: 28.71 J/g) and mercury (melting point: −38.83 °C, melting enthalpy: 11.44 J/g). About 10 mg of crumbled sample were put into hermetic stainless-steel pans (Perkin Elmer, Waltham, MA, USA), quickly cooled from 30 °C to −80 °C and then heated from −80 °C to 120 °C at 5 °C/min using an empty pan as reference. Peak temperature (T_p, °C) and enthalpy (ΔH, Jg^{-1}) of the transitions observed upon heating were obtained by Universal Analysis Software (version 3.9A, TA Instruments). Only the transitions observed upon heating were taken into consideration being more informative for the purpose of this study. At least triplicates were analysed for each condition.

2.5. Molecular mobility during storage (^1H NMR relaxometry)

The ^1H molecular mobility and dynamics of cookies during storage was studied with a low-resolution ^1H NMR spectrometer (20 MHz, the miniSpec, Bruker Biospin, Milano, Italy) working at 25.0 ± 0.1 °C. About 2–3 g of crumbled cookies were put into a 10 mm NMR tube, sealed with Parafilm® and analysed with a ^1H Free Induction Decay (FID) and ^1H transverse relaxation time (^1H T_2) experiments, the former to observe less mobile protons and the latter to observe the more mobile ones. The recycle delay was set at 1 s ($\geq 5\ T_1$) for both experiments. The FID curves were obtained operating with 32 scans, a dwell time to 7 μs, an acquisition window to 0.5 ms and 900 data points. Experimental curves were fitted (Sigmaplot, v10, Systat Software Inc. USA) using a multi-component model (exponential and Gaussian, modified with a Pake function; (Derbyshire et al., 2004), MATLAB 2016a, The MathWorks Inc. USA)

$$F(t) : y0 + A * exp\left(-t/T_A\right) + B * \sin[(c * t) \Big/ (c * t) * \exp\left[-\left(t/T_B\right)^2 * 0.5\right] \quad \text{(Equation 1)}$$

where $y0$ is the FID decay offset, A and B are the intensities of each relaxation component, T_A and T_B the apparent relaxation times.

An interpulse spacing of 0.087 ms, 32 scans and 16000 data points were the parameters used for the ^{1}H T_2 experiment. Quasi-continuous distributions of relaxation times were obtained using the UPENWin software (Alma Mater Studiorum, Bologna, Italy), setting up default values for all UPEN parameters (except LoXtrap = 1, to avoid extrapolation of times shorter than the first experimental point). ^{1}HT_2 curves were also fitted with a discrete multiexponential model (Sigmaplot, v.10, Systat Software Inc. USA) to obtain relaxation times (ms) and relative abundances (%) of each proton population. At least twelve ^{1}H FID and ^{1}H T_2 experimental curves were performed for each sample and storage time.

2.6. Sensory evaluation

Quantitative Descriptive Analysis (QDA) was performed by a panel of 15 assessors, according to Giarnetti et al. (2015). A lexicon of 8 descriptors, regarding 4 characteristics (odour, surface appearance, hardness and aroma), was defined through consensus sessions (Table 2). Samples were analysed in duplicate for each storage time.

2.7. Statistical analysis

Two-way ANOVA was performed taking in consideration two fixed factors: formulation (FORM) and storage time (ST). The partition of total variance of sum squares (SS%) was computed to evaluate the contribution of single factors and their interaction in the variability of each parameter (IBM SPSS v.23, New York, USA). A one-way-analysis of variance (ANOVA) at $p \leq 0.05$ was carried out to verify significant differences in the evaluated parameters among formulations and during storage. Significant differences among the mean values were calculated using Duncan's test ($p \leq 0.05$). Pearson correlation coefficients were calculated among all variables considering 95% and 99% confidence levels ($p < 0.05$ and $p < 0.01$).

3. Results and discussion

3.1. Fat contents

All butter substitution levels showed, as expected, significant decreases of the saturated fat contents, resulting 14.4, 11.3, 9.0 and 6.6 g 100 g^{-1} in EFG0, EFG20, EFG40 and EFG50, respectively (Fig. 1). According to EU Regulations (Regulation (EC) No 1924/2006), EFG40 and EFG50, with a reduction of 37.5 and 54.2 g/100g, could be labelled for "reduced saturated fat content", showing reductions above the reference threshold of 30 g/100g.

Table 2
Descriptors adopted for the sensory evaluation of Cookies.

Odour	Butter Toasted Rancid
Surface appearance	Porosity
Hardness	Breakage resistance
Aroma	Butter Toasted Rancid

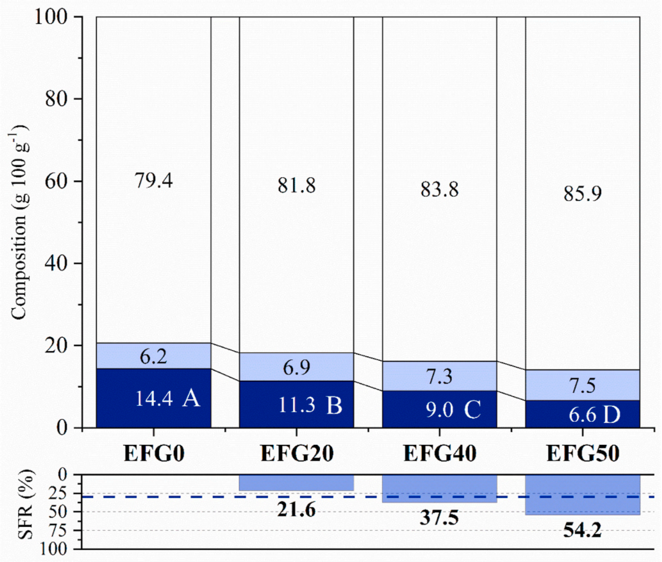

Fig. 1. Composition of cookies with different fat substitution levels and focus on the saturated/unsaturated lipid fraction.
Different letters indicate significant differences in saturated fat content at $p < 0.05$. ■ Saturated Fats 🟦 Unsaturated Fats □ Other Components. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG; SFR, saturated fats reduction. The blue dashed line indicates the value of 30% reduction, as reference for nutritional labelling. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.2. Physico-chemical characterization during storage

3.2.1. Moisture content and water activity

Table 3 reports the values of moisture content (MC) and water activity (a_w) for the different cookie's formulations. Both these parameters are involved in the stability of cookies during storage, mainly

Table 3
Moisture content and water activity of cookies with different fat substitution levels during storage*.

