Critical Reviews in Food Science and Nutrition

ISSN: 1040-8398 (Print) 1549-7852 (Online) Journal homepage: http://www.tandfonline.com/loi/bfsn20

Oleogels, a promising structured oil for decreasing saturated fatty acid concentrations: Production and food-based applications

Halime Pehlivanoğlu, Mehmet Demirci , Omer Said Toker , Nevzat Konar, Salih Karasu & Osman Sagdic

CRITICAL REVIEWS IN FOOD SCIENCE AND NUTRITION
http://dx.doi.org/10.1080/10408398.2016.1256866

Taylor & Francis
Taylor & Francis Group

Check for updates

Oleogels, a promising structured oil for decreasing saturated fatty acid concentrations: Production and food-based applications

Halime Pehlivanoğlu[a], Mehmet Demirci [a], Omer Said Toker [b], Nevzat Konar[c], Salih Karasu [b], and Osman Sagdic [b]

[a]Food Engineering Department, İstanbul Sabahattin Zaim University, Istanbul, Turkey; [b]Food Engineering Department, Chemical and Metallurgical Engineering Faculty, Yıldız Technical University, İstanbul, Turkey; [c]Food Engineering Department, Engineering and Architecture Faculty, Siirt University, Siirt, Turkey

ABSTRACT

Oils and fats are widely used in the food formulations in order to improve nutritional and some quality characteristics of food products. Solid fats produced from oils by hydrogenization, interesterification, and fractionation processes are widely used in different foodstuffs for these aims. In recent years, consumer awareness of relation between diet and health has increased which can cause worry about solid fat including products in terms of their high saturated fatty acid and trans fatty acid contents. Therefore, different attempts have been carried out to find alternative ways to produce solid fat with low saturated fatty acid content. One of the promising ways is using oleogels, structuring oils with oleogelators. In this review, history, raw materials and production methods of the oleogels and their functions in oleogel quality were mentioned. Moreover, studies related with oleogel usage in different products were summarized and positive and negative aspects of oleogel were also mentioned. Considering the results of the related studies, it can be concluded that oleogels can be used in the formulation of bakery products, breakfast spreads, margarines, chocolates and chocolate-derived products and some of the meat products.

KEYWORDS
Oleogel; production; applications; food; saturated fatty acid

Introduction

Fats and oils are indispensible part of our diet due to the several reasons like being energy source, solvent for some valuable nutrients (vitamins and vitamin precursors) and bioactive compounds and flavor carriers. Fats bring processed foods in flavor, mouth feel, palatability, texture and aroma. Vaclavik and Christian (2014) stated the functions of fat as follows: improving or modification of flavor and texture, leavening batters and doughs, contributing flakiness and tenderness, emulsification, transferring of heat such as in frying, prevent sticking and providing satiety. Therefore, fats play an important role in production of the product with desired quality, determining attractiveness of the corresponding food material. After understanding the technological importance in the food industry, researchers and research development experts in the industry have tended towards to investigate the ways for the cheaply production of solid fats with vegetable oil. Different methods have been improved so far. Hydrogenization, interesterification and fractionation methods are widely used for transforming oils to fats. The fabricated solid fats are widely used as margarine and breakfast spreads in our breakfast, bakery products, chocolate products etc. Usage of these solid fats in food products give consumers, aware of relation between diet and health, worry in two aspects. The former is associated with higher saturated fatty acid content of such solid fats. Consumption of saturated fatty acids at higher amount increases the risk of cardiovascular diseases, obesity and diabetes (Muguerza et al., 2002). Therefore, it is suggested by The World Health Organization (WHO) that 10% of required energy is supplied from saturated fats (World Health Organization, 2002). The latter main problem is resulted from trans fatty acids content, formed as a result of interesterification process, which have adverse effects on coronary heart disease, cancer, diabetes and blood lipoprotein profiles (Mozaffarain et al., 2009; Uauy et al., 2009; Brouwer et al., 2010). Regarding those adverse effects of both saturated and trans fatty acids, efforts have been exerted to decrease their content for a long time in the food industry.

