



Potential application of pomegranate seed oil oleogels based on monoglycerides, beeswax and propolis wax as partial substitutes of palm oil in functional chocolate spread



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ABSTRACT

In this research, the effectiveness of pomegranate seed oil oleogel as a partial replacement of fat phase in chocolate spread was studied. Monoglyceride (MG), beeswax (BW) and propolis wax (PW) have been used as structuring agents at 5 g/100 g concentration to gel pomegranate seed oil. The oleogels were then combined with palm oil at 1:1 ratio. Different techniques, including polarized light microscopy, synchrotron XRD, mechanical analyses, and oil binding capacity were used to study the physical and mechanical properties of the palm oil-oleogel systems and chocolate spreads. Results highlighted that MG, BW and PW chemical nature led to the formation of different crystalline network in palm oil-oleogel systems. Chocolate spreads containing palm oil-oleogels showed an increase in the mechanical parameters in the order of PW < BW < MG. This trend might be attributed to the chemical composition of oleogelators and physical bonds formed in the samples. During storage, crystal transformation in MG and structural reorganization in waxes (PW and BW) samples showed a gradual decrease and increase in hardness, respectively. These findings could provide useful information in the application of pomegranate seed oil oleogel for novel confectionery products engineering.

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1. Introduction

The health benefits associated with the consumption of polyunsaturated fatty acid (PUFAs) and especially conjugated linoleic acid (CLA) and conjugated linolenic acid (CLN) are well demonstrated in the literature (Chojnacka et al., 2016). One of the most important oils containing conjugated fatty acids is pomegranate seed oil (PSO). The specific trienoic fatty acid found in PSO is punicic acid (PA) which is a polyunsaturated fatty acid (18:3) also called 9-cis, 11-trans, 13-cis, octadecatrienoic acid. PA is referred as a “super CLA” whose effect is even higher than that of an ordinary CLN (Aruna, Venkataramanamma, Singh, & Singh, 2016). This unique conjugated fatty acid presents several potential health benefits such as cholesterol lowering, antidiabetic, anti-inflammatory and

anticarcinogenic properties (Kýralan, Gölükçü, & Tokgöz, 2009).

The introduction of PSO, and thus PA, in fat-based foods as substitute (partial or total) of saturated fats could allow the development of new products with improved health properties, not only thanks to the presence of essential fatty acids but also by reducing the total level of saturated fats. As well known, consumption of saturated fatty acids at higher amount leads to negative health implications, including obesity, cardiovascular diseases (CVD), high cholesterol, cancer and type II diabetes (Micha & Mozaffarian, 2010).

Chocolate spreads could be a good candidate for the enrichment with PSO accompanied with the reduction of saturated fats in the formulation. Chocolate spread is a suspension of solid particles embedded into a fat crystal network (i.e. higher than 40 g/100 g) composed of saturated fats such as palm oil, coconut oil and cocoa butter. This product is widely used directly by consumers as delicious confectionary product or by the food industry as filling ingredient in other formulations such as biscuits and cakes

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(Manzocco, Calligaris, Camerin, Pizzale, & Nicoli, 2014). The sensory performances of chocolate spreads are strictly related to the presence of a fat crystal network providing texture, mouthfeel and flavor to the product (Marangoni et al., 2012). Thus, the partial substitution of solid hardstock fat with liquid oil could greatly affect the chocolate spread performances and thus the product quality (Anese et al., 2016; Doan et al., 2016). An emerging strategy is to substitute solid hardstock fat rich in saturated fats with unsaturated oils solidified thanks to molecules forming self-assembly networks. These systems are called oleogels that are self-standing, thermoreversible, anhydrous and viscoelastic materials structured by a three-dimensional supramolecular network of self-assembled molecules with limited solubility in an organic liquid (Co & Marangoni, 2012; Patel & Dewettinck, 2016). A wide number of different oleogelators has been proposed in the literature to gel oils. Among others, natural waxes and saturated monoglycerides have been indicated as particularly promising for food applications (Da Pieve, Calligaris, Co, Nicoli, & Marangoni, 2010; Doan, Van de Walle, Dewettinck, & Patel, 2015; Ögütçü & Yilmaz, 2015). Waxes deriving from different natural sources, such as candelilla wax, carnauba wax, rice bran wax, beeswax and propolis wax, contain long-chain fatty acid esters able to crystallize forming a three-dimensional network entrapping liquid oil (Doan et al., 2015; Fayaz, Goli, Kadivar, et al., 2017). Similarly, monoglycerides (MGs) are able to self-assemble into inverse bilayer nanostructures organized at micro-level into lamellar platelets that finally interact immobilizing liquid oil (Da Pieve et al., 2010).

Recently, the application of oleogels in food products for the reduction of saturated fatty acids as well as the delivery of essential polyunsaturated fatty acids has been studied in ice cream (Zulim Botega, Marangoni, Smith, & Goff, 2013), chocolate containing products (Doan et al., 2016), bakery products (Anese et al., 2016; Patel et al., 2014; Stortz & Marangoni, 2013), frankfurters (Zetzl, Marangoni, & Barbut, 2012) and margarine (Hwang et al., 2013). Results demonstrated that the novel structural approach could be a pursuable strategy even though a careful re-examination of formulation and processing conditions should be done. In the development of the new functional products, the knowledge of the structural behavior of the oleogel in the formulation is fundamental to obtain a product with adequate textural and sensory properties.