MC (%)					
	t0	t7	t15	t30	t60
EFG0	5.30 ± 0.13 ab/AB	5.43 ± 0.10 bc/A	5.49 ± 0.04 bc/A	5.33 ± 0.13 b/A	5.13 ± 0.04 b/B
EFG20	4.86 ± 0.13 b/B	5.12 ± 0.10 c/A	5.14 ± 0.06 c/A	5.05 ± 0.10 b/A	5.15 ± 0.13 b/A
EFG40	5.54 ± 0.30 a/B	5.76 ± 0.26 ab/AB	6.02 ± 0.43 ab/A	5.61 ± 0.36 ab/B	5.68 ± 0.26 a/AB
EFG50	5.52 ± 0.05 a/C	5.97 ± 0.01 a/B	6.42 ± 0.05 a/A	6.03 ± 0.05 a/B	6.05 ± 0.00 a/B
a_w					
	t0	t7	t15	t30	t60
EFG0	0.439 ± 0.009 a/B	0.387 ± 0.009 b/C	0.359 ± 0.011 c/D	0.459 ± 0.008 ab/A	0.468 ± 0.005 a/A
EFG20	0.413 ± 0.003 b/B	0.346 ± 0.017 c/C	0.344 ± 0.016 c/C	0.455 ± 0.004 b/A	0.462 ± 0.004 a/A
EFG40	0.433 ± 0.006 a/B	0.417 ± 0.009 a/C	0.394 ± 0.006 b/D	0.458 ± 0.015 ab/A	0.460 ± 0.005 a/A
EFG50	0.431 ± 0.003 a/B	0.427 ± 0.004 a/B	0.434 ± 0.010 a/B	0.474 ± 0.004 a/A	0.468 ± 0.004 a/A

*Different small letters in the same column indicate significant differences among formulations at the same storage time ($p < 0.05$); different capital letters in the same row indicate significant differences during storage for the same formulation ($p < 0.05$). Abbreviations: MC, Moisture content; a_w, water activity. EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. t0, t7, t15, t30, t60, days of storage.

influencing chemical reactions (e.g. lipid oxidation) and rheological properties of the products.

According to the results of the two-way ANOVA, MC was mostly affected by the effect of formulation (FORM) (SS% 77.5, $p \leq 0.001$). In particular, MC increased for higher levels of EFG substitution, in accordance with Giarnetti et al. (2015), because of the higher water content of EFG in comparison to butter. Moreover, the presence of inulin in EFG, able to retain water, would not allow large water evaporation during baking (Rodríguez-García, Laguna, Puig, Salvador, & Hernando, 2013). This trend was observed throughout the all storage period.

On the other hand, a_w was more influenced by the effect of storage time (ST) (SS% 70.5, $p \leq 0.001$). a_w decreased until T15 and then increased up to ~0.46–0.47 at T60, for all the formulations. These fluctuations may be ascribed to a water redistribution among the cookies components (Paciulli et al., 2018). EFG50 did not show any a_w decrease in the first 15 days of storage, probably because of the higher presence of inulin that bound water.

3.2.2. Dimensions and volume

The presence of fat in cookies contributes to their expansion during baking (Rodriguez Garcia et al., 2013). In this study, increasing the EFG content, shortbread cookies decreased in diameter and increased in height (data not shown). Low spread was already reported for fat reduced short dough biscuits and related to the minor fluidity of the dough, due to lower amount of fat that melts during baking (Rodriguez Garcia et al., 2013). Moreover, fat coats proteins and starch granules, making them less accessible to water. Consequently, the higher gluten hydration in fat reduced formulations hardens the doughs leading to less spread biscuits (Rodríguez-García et al., 2013).

The measured specific volume of the cookies resulted significantly affected by FORM (SS% ~59, $p \leq 0.001$), while ST did not have any effect, as expected. The measured volume was 2.27 cm^3/g for EFG0, 2.43 cm^3/g for EFG20 and increased up to 2.7 cm^3/g for EFG40 and EFG50. Generally, one of the major roles of fats in cookie doughs is aeration, consisting in the presence of more air cells that leads to increased volume (Rodríguez-García et al., 2013). In this study an opposite trend was observed; it may be explained with the higher level of water and lower amount of lipids in the EFG richer doughs, probably responsible for a larger gluten network development and thus for the higher cookies volumes.

3.2.3. Texture and colour

High fats and low water levels have definitely a key role on limiting the gluten development in shortbread cookies, phenomenon responsible for the brittle structure (Devi & Khatkar, 2018).

Table 4 reports the values of Hardness (N) measured on the shortbread cookies. The two-way ANOVA revealed that the most important factor driving the evolution of textural parameter was FORM (SS% 74, $p \leq 0.001$). Hardness progressively increased with the EFG substitution level. Hardness resulted negatively correlated with both total fat content ($R = -0{,}990$; $p < 0.05$) and saturated fats ($R = -0{,}980$; $p < 0.05$), while it resulted positively correlated with unsaturated fats ($R = 0{,}980$; $p < 0.05$). Devi and Khatkar (2018) found the same correlations studying the effect of fatty acids composition on textural properties of dough and cookie quality. Saturated fats contribute to superior air incorporation during dough mixing, resulting in softer cookies after baking. Moreover, saturated fats cover proteins reducing their entanglement and leading to the formation of a more brittle structure. Moreover, the EFG richer formulations, carrying more water and less fats, allowed a higher hydration of the flour particles; it led to a higher gluten network development in the dough and a consequent hardening of the cookies (Rodríguez-García et al., 2013).

Table 4
Hardness (N) of cookies with different fat substitution levels during storage.

	Hardness (N)				
	t0	t7	t15	t30	t60
EFG 0	12.30 ± 0.67 b/B	13.88 ± 1.68 a/AB	14.40 ± 2.40 b/A	13.48 ± 2.17 a/AB	12.58 ± 1.50 a/B
EFG 20	13.92 ± 1.64 ab/A	13.87 ± 1.83 a/A	13.97 ± 1.50 b/A	13.44 ± 1.38 a/AB	11.80 ± 1.50 a/B
EFG 40	14.82 ± 1.42 a/A	14.02 ± 1.31 a/AB	14.77 ± 2.34 ab/A	13.75 ± 1.83 a/AB	12.59 ± 1.96 a/B
EFG 50	15.63 ± 3.45 a/A	16.04 ± 3.65 a/A	16.84 ± 2.69 a/A	14.01 ± 1.58 a/AB	12.21 ± 2.10 a/B

*Different superscript small and capital letters indicate significant differences ($p \leq 0.05$) among formulations and different storage time, respectively. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. t0, t7, t15, t30, t60, days of storage.

The effect of ST on cookies hardness was less evident (SS% 12.33, $p \leq 0.01$), indeed only a slight decrease of force was observed after long storage times of (30–60 days). Textural changes observed on this kind of products during storage have been mainly attributed to phenomena of moisture migration and its redistribution within the sample components (Paciulli et al., 2018).

Table 5 reports the colour parameters measured on the shortbread cookies. As expected, the colour parameters were mainly affected by FORM (SS% 54.11, 55.79, 71.33, $p \leq 0.001$ for L^*, a^* and b^*, respectively). The influence of formulation therefore, had the largest influence on yellowness, represented by the parameter b^*. This colour index showed a slight reduction as the EFG content increased. This may be due to the presence of inulin in EFG which, providing reducing sugar, may accelerate the Maillard reaction leading to the formation of brown melanoidin (Laguna, Primo-martín, Varela, Salvador, & Sanz, 2014). On the other hand, the colour of cookies resulted almost stable during storage. The total colour difference, ΔE, is reported to be not perceptible to the human eye when lower than 3. In this study very low values of ΔE were registered, indicating that the colour differences due to either formulation or storage were not detectable by the consumers.

3.3. Thermal analysis

DSC was used to study the thermal properties of shortbread cookies. Table 6 reports peak temperatures (T_p) and enthalpies (ΔH) registered for each thermal event observed during heating.