One of the up-and-coming ways to decrease saturated fatty acid content is using oleogels in the formulations. Oloegels are the structured oils prepared by oleogelation of liquid oil using oleogelators such as vegetable waxes, monodiglycerides, alcohols or esters of fatty acids, phospholipids and phytosterols (Pérez-Monterroza et al., 2014). The findings of oleogel-related studies showed that oleogel can be used in the formulations of cake, cookies, meat products, chocolate and ice cream to decrease saturated fatty acid contents. However, further studies are needed to obtain detailed information in terms of usage possibilities of them. According to results of previous studies, the quality characteristics of the oleogel-including samples have been found as at acceptable limits. Oleogel can be also used to eliminate some quality defects such as preventing or decreasing fat migration substantial for fat bloom and

CONTACT Halime Pehlivanoğlu halime.pehlivanoglu@izu.edu.tr Food Engineering Department, İstanbul Sabahattin Zaim University, Istanbul, Turkey.
Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/bfsn.

increasing melting temperature of the chocolate or chocolate-derived products as well as reducing saturated fatty acid content of the corresponding products (Hughes et al., 2009).

By considering those positive impacts of oleogels on the quality of food products, it seems as promising ingredient which could have industrial applications in the future. Therefore, in the present study, we reviewed the oleogel in the following aspects: (i) history of oleogel, (ii) production methods of oleogel, and (iii) applications of oleogel. This review has potential in directing of future studies related with oleogel and applications of them in the food industry. Understanding the functions of raw materials used in the oleogel and production steps in the quality of oleogels can enable to fabricate the healthier products with desired quality and with lower cost, which is very important in food industry.

History, production and applications

Why oleogel?

For children %35-40, for young people %30–35 and for adults %25–40 of their daily calorie need is supplied by fats in developed countries; but this rate is almost %5 in undeveloped countries (Başoğlu, 2014). Both FAO and WHO reported that %15–30 of total calorie needs to be taken by fats and also significant amount of these fats should be vegetable liquid oils. Additionally, Demirci (2003) indicated that consumption of saturated fatty acid should not exceed % 10 of total calorie in a diet in terms of healthy life. When considering the human diet, consumption of the fast and snacks foods like cakes, chocolates, biscuits, pastries etc. have been increased in a recent years. Saturated fats have important role in improving of the some quality attributes such as mouth feel and textural properties. Formulation of the vegetable oil in these product have attracted due to their nutritional and health benefit, and economic advantage. On the other hand, using of the unsaturated oils instead of saturated fats in these type products cause technical problems like textural weakness and oil leakage. Therefore saturated fat produced from vegetable source has gain importance recently (Stortz et al., 2012).

Solid like lipids is preferred in a processing of some industrial products like pasta, patty and cake due to their some specific characteristics such as better oxidative stability, solid lipid functionality and alternative usage to butter. Because such characteristics affect the texture, spreadability, snap, the shelf life, and flavor of the final product, applications of solid lipid in food industry are important. As known there are widely used three methods to transform vegetable oils into solid fat due to economical purposes and technological function of solid fats in the food industry. These are hydrogenation, interesterefication and fractionation (Başoğlu, 2014).

Hydrogenation process is divided into three groups which include full hydrogenation (full saturation of whole double bounds in fatty acid chains), partial hydrogenation (to set melting point of fats below body temperature), and selective hydrogenation (reacting fatty acids according to their double bound number from 3 double bounds to 1 double bound). Hydrogenation includes three reactions which are saturation of double bonds between carbons, changing *cis* geometric isomers into *trans* formation and the formation of new positional isomers in fatty acid chains. Melting point of fats increase by saturation and isomerization (*cis* to *trans*). In cooling hydrogenated fats below their melting point will pave the way for an initial effect to crystallization of the material. Final semisolid fat system has trapping effect for liquid oil (Narine and Marangoni, 1999a; Marangoni, 2000; Marangoni and Rogers, 2003). Interestereficaton technique based on that replacement of fatty acid radicals as intermolecular or intramolecular in glyceride molecules without any isomer transform. Interestereficationhas two types, these are enzymatic and chemical interestereficaton. Fractioning process is separated components of liquid or solid oils which have high melting points using low melting points and has three types; chromatography, friction crystallization and liquid/liquid extraction (Başoğlu, 2014).