Based on these considerations, the aim of this research was to investigate the application of pomegranate seed oil oleogels as co-structurants with palm oil (PO) in chocolate spreads to obtain a functional food enriched with PSO with a reduced saturated fat content. To this purpose, PSO oleogels containing 5 g/100 g of saturated monoglyceride, beeswax and propolis wax were considered as partial replacers of palm oil (50% of replacement) in chocolate spreads. It should be noted that the oil gelling properties of these three structurants at the selected concentration have been already demonstrated by different authors (Da Pieve et al., 2010; Fayaz, Goli, & Kadivar, 2017; Fayaz, Goli, Kadivar, et al., 2017; Yilmaz & Ögütçü, 2014).

The PO-oleogel mixtures as well as chocolate spreads containing these mixtures were characterized by using different techniques, including polarized light microscopy, synchrotron X-ray diffraction, mechanical and rheological analyses and oil binding capacity.

2. Materials and methods

2.1. Materials

Myverol™ saturated monoglyceride (MG) (fatty acid composition: 1.4% C_{14:0}, 59.8% C_{16:0}, 38.8% C_{18:0}; melting point 68.05 ± 0.5 °C) was from Kerry Bioscience (Bristol, United Kingdom), beeswax (BW) and propolis was from Espadana

Mokamel Co. (Isfahan, Iran), pomegranate seed oil (PSO) from Dastchinali Co. (Isfahan, Iran) and palm oil (PO) (saturated fatty acid 48.45% w/w; melting point 26.89 ± 0.10 °C) was from Unigrà (Conselice, Italy). Sugar and defatted cocoa powder were purchased in a local market. Propolis wax (PW) was extracted from propolis according to Fayaz, Goli, and Kadivar (2017) procedure.

2.2. Oleogel preparation

The oleogelators were dispersed in PSO at a concentration of 5 g/100 g. The mixture was heated at 80 °C under magnetic stirring in a temperature controlled water bath. Just after oleogelator melting, the mixtures were maintained at 80 °C for at least 10 min and subsequently quiescently cooled at 20 °C to allow gel formation. Samples were stored at 20 °C for 24 h before analysis.

2.3. Oleogel-palm oil mixture preparation

The oleogels were mixed with PO (1:1 w/w) at 80 °C in a temperature controlled water bath under magnetic stirring until the melting of all components and then cooled and stored at 20 °C for 24 h before analysis and usage in chocolate spreads.

2.4. Chocolate spread preparation

The chocolate spreads were prepared according to the methodology reported by Manzocco et al. (2014) and Doan et al. (2016), with minor modifications. In particular, the samples consisted of 40 g/100 g fat, 50 g/100 g sugar with fineness of 0.25 mm or lower (sugar grounded and sifted with a 60-mesh sieve) and 10 g/100 g cocoa powder. Fat phase (oleogel-palm oil mixture) was heated at 80 °C until complete melting in a temperature controlled water bath, then dry ingredients were dispersed in the molten fat phase and manually stirred with a spatula for 2 min until a homogeneous paste-like spread was obtained while maintaining temperature at 80 °C. Chocolate spreads were cooled to 20 °C. Analyses were carried out 24 h after preparation and during storage at 20 °C.

2.5. Analytical determinations

2.5.1. Polarized light microscopy

The microstructure of oleogels and OG-PO mixtures were studied using a polarized light (PL) optical microscope (Leica DM 2000; Leica Microsystems, Heerburg, Switzerland) connected with a Leica EC3 digital camera (Leica Microsystems). One drop of sample was placed in the middle of a glass slide and a glass cover slip was centered above the drop. The samples were analyzed at 20 °C using a 200 × magnification. Micrographs were acquired and processed using the application software Leica Suite LAS EZ (Leica Microsystems).

2.5.2. Synchrotron XRD analysis

Synchrotron X-ray diffraction patterns were recorded at the X-ray diffraction beam-line 5.2 of the Synchrotron Radiation Facility Elettra in Trieste (Italy). The X-ray beam emitted by the wiggler source on the Elettra 2 GeV electron storage ring was monochromatized by a Si (111) double crystal monochromator, collimated by a double set of slits giving a spot size of 0.2 × 0.2 mm. A drop of sample was lodged into a nylon pre-mounted cryoloop 20 µm (0.7–1.0 mm) (Hampton Research HR4-965, AlisoVejito, CA, USA). Analyses were performed at 20 °C controlling the temperature by a 700 series cryocooler (Oxford Cryosystems, Oxford, UK). Data were collected at a photon energy of 8.856 keV ($\lambda = 1.4 \text{ \AA}$), using a 2 M Pilatus silicon pixel X-ray detector (DECTRIS Ltd., Baden, Switzerland). Bidimensional patterns collected with Pilatus were

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