Starting from the lower temperatures, the first detectable event for all the formulations was an exothermic peak (0) around −20 °C (Fig. 2, data not shown). The presence of an exothermic peak during heating may be attributed to a solid–solid polymorphic transformation resulting from the rearrangement of a portion of the fat crystals formed during cooling into more thermodynamically stable polymorphic forms (Barba, Arrighetti, & Calligaris, 2013). Chiavaro, Vittadini, and Corradini (2007) reported that inulin shows a glass transition upon heating between −25 and −30 °C. In the present work, the glass transition of inulin contained in EFG was not visible, probably because its overlapping with peak 0.

Further temperature increase caused other two endothermic events, both associated to fat melting (Chevallier, Colonna, Valle, & Lourdin, 2000): the first one at about 18 °C (1), followed by the second one at about 40 °C (2). T_p and the ΔH for the transitions 1 and 2 (Table 6) resulted predominantly affected by FORM (SS% T_{p1}: 96.07 $p < 0.001$; ΔH_1: 82.02 $p < 0.001$; T_{p2}: 45.47 $p < 0.01$; ΔH_2: 72.15 $p < 0.001$). Indeed, as the level of EFG substitution increased, T_{p1} and ΔH_1 significantly decreased (Table 6). T_{p1} reduction may be attributable to the increase of unsaturated triglycerides carried by EFG ($R = 0.993$, $p < 0.01$), known to melt at lower temperatures (Chiavaro, Vittadini, Rodriguez-Estrada, Cerretani, & Bendini, 2008). ΔH_1 reduction may be ascribable to the lower fat content of the EFG-enriched formulations (Fig. 1) ($R = 0.991$, $p < 0.01$). Focusing on the transition 2, ΔH_2 resulted correlated to the saturated fat fraction ($R = 0.954$, $p < 0.05$), indeed it decreased in the EFG enriched formulations. T_p as well as ΔH of the transitions 1 and 2 were highly correlated with volume and hardness of

Table 5
L*, a^*, b* colour parameters of cookies with different fat substitution levels during storage.

		t 0	ΔE	t7	ΔE	t15	ΔE	t30	ΔE	t60	ΔE
EFG-0	L* a* b*	74.18 ± 1.87 a/A 9.81 ± 1.04 b/AB 35.24 ± 1.37 a/A	–	72.29 ± 1.95 a/AB 10.65 ± 1.75 bc/B 35.58 ± 1.68 ab/A	–	72.79 ± 2.75 b/A 10.33 ± 1.53 b/AB 35.38 ± 1.07 a/A	–	70.84 ± 3.18 b/B 9.89 ± 1.45 a/AB 35.36 ± 1.06 a/A	–	73.56 ± 1.97 ab/A 9.48 ± 1.48 b/B 36.25 ± 1.55 a/A	–
ΔE		–		2.2		1.5		2.7		1.1	
EFG-20	L* a* b*	70.01 ± 2.98 b/B 11.05 ± 0.86 a/AB 34.46 ± 1.21 ab/BC	4.4	70.95 ± 1.78 a/AB 11.86 ± 1.30 a/A 35.61 ± 1.09 a/A	1.7	72.57 ± 1.77 b/A 10.31 ± 1.18 b/B 34.82 ± 0.82 ab/C	0.7	71.66 ± 2.07 ab/AB 10.31 ± 1.18 a/B 35.30 ± 1.57 a/AB	0.7	71.65 ± 3.68 b/AB 10.76 ± 1.89 a/AB 35.53 ± 1.22 a/A	2.3
ΔE		–		1.8		2.6		2.0		2.2	
EFG-40	L* a* b*	73.38 ± 1.93 a/A 9.66 ± 1.02 b/A 34.03 ± 1.55 ab/A	1.3	72.42 ± 3.52 a/A 10.12 ± 0.85 c/A 34.38 ± 1.30 c/A	1.6	74.68 ± 0.99 a/A 9.43 ± 0.81 b/AB 34.16 ± 1.53 ab/A	2.6	73.43 ± 1.16 a/A 9.54 ± 0.82 a/AB 34.90 ± 0.88 ab/A	2.1	74.87 ± 2.37 a/A 8.73 ± 1.31 b/B 34.89 ± 1.58 b/A	2.0
ΔE		–		1.3		1.1		0.6		1.7	
EFG-50	L* a* b*	70.65 ± 3.46 b/B 10.71 ± 1.00 a/AB 33.44 ± 1.06 b/A	4.1	71.45 ± 1.32 a/AB 11.36 ± 0.60 ab/A 34.66 ± 1.15 bc/A	2.0	71.50 ± 2.05 b/AB 11.50 ± 1.04 a/A 33.69 ± 1.13 c/A	2.0	72.77 ± 2.35 a/A 10.31 ± 1.33 a/BC 34.44 ± 1.04 b/A	2.3	72.69 ± 1.39 b/AB 9.82 ± 0.94 ab/C 34.51 ± 1.51 c/A	2.8
ΔE		–		1.6		1.4		2.2		2.2	

*Different small and capital letters indicate significant differences ($p \leq 0.05$) among formulations and storage times, respectively. ΔE in column indicate colour differences of the formulations in comparison at EFG0 at the same time of storage; ΔE in raw indicate colour differences among the time of storage in comparison to t0 for the same formulation. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. t0, t7, t15, t30, t60, days of storage. L^*, lightness; a^*, redness; b^*, yellowness.

Table 6
Tp (°C) and ΔH (J/g) for thermal transition Fus1. Fus2. Fus3 in different formulations of cookies during storage*.