Although saturated and trans fats have positive effects on the quality properties of foods, they have negative attributes to people's health (Mensink et al., 2003). Excessive consumption of saturated fats can increase obesity risk as well as cardiovascular health problems (American Heart Association Statistics Committee and Stroke Statistics Subcommittee, 2012). While these fatty acids increase the amount of LDL cholesterol, on the other hand decrease the amount of HDL cholesterol in metabolism (Stauffer, 1996; O'Brien, 2004; Vandana et al., 2011). Intake of high amounts of trans and saturated fat has negative effect on metabolic order (Riccardi et al., 2004; Micha and Mozaffarian, 2009; Lakmali et al., 2011; Ambrosini, 2014).

Another known method to transform liquid oils into solid fats is by either adding saturated fat or *trans* fats. Most significant disadvantage of this method is that the end product contains high amount of saturated or *trans* fats (Marangoni and Garti, 2011). When considering the fact that many food products include solid fats fabricated by one of such methods, those widely consumed products are rich in saturated fatty acids and some of them include trans fatty acids depending on the fat type in the formulation and production methods of the corresponding product. Therefore, the development of novel methods to structure oil with different ways to fabricate solid fats with low saturated fatty acids and no *trans* fatty acids has drawn attention. That is the reason of why oleogels are promising subject in recent years. Nowadays, oleogels have been studied to determine possible alternative usages of them in the food industry.

What is oleogel and its history?

Organogelation or oleogelation is a novel definition for forming liquid oils into a gel-like structure. In other words, oleogelation which recently has been of great interest in many areas like pharmaceutic, food, cosmetic and petro chemistry etc. (Terech and Weiss, 1997; Dassanayake et al., 2011; Marangoni and Garti, 2011) is transformation of a liquid oils into a gel like structure which has features (rheological properties, viscoelasticity, spreadibility, and firmness etc.) of a solid fat without containing large amount of saturated fats (Rogers et al., 2009; Botege, 2012; Co and Marangoni, 2012; Stortz et al., 2012; Patel et al., 2014a). The gels fabricated as a result of oleogelation are called as oleogels.

Oleogels can be described as a complex microstructured system where an organic liquid is entrapped within a thermo-reversible, 3-dimensional gel-network (Hinze et al., 1996;

Sánchez et al., 2008; Stortz et al., 2014). This system provides to obtain oleogels with solid-like properties (Patel, 2015) by ability of entrapping liquid oils (Van Esch and Feringa, 2000; Marangoni, 2004) in a three-dimensional, thermo-reversible gel network with a gelling agent (Marangoni, 2012).

After brief description of oleogels, history of oleogels was mentioned in this section. Historical development of oleogels is based on the term of gel defined by Thomas Graham in 1861. After 65 years, Jordan Lloyd made a definition 'the colloid condition or 'gel', and now that is also widely used. According to Lloyd's gel definition, all gels must be have at least two major components, a liquid phase and a gelling agent and that system has similar mechanical properties with a solid. After Lloyd, Herman and Ferry developed the definition but major feature like two components-including system (liquid phase and gelling agent) was also the same (Weiss, 2006; Marangoni and Garti, 2011). Hermans offered that gels display mechanical properties of a solid like material and both phase of system (dispersion and dispersed phase) must extend throughout the whole system. On the other hand, Ferry came up with an illustrative definition of a gel which was a system showing unstable flow.

Emergence of the importance of the solid fats in the food industry has induced researchers to seek alternative ways to produce solid phase from liquid. For the food industry, structuring liquids into solid-like materials were successfully used since 1903 when Wilhelm Norman produced a solid fat using liquid animal fat by hydrogenation. After that in a company, designed by Norman, where solid fats were fabricated by using whale liquid oils. But first production of solid fat from vegetable based liquid oil dated to be 1911, which was accepted as first description for shortening. An American company named by Procter and Gamble achieved solid like oils by using cotton oil with hydrogenation technology (Marangoni and Garti, 2011). Between 1920 and 1940 many researches had been carried out to produce margarines and shortenings with desired properties such as melting point, softness and easy machinability (Başoğlu, 2014). Thus hydrogenation technology-based products resulted in new approaches for the production of structured special products containing vegetable oils (O'Brien, 2003). Hydrogenated oils are also widely produced in our times for fabrication of the different products such as margarine, shortenings, coating fats and frying oils (Başoğlu, 2014).