	Fus1 T_p (°C)					Fus1 ΔH (J/g)				
	t0	t7	t15	t30	t60	t0	t7	t15	t30	t60
EFG	18.78 ± 0.13 a/AB	19.26 ± 0.26 a/A	18.46 ± 0.21 a/B	18.82 ± 0.44 a/AB	18.54 ± 0.28 a/B	9.76 ± 0.08 a/AB	6.99 ± 3.73 a/B	11.34 ± 0.29 a/A	10.33 ± 0.39 a/AB	9.80 ± 0.62 a/AB
EFG20	17.76. ± 0.4 b/A	18.20 ± 0.12 b/A	17.72 ± 0.28 a/A	17.91 ± 0.03 b/A	17.80 ± 0.15 b/A	8.02 ± 1.16 a/A	7.60 ± 0.53 a/A	8.83 ± 0.76 b/A	8.22 ± 0.22 b/A	8.35 ± 0.08 b/A
EFG40	16.36 ± 0.34 c/A	16.62 ± 0.16 c/A	16.62 ± 0.41 b/A	16.67 ± 0.51 c/A	16.15 ± 0.11 c/A	5.76 ± 0.75 b/B	6.31 ± 0.12 a/AB	7.33 ± 0.69 c/A	6.17 ± 0.54 c/AB	6.55 ± 0.43 c/AB
EFG50	15.51 ± 0.42 d/A	15.37 ± 0.14 d/A	14.94 ± 0.81 c/A	15.97 ± 0.28 c/A	15.52 ± 0.01 d/A	4.67 ± 0.38 b/A	4.12 ± 0.43 a/A	5.49 ± 0.97 d/A	4.53 ± 0.25 d/A	4.98 ± 0.49 d/A
	Fus2 T_p (°C)					**Fus2 ΔH (J/g)**				
	t0	t7	t15	t30	t60	t0	t7	t15	t30	t60
EFG0	40.13 ± 0.33 b/A	38.85 ± 2.15 a/A	39.18 ± 0.20 a/A	39.74 ± 0.32 ab/A	39.04 ± 0.27 a/A	3.06 ± 0.17 a/B	1.90 ± 1.01 a/AB	3.22 ± 0.07 a/A	2.90 ± 0.25 a/AB	3.00 ± 0.25 a/AB
EFG20	38.75 ± 0.44 c/A	39.17 ± 0.09 a/A	38.73 ± 1.12 a/A	38.61 ± 0.62 b/A	38.70 ± 0.62 a/A	1.79 ± 0.31 b/A	1.60 ± 0.24 a/A	1.95 ± 0.01 b/A	1.75 ± 0.14 b/A	1.95 ± 0.02 b/A
EFG40	40.10 ± 0.52 b/AB	39.62 ± 0.19 a/AB	40.37 ± 1.35 a/AB	40.88 ± 0.82 a/A	38.88 ± 0.27 a/B	1.21 ± 0.31 b/A	1.21 ± 0.16 a/A	1.46 ± 0.25 c/A	1.17 ± 0.09 c/A	1.52 ± 0.10 c/A
EFG50	42.03 ± 0.32 a/A	39.71 ± 0.04 a/CD	40.66 ± 0.07 a/B	40.20 ± 0.23 a/BC	39.26 ± 0.50 a/D	2.52 ± 0.29 a/A	1.09 ± 0.18 a/B	1.17 ± 0.10 c/B	1.06 ± 0.06 c/B	1.10 ± 0.09 d/B
	Fus3 T_p (°C)					**Fus3 ΔH (J/g)**				
	t0	t7	t15	t30	t60	t0	t7	t15	t30	t60
EFG0	n.d.	56.33 ± 0.74 a/A	57.68 ± 1.80 ab/A	55.29 ± 2.44 ab/A	55.66 ± 0.34 b/A	n.d.	0.60 ± 0.04 a/B	1.08 ± 0.06 a/A	0.52 ± 0.17 b/B	0.62 ± 0.02 c/B
EFG20	n.d.	54.31 ± 0.42 a/AB	55.08 ± 0.34 c/A	54.31 ± 0.07 b/AB	53.47 ± 0.35 c/B	n.d.	0.69 ± 0.19 a/AB	1.04 ± 0.11 a/A	0.72 ± 0.04 ab/AB	0.39 ± 0.1 c/B
EFG40	n.d.	54.25 ± 0.45 a/C	56.50 ± 0.57 bc/A	54.71 ± 0.85 b/BC	56.06 ± 0.11 b/AB	n.d.	0.78 ± 0.08 a/B	0.87 ± 0.18 ab/B	0.46 ± 0.07 b/C	1.49 ± 0.01 a/A
EFG50	n.d.	57.17 ± 3.42 a/A	59.81 ± 0.38 a/A	58.52 ± 0.11 a/A	59.03 ± 0.63 a/A	n.d.	0.54 ± 0.24 a/B	0.65 ± 0.03 b/B	0.80 ± 0.14 a/AB	1.21 ± 0.15 b/A

*Different small and capital letters indicate significant differences ($p \leq 0.05$) among formulations and storage time, respectively. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. t0, t7, t15, t30, t60, days of storage. Fus, fusion; T_p, peak temperature; H, enthalpy.

shortbread cookies, showing how thermal phenomena can explain the changes in mechanical and structural properties of the material.

The last endothermic transition (3), peaking at about 55 °C, was previously attributed, in low moisture systems, to the physical aging of amorphous starch (Chung & Lim, 2006). It cannot be ascribed to starch retrogradation, due to the very low water content of cookies (Slade & Levine, 1994), but rather to a moisture redistribution phenomenon (Kulp, Olewnik, Lorenz, & Collins, 1991). It is a structural relaxation phenomenon characterized by the movement of amorphous starch at a temperature below its glass transition. It is reported that the enthalpy of

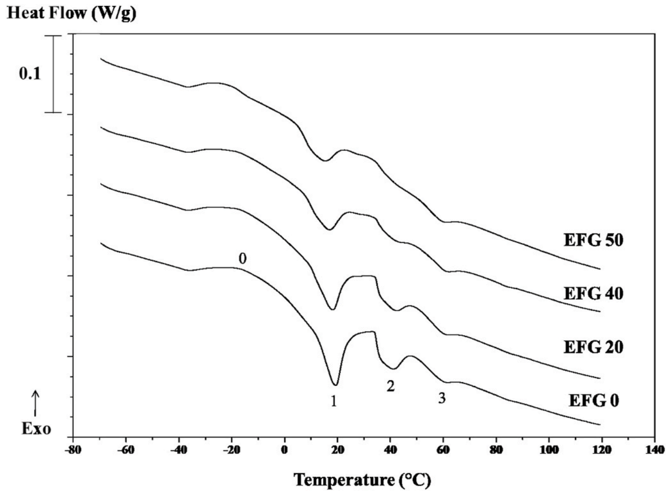

Fig. 2. DSC heating curves of cookies with different fat substitution levels after 60 days of storage (t60).
Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50 % butter substitution with EFG.

this endotherm increases with increasing aging time and moisture contents (Shogren, 1992). It may justify the high dependence of the enthalpy relaxation phenomenon from the interaction FORM*ST (SS% 67.45, P $<$ 0.001).

3.4. *Molecular mobility during storage (^{1}H NMR relaxometry)*

^{1}HNMR was used to describe the influence of formulation and storage on proton molecular mobility and dynamics. Fig. 3a reports the evolution of ^{1}H FID curves for EFG0 during storage. The line-shape of the curves indicated the presence of a very fast decay in the first part and the presence of a hump in the second part with no differences due to the storage times. A similar behaviour was also observed in the EFG-containing samples (Fig. 3b). This shape was previously attributed to the crystalline phase (Derbyshire et al., 2004) that in these samples may arise from starch, sucrose and butter domains. Two protons populations (FIDA and FIDB) and their relative abundances (%FIDA and %FIDB) were obtained by fitting the experimental curves. Only ^{1}H belonging to population A has been considered for the discussion (Table 7), because population B showed ^{1}H relaxing at times comparable to those of protons of less mobile ^{1}HT$_2$ population. Focusing on the relaxation time of FID A (Table 7), it resulted more affected by FORM (SS% $\sim$ 57, p $\leq$ 0.001) than by ST; it showed a significant but very small increase with the level of butter substitution, indicating a slight higher mobility of this proton domain. This may be hypothetically related to the increased

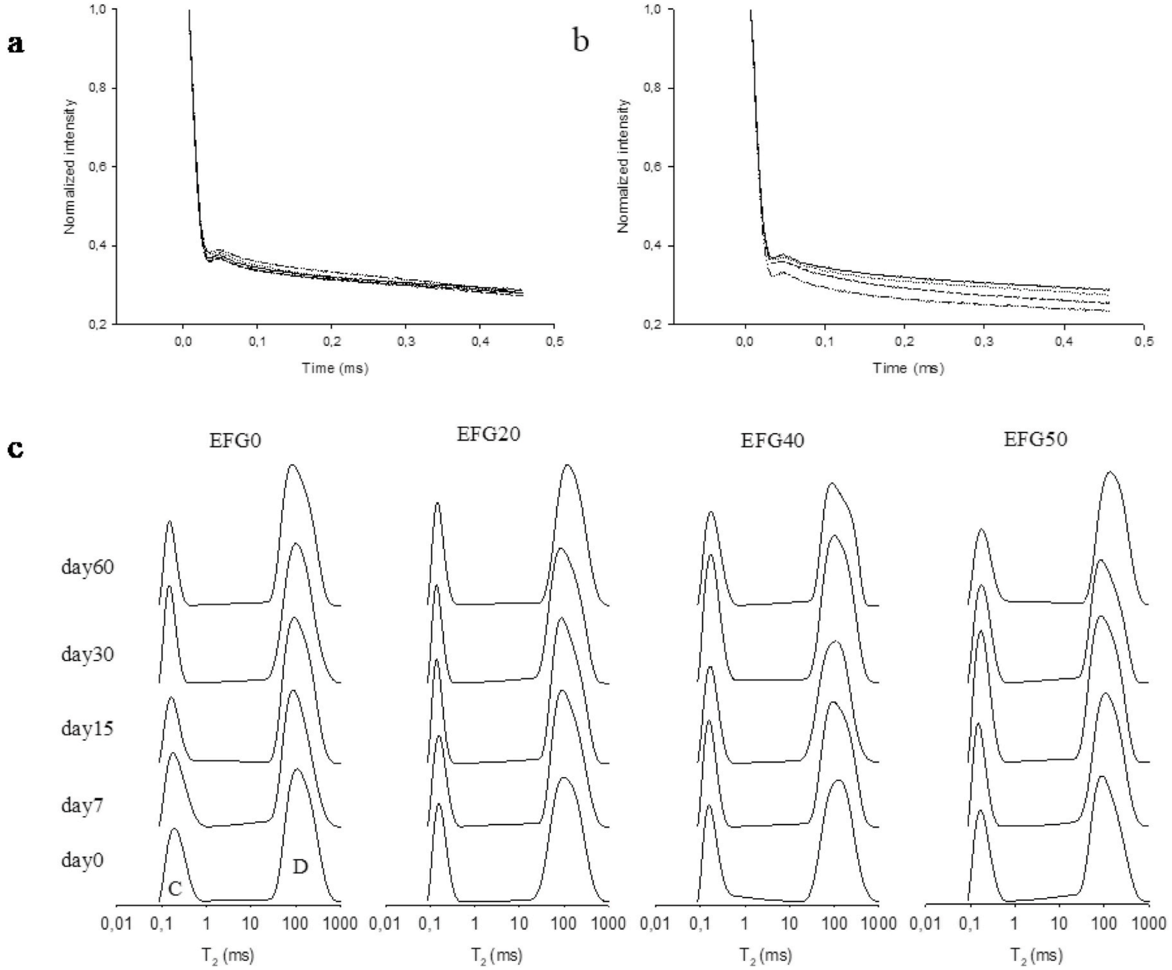

Fig. 3. ^{1}H FID relaxation times, a) comparison among different days of storage for the formulation EFG-0. b) comparison among different formulations at time 0. c) ^{1}H T$_2$ distributions in cookies with different fat substitution levels during 60 days of storage.
a)—— EFG0-t0; ········ EFG0-t7; ---- EFG0-t15; –·– EFG0-t30; — — EFG0-t60. b) —— EFG0-t0; ········ EFG20-t0; ---- EFG40-t0; –·– EFG50-t0. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG.

Table 7
Relaxation times (FID A, T_2C, T_2D) and relative abundances (FID PopA, PopC, PopD) of cookies formulations during storage.

	FID A (μsec)					FID PopA (%)				
	t 0	t7	t15	t30	t60	t 0	t7	T15	T30	T60
EFG0	15.52 ± 0.44 ab/A	15.41 ± 0.69 b/AB	15.16 ± 0.32 c/BC	15.13 ± 0.37 b/BC	14.94 ± 0.32 b/C	88.02 ± 1.52 a/A	84.18 ± 1.73 b/B	86.70 ± 2.79 a/AB	78.22 ± 9.00 b/C	84.66 ± 4.25 a/AB
EFG20	14.88 ± 1.92 b/A	15.67 ± 0.57 ab/A	15.64 ± 0.51 b/A	15.06 ± 0.30 b/A	15.19 ± 0.61 ab/A	85.50 ± 1.27 b/A	86.38 ± 0.59 a/A	86.27 ± 1.19 a/A	73.41 ± 5.92 c/C	82.27 ± 4.74 ab/B
EFG40	16.01 ± 0.34 a/A	15.76 ± 0.51 ab/A	15.92 ± 0.32 a/A	15.69 ± 0.21 a/AB	15.39 ± 0.60 a/B	84.64 ± 2.03 b/B	85.71 ± 0.44 a/A	85.82 ± 1.00 ab/A	85.46 ± 0.94 a/AB	85.34 ± 0.96 a/AB
EFG50	16.10 ± 0.30 a/A	15.88 ± 0.58 a/ABC	16.00 ± 0.30 a/AB	15.68 ± 0.48 a/BC	15.58 ± 0.68 a/C	85.69 ± 0.45 b/A	84.84 ± 0.65 b/A	84.88 ± 1.13 b/A	81.40 ± 4.31 ab/B	79.99 ± 7.63 b/B
	T_2C (msec)					**PopC (%)**				
	t0	t7	t15	t30	t60	t0	t7	t15	t30	t60
EFG0	0.16 ± 0.02 a/A	0.17 ± 0.01 a/A	0.16 ± 0.02 a/A	0.16 ± 0.02 a/A	0.17 ± 0.02 b/A	20.60 ± 5.41 c/B	26.39 ± 4.23 bc/A	17.54 ± 3.05 d/C	20.56 ± 2.53 c/B	18.95 ± 2.35 c/BC
EFG20	0.16 ± 0.01 a/AB	0.15 ± 0.01 b/C	0.15 ± 0.02 a/BC	0.15 ± 0.01 a/BC	0.17 ± 0.01 b/A	25.29 ± 2.86 b/A	24.28 ± 2.31 c/AB	22.54 ± 3.30 c/B	22.66 ± 3.12 b/B	23.14 ± 2.57 b/AB
EFG40	0.17 ± 0.01 a/A	0.16 ± 0.01 b/A	0.16 ± 0.02 a/A	0.16 ± 0.02 a/A	0.17 ± 0.02 b/A	33.42 ± 4.87 a/A	28.79 ± 2.64 b/BC	30.37 ± 3.92 b/B	28.95 ± 2.28 a/BC	26.60 ± 5.52 a/C
EFG50	0.16 ± 0.01 a/B	0.17 ± 0.02 a/B	0.16 ± 0.01 a/B	0.16 ± 0.02 a/B	0.19 ± 0.02 a/A	31.64 ± 2.72 a/AB	33.32 ± 5.24 a/A	32.82 ± 2.54 a/A	29.91 ± 2.47 a/BC	26.95 ± 1.70 a/C
	T_2D (msec)					**PopD (%)**				
	t0	t7	t15	t30	t60	t0	t7	t15	t30	t60
EFG0	98.14 ± 14.22 a/A	95.23 ± 8.35 b/A	88.47 ± 8.51 b/B	99.48 ± 4.00 a/A	97.14 ± 8.43 a/A	79.40 ± 5.41 a/B	74.72 ± 4.79 ab/C	82.47 ± 3.05 a/A	79.44 ± 2.53 a/B	81.05 ± 2.35 a/AB
EFG20	95.17 ± 9.89 a/AB	92.52 ± 7.94 b/AB	91.91 ± 6.54 b/B	93.10 ± 4.16 b/AB	100.22 ± 12.74 a/A	74.71 ± 2.86 b/B	75.72 ± 2.31 a/AB	77.46 ± 3.30 b/A	77.34 ± 3.12 a/A	76.86 ± 2.57 b/AB
EFG40	104.89 ± 14.89 a/AB	111.46 ± 12.32 a/A	100.55 ± 11.07 a/BC	91.97 ± 6.94 b/C	95.62 ± 10.86 a/BC	66.49 ± 4.24 c/C	71.35 ± 2.70 b/AB	69.12 ± 3.94 c/B	70.88 ± 2.26 b/AB	73.24 ± 5.67 c/A
EFG50	96.33 ± 13.16 a/BC	111.33 ± 14.44 a/A	94.55 ± 7.48 ab/BC	90.34 ± 4.34 b/C	105.03 ± 18.49a/AB	67.86 ± 2.42 c/BC	66.68 ± 5.24 c/C	66.86 ± 2.53 c/C	70.06 ± 0.58 b/B	72.54 ± 1.93 c/A