Application of the hydrogenation caused some health problem related to cardiovascular diseases due to high consumption of the saturated and trans fatty acid although it overcome technical problems related to using of the vegetable oil in some food products. Formulated of the animal and vegetable saturated fat was not recommended by American Heart Association. For this reason, new methods in structuring of the vegetable oil were required. Scientists have focused on new techniques to develop unsaturated fat alternatives for overcoming of health and technical problems. From these techniques, oleogelation have gained importance because liquid oil is converted into gel like structure without changing of the chemical properties of the vegetable oil by oleogelation. A few studies related to oleogels were performed by the end of the nineties. Interest in oleogels in food application has increased for last ten years (since 2006).

Oleogel production methods

Oloegels are formed with three different systems which are (i) crystalline particles and self-assembled structures of low molecular weight, (ii) self-assembled structures of polymers or polymeric strands, and (iii) miscellaneous (Gronwald et al., 2002; Pernetti et al., 2007a; Bot et al., 2009; Hughes et al., 2009; Co and Marangoni, 2012; Patel, 2015).

At first formation, gelation occurs by trapping the liquid oil phase into triacylglyserol (TAG) particles (Marangoni, 2004; Bot et al., 2007). Also diacylglycerols (DAG), monoacylglycerols (MAG), and fatty acids are capable of formation of similar structures like triacylglyserol (TAG) (Ojijo et al., 2004; Wright and Marangoni, 2006; Pernetti et al., 2007b; Calligaris et al., 2010; Da Pieve et al., 2010; Lupi et al., 2012; Patel, 2015). Self-assembled fibrous networks (SAFIN) occur by using gelling agent that has low molecular weight (Abdallah et al., 2002) like phytosterols (oryzanol, 12-hydroxystearic acid (Rogers et al., 2008) and ricinoleic acid (Wright and Marangoni, 2006). Helical and twisted crystalline ribbons are obtained by this system (Marangoni and Garti, 2011; Sahoo et al., 2011; Patel, 2015).

In the second method, gelation is formed by self-assembled structures which are using polymer or polymeric strands like ethyl cellulose (Marangoni and Garti, 2011; Patel, 2015). Ethylcellulose which is a hydrophobic cellulose, is chemically derivatived from cellulose. Polysaccharides and proteins which also hydrophobic molecules could be also used for gelling agent (Guenet, 2008; Patel, 2015). The last method where some researchers used inorganic particles to achieve gel network (Eitel, 1975). Patel et al. (2015) used fumed silica for gelling of sunflower oil.

General production methods of oleogel, as summarized in Fig. 1, include melting of oleogelators, heating oils to melting temperature of oleogelators, mixing of oleogelator and oil and cooling of the formed gels.

As known, for formation of oleogels two materials are required. One of them is oil and the other one is oleogelator used for gelation purposes. In this part of the review, the information will be given about oils and oleogelators used in the oleogel production and their importance in oleogel characteristics.

Oils

Oil type is important factor affecting rheological, textural properties and thermal properties of the oleogels in addition to oleogelator type and concentration. Various type of vegetable oil such as, sun flower oil, corn oil, olive oil, canola oil, and hazelnut oil can be used to prepare oleogels (Ogutcu et al., 2015; Öğütcü and Yılmaz, 2015a; Patel and Dewettinck, 2015; Jang et al., 2015).

Effect of the oil type on the rheological, textural, and thermal properties of the oleogels was reported by Patel (2015). Five different oil types (rape seed oil, rice bran oil, corn oil, sun flower oil, and high oleic sun flower oil), which has a different saturated and unsaturated fatty acid levels, were selected to observe the influence of fatty acid composition on the gelling ability of the wax. It was reported that minimum gelling concentration was significantly affected from saturated and unsaturated levels of the oils and lower gelling concentrations were obtained when vegetable oils with higher proportion of