*Different small and capital letters indicate significant differences ($p \leq 0.05$) among formulations and storage time, respectively. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. t0, t7, t15, t30, t60, days of storage. FID, free induction decay; Pop, population.

moisture content carried by EFG, as revealed by the correlation found between FID A relaxation time and the cookies moisture content ($R = 0.631$, $p < 0.01$). Moreover, the correlation between FID A and Hardness ($R = 0.557$, $p < 0.05$) suggests a possible influence of the mobility of this class of protons on texture modification. Conversely, the relative abundance FID PopA (Table 7) didn't show a clear trend of modification neither for FORM nor for ST.

The evolution of ^1H T_2 distributions is reported in Fig. 3c. The presence of two protons populations C and D was evidenced in all the samples. C was the less abundant and more rigid population, encompassing ~17–33% of the total observed protons and relaxing at ~0.16–0.19 ms. D was the more abundant and mobile population relaxing in the ~88–111 ms range. In a previous work, Serial et al. (2016) found the presence of three ^1H T_2 populations in biscuits, relaxing at about 0.23, 5 and 80 ms, respectively; the population at intermediate mobility showed very low intensity. The differences respect to this study could be ascribable to different samples formulations, different software used for data fitting and different data acquisition temperature. The less mobile population was attributed to water protons tightly associated with CH protons of starch and gluten, while the more mobile population to protons belonging to margarine (Serial et al., 2016). In this study, while T_{2C} and T_{2D} did not relevantly change with respect to FORM or ST, the relative abundances of PopC and PopD (Table 7) resulted mainly influenced by FORM (SS% ~78 and ~80%, respectively). Indeed, for increasing level of EFG in the cookies, PopC increased while PopD dependently decreased (Table 7). These changes could be related to a decrease of the lipid phase in EFG-containing samples resulting in a decrease of PopD abundance. Moreover, Serial and colleagues studied the protons mobility of fiber containing biscuits with 2D-NMR; they hypothesized a relation between the increase of PopC abundance and the presence of fibre (Serial et al., 2016). These authors assumed that, being fibre able to retain water, it may affect the dough viscosity and in turn the biscuits spread and hardness. In this study both PopC and PopD resulted correlated to Hardness: while PopC resulted positively correlated ($R = 0.647$, $P < 0.01$), PopD showed a negative correlation ($R = -0.653$, $p < 0.01$). These results confirm the relation among the macroscopic structure of the final product and the interactions established at molecular level, involving water and fats with other biopolymers of the system.

3.5. Sensory evaluation of cookies during storage

Fig. 4 reports the results of the sensory scores assigned by the panellists to the shortbread cookies with different levels of fat substitution along 60 days of storage. Increasing fat substitution determined, as expected, decreased butter odour and aroma perception. On the other hand, this led to a slight increasing perception of toasted odour due to the formation of Maillard reaction products, as also observed from the colour analysis (Table 5). Looking at the structural features, the perceived porosity of the shortbread cookies decreased as EFG levels increased. This apparent discordance with the results of specific volume could be explained with different size distributions of the pores, as previously observed (Giarnetti et al., 2015). In fact, fat replacement with the EFG led to a larger number of smaller pores, as pointed out by image analysis (Giarnetti et al., 2015). This could be related to lower amounts of gases incorporated in the cookies when lower amounts of fatty shortenings were used (Forker, Zahn, & Rohm, 2012). It is reported that plastic fats, such as shortenings, are able to entrap and retain gases deriving from water vaporisation and from the decomposition of the used chemical leavening agents (Chevallier, Della Valle, Colonna, Broyart, & Trystram, 2002), while liquid oils are not as effective (Dapčević Hadnađev, Hadnađev, Pojić, Rakita & Krstonošić, 2015).

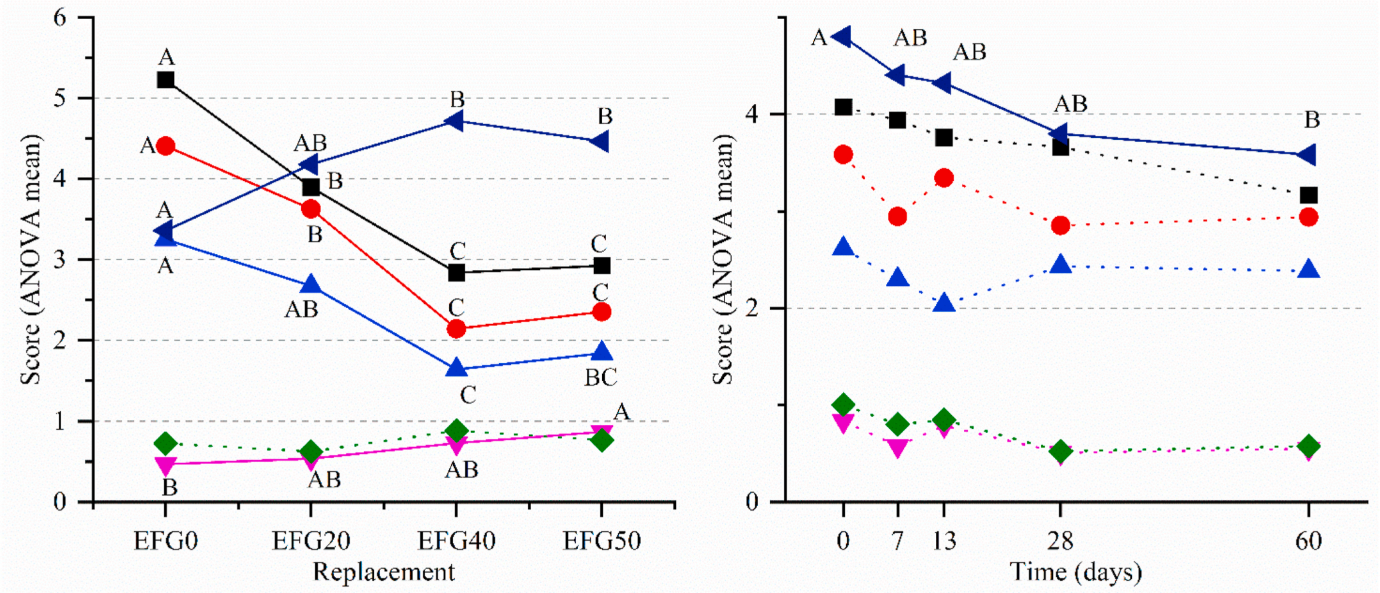

Fig. 4. Sensory scores assigned to cookies with different fat substitution levels (FORM) and during 60 days of storage (ST).
—■—Butter Odour —●— Butter Aroma —▲—Porosity —▼— Toasted Odour ···◆··· Toasted Aroma —◀—Hardness. Abbreviations: EFG, emulsion filled gel; EFG0, EFG20, EFG40, EFG50, cookies with 0, 20, 40, 50% butter substitution with EFG. Different letters indicate significant differences among mean scores $p < 0.05$. Dotted lines indicate no significant effect of the independent variable.

Moreover, water was hypothesized to favour a more uniform distribution of the leavening agent (Giarnetti et al., 2015). The higher hardness scores assigned to EFG40 and EFG50, in accordance with the texture results (Table 4), can also be explained by the gluten formation, that does not usually occur in shortbread doughs (Pareyt, Wilderjans, Goesaert, Brijs, & Delcour, 2008). It should not be neglected, moreover, that fat replacement led to a decrease of cookies spread and increase in their height, that has an obvious influence on the perception of breaking strength and, therefore, of hardness.

Sixty days of storage in controlled conditions induced slight sensory changes in cookies. Panellists reported a decrease of cookie hardness, as confirmed by the instrumental analysis (Table 4). A decrease of breaking strength, also measured by three-point bending tests, is usually reported in stored cookies, with or without fat replacement (Forker et al., 2012).

No rancid off-flavour was reported by the panellists up to 60 days of storage, in any of the cookie formulations (data not shown).

4. Conclusions

Shortbread cookies formulated by replacing butter with up to 50 g/100g EFG showed good stability during 60-days storage. Indeed, storage did not particularly affect the physicochemical properties of cookies, as instead the formulation did. Fats reduction, especially saturated ones, led to taller and narrower cookies with harder consistency, already at 40 g/100g replacement level. These structural changes were correlated with the thermal and ^{1}H-NMR parameters, which highlighted the key role of the state of lipids and water on the cookies' properties. The effect of storage was well observed only for texture, as also perceived by the panellists; it underwent slight softening, in relation to moisture absorption from the environment or internal water redistribution. The use of a packaging with high moisture barrier properties would further stabilize these products which, starting from 40 g/100g fat replacement, could be labelled for "reduced saturated fat content".

CRediT authorship contribution statement

Maria Paciulli: Conceptualization, Formal analysis, Validation, Data curation, Writing - original draft, revision. **Paola Littardi:** Formal analysis, Validation, Data curation, Writing - original draft, writing original draft. **Eleonora Carini:** Conceptualization, Visualization, Writing - review & editing. **Vito Michele Paradiso:** Conceptualization, Project administration, Writing - review & editing. **Maria Castellino:** Data curation, Formal analysis, Validation. **Emma Chiavaro:** Conceptualization, Project administration, Supervision, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

AACC, American Association of Cereal Chemists. (2000). *Approved methods of the AACC* (10th ed.). St. Paul, USA: Am Assoc Cereal Chem.

AOAC. (2000). *Official methods of analysis* (17th ed.). Gaithersburg, MD: AOAC INTERNATIONAL.

Barba, L., Arrighetti, G., & Calligaris, S. (2013). Crystallization and melting properties of extra virgin olive oil studied by synchrotron XRD and DSC. *European Journal of Lipid Science and Technology, 115*(3), 322–329. https://doi.org/10.1002/ejlt.201200259.

Caponio, F., Summo, C., Paradiso, V. M., Pasqualone, A., & Gomes, T. (2009). Evolution of the oxidative and hydrolytic degradation of biscuits' fatty fraction during storage. *Journal of the Science of Food and Agriculture, 89*(8), 1392–1396. https://doi.org/10.1002/jsfa.3600.

Cerretani, L., Bendini, A., Rinaldi, M., Paciulli, M., Vecchio, S., & Chiavaro, E. (2012). DSC evaluation of extra virgin olive oil stability under accelerated oxidative test: Effect of fatty acid composition and phenol contents. *Journal of Oleo Science, 61*(6), 303–309. https://doi.org/10.5650/jos.61.303.

Chevallier, S., Colonna, P., Valle, G. Della, & Lourdin, D. (2000). Contribution of major ingredients during baking of biscuit dough systems. *Journal of Cereal Science, 31*, 241–252. https://doi.org/10.1006/jcrs.2000.0308.

Chevallier, S., Della Valle, G., Colonna, P., Broyart, B., & Trystram, G. (2002). Structural and chemical modifications of short dough during baking. *Journal of Cereal Science, 35*, 1–10. https://doi.org/10.1006/jcrs.2001.0388.

Chiavaro, E., Vittadini, E., & Corradini, C. (2007). Physicochemical characterization and stability of inulin gels. *European Food Research and Technology, 225*(1), 85–94. https://doi.org/10.1007/s00217-006-0385-y.

Chiavaro, E., Vittadini, E., Rodriguez-Estrada, M. T., Cerretani, L., & Bendini, A. (2008). Monovarietal extra virgin olive oils. Correlation between thermal properties and chemical composition: Heating thermograms. *Journal of Agricultural and Food Chemistry, 56*(2), 496–501. https://doi.org/10.1021/jf072680w.

Chiu, S., Williams, P. T., & Krauss, R. M. (2017). Effects of a very high saturated fat diet on LDL particles in adults with atherogenic dyslipidemia: A randomized controlled trial. *PloS One, 12*(2), 1–14. https://doi.org/10.1371/journal.pone.0170664.

Chowdhury, R., Warnakula, S., Kunutsor, S., Crowe, F., Ward, H. A., Johnson, L., et al. (2014). Association of dietary, circulating, and supplement fatty acids with coronary risk: A systematic review and meta-analysis. *Annals of Internal Medicine, 160*(6), 398–406. https://doi.org/10.7326/M13-1788.

Chung, H., & Lim, S. (2006). Physical aging of amorphous starches (a review). *Starch - Stärke, 58*, 599–610. https://doi.org/10.1002/star.200600547.

Commission Internationale de l'Eclairage. (1978). *Recommendations on uniform color spaces, color-difference equations, psychometric color therms*. CIE.

Curti, E., Federici, E., Diantom, A., Carini, E., Pizzigalli, E., Wu Symon, V., et al. (2018). Structured emulsions as butter substitutes: Effects on physicochemical and sensory attributes of shortbread cookies. *Journal of the Science of Food and Agriculture, 98*(10), 3836–3842. https://doi.org/10.1002/jsfa.8899.

Dapčević Hadnađev, T., Hadnađev, M., Pojić, M., Rakita, S., & Krstonošić, V. (2015). Functionality of OSA starch stabilized emulsions as fat replacers in cookies. *Journal of Food Engineering, 167*, 133–138. https://doi.org/10.1016/j.jfoodeng.2015.02.002.

Derbyshire, W., Van Den Bosch, M., Van Dusschoten, D., MacNaughtan, W., Farhat, I. A., Hemminga, M. A., et al. (2004). Fitting of the beat pattern observed in NMR free-induction decay signals of concentrated carbohydrate-water solutions. *Journal of Magnetic Resonance, 168*(2), 278–283. https://doi.org/10.1016/j.jmr.2004.03.013.

Devi, A., & Khatkar, B. S. (2018). Effects of fatty acids composition and microstructure properties of fats and oils on textural properties of dough and cookie quality. *Journal of Food Science & Technology, 55*(1), 321–330. https://doi.org/10.1007/s13197-017-2942-8.

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 and Bioprocess Technology, 5*(6), 2497–2505. https://doi.org/10.1007/s11947-011-0536-4.

Giarnetti, M., Paradiso, V. M., Caponio, F., Summo, C., & Pasqualone, A. (2015). Fat replacement in shortbread cookies using an emulsion filled gel based on inulin and extra virgin olive oil. *Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology, 63*(1), 339–345. https://doi.org/10.1016/j.lwt.2015.03.063.

Kulp, K., Olewnik, M., Lorenz, K., & Collins, F. (1991). Starch functionality in cookie systems. *Starch - Stärke, 43*(2), 53–57. https://doi.org/10.1002/star.19910430205.

Laguna, L., Primo-martín, C., Varela, P., Salvador, A., & Sanz, T. (2014). LWT - food Science and Technology HPMC and inulin as fat replacers in biscuits : Sensory and instrumental evaluation. *Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology, 56*(2), 494–501. https://doi.org/10.1016/j.lwt.2013.12.025.

Mantzouridou, F., Spanou, A., & Kiosseoglou, V. (2012). An inulin-based dressing emulsion as a potential probiotic food carrier. *Food Research International, 46*(1), 260–269. https://doi.org/10.1016/j.foodres.2011.12.016.

Natalicchio, A., Piro, S., Gastaldelli, A., Targher, G., Laviola, L., Perseghin, G., et al. (2018). *Documento SID - Acidi grassi e diabete. SID - Società Italiana di Diabetologia*.

Nourbehesht, N., Shekarchizadeh, H., & Soltanizadeh, N. (2018). Investigation of stability, consistency, and oil oxidation of emulsion filled gel prepared by inulin and rice bran oil using ultrasonic radiation. *Ultrasonics Sonochemistry, 42*, 585–593. https://doi.org/10.1016/j.ultsonch.2017.12.029. October 2017.

Paciulli, M., Rinaldi, M., Cavazza, A., Ganino, T., Rodolfi, M., Chiancone, B., et al. (2018). Effect of chestnut flour supplementation on physico-chemical properties and oxidative stability of gluten-free biscuits during storage. *Lebensmittel-Wissenschaft & Technologie, 98*, 451–457. https://doi.org/10.1016/j.lwt.2018.09.002. September.

Paradiso, V. M., Giarnetti, M., Summo, C., Pasqualone, A., Minervini, F., & Caponio, F. (2015). Production and characterization of emulsion filled gels based on inulin and extra virgin olive oil. *Food Hydrocolloids, 45*, 30–40. https://doi.org/10.1016/j.foodhyd.2014.10.027.

Pareyt, B., Wilderjans, E., Goesaert, H., Brijs, K., & Delcour, J. A. (2008). The role of gluten in a sugar-snap cookie system: A model approach based on gluten-starch blends. *Journal of Cereal Science, 48*(3), 863–869. https://doi.org/10.1016/j.jcs.2008.06.011.

Parks, E., Yki-Järvinen, H., & Hawkins, M. (2017). Out of the frying pan: Dietary saturated fat influences nonalcoholic fatty liver disease. *Journal of Clinical Investigation, 127*(2), 454–456. https://doi.org/10.1172/JCI92407.

Patel, A. R., & Dewettinck, K. (2016). Edible oil structuring: An overview and recent updates. *Food & Function, 7*(1), 20–29. https://doi.org/10.1039/C5FO01006C.

Pintado, T., Herrero, A. M., Jiménez-Colmenero, F., Pasqualin Cavalheiro, C., & Ruiz-Capillas, C. (2018). Chia and oat emulsion gels as new animal fat replacers and healthy bioactive sources in fresh sausage formulation. *Meat Science, 135*, 6–13. https://doi.org/10.1016/j.meatsci.2017.08.004. November.

Pintado, T., Herrero, A. M., Ruiz-Capillas, C., Triki, M., Carmona, P., & Jiménez-Colmenero, F. (2016). Effects of emulsion gels containing bioactive compounds on sensorial, technological, and structural properties of frankfurters. *Food Science and Technology International, 22*(2), 132–145. https://doi.org/10.1177/1082013215577033.

Regulation (EC) No 1924/2006. (2006). *On nutrition and health claims made on foods*.

Rodríguez-García, J., Laguna, L., Puig, A., Salvador, A., & Hernando, I. (2013). Effect of fat replacement by inulin on textural and structural properties of short dough biscuits. *Food and Bioprocess Technology, 6*(10), 2739–2750. https://doi.org/10.1007/s11947-012-0919-1.

Serial, M. R., Blanco Canalis, M. S., Carpinella, M., Valentinuzzi, M. C., León, A. E., Ribotta, P. D., et al. (2016). Influence of the incorporation of fibers in biscuit dough on proton mobility characterized by time domain NMR. *Food Chemistry, 192*, 950–957. https://doi.org/10.1016/j.foodchem.2015.07.101.

Shoaib, M., Shehzad, A., Omar, M., Rakha, A., Raza, H., Sharif, H. R., et al. (2016). Inulin: Properties, health benefits and food applications. *Carbohydrate Polymers, 147*, 444–454. https://doi.org/10.1016/j.carbpol.2016.04.020.

Shogren, R. L. (1992). *Effect of moisture content on the melting and subsequent physical aging of cornstarch* (Vol. 19, pp. 83–90).

Slade, L., & Levine, H. (1994). Structure-function relationships of cookie and cracker ingredients. *The Science of Cookie and Cracker Production, 9*, 23–141.

de Souza, R. J., Mente, A., Maroleanu, A., Cozma, A. I., Ha, V., Kishibe, T., et al. (2015). Intake of saturated and trans unsaturated fatty acids and risk of all cause mortality, cardiovascular disease, and type 2 diabetes: Systematic review and meta-analysis of observational studies. *BMJ, h3978*. https://doi.org/10.1136/bmj.h3978.

World Health Organization. (2015). *Fact sheet No 394*. Healthy Diet Issue September. Available from: http://who.int/mediacentre/factsheets/fs394/en/ (Last accessed on 2015 Sep 16).