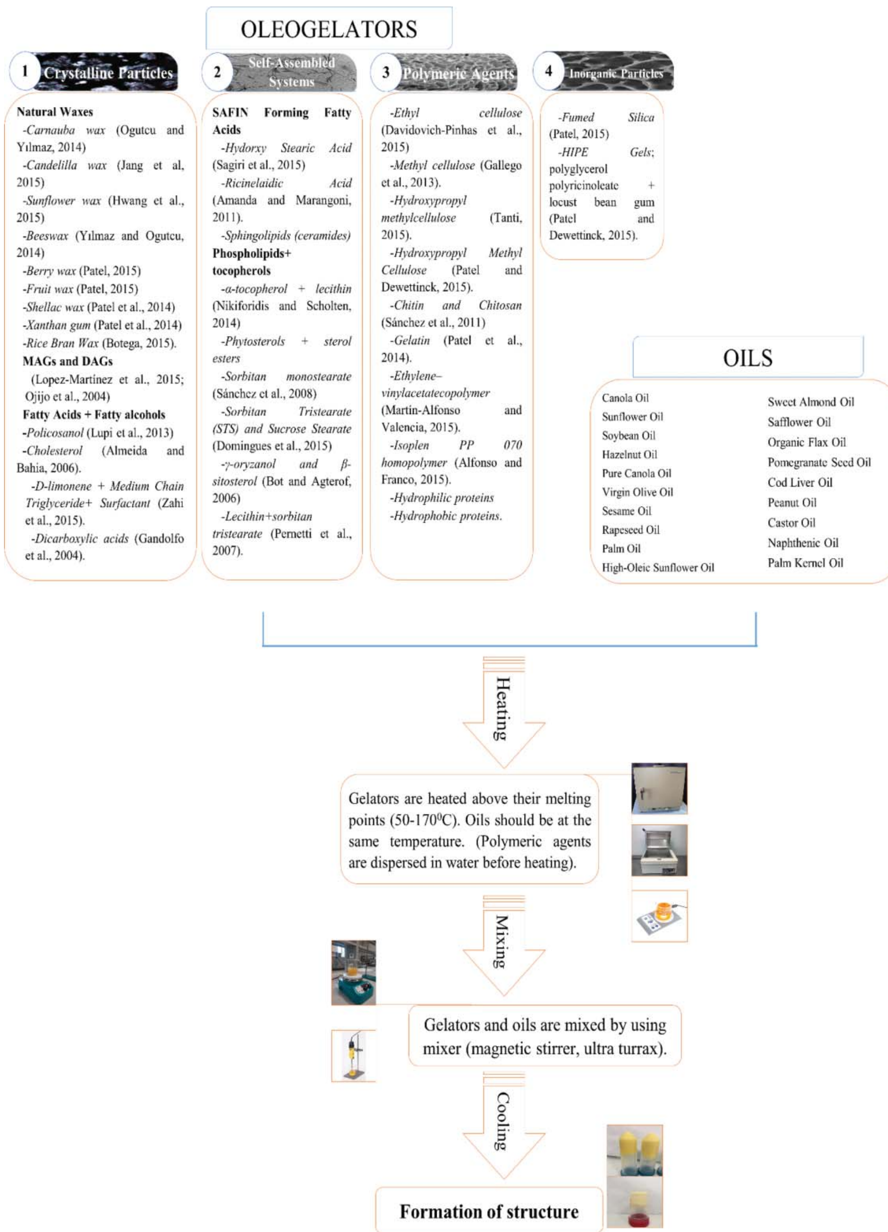

Figure 1. General production process of the oleogels.

saturated fatty acid (SAFA) were used. This result was linked to different reasons;

1. The higher level of SAFA content (consequently higher rate of high melting TAGs) gives rise to the strengthening of the oleogels structure,
2. The lower level of low melting TAGs depletes the solvency effect of liquid oils forcing the formation of higher crystalline mass of wax.

It was also stated that thermal characteristics of the edible oil were affected by the degree of the oil saturation. They found that minimum gelling concentration of the bees wax was significantly lower in rice bran oil (highest saturated level) when a compared to rape seed oil (highest unsaturated level). They also observed that significantly higher latent heat for crystallization of bees wax was found in rice bran oil when a compared to rapeseed oil. It was confirmed that the higher crystalline mass of the wax was formed in rice bran oil when a compared to rape seed oil at same concentration of bees wax.

Similar results were also found by Dassanayake et al. (2012). They investigated effect of oil type on the melting and crystallization temperature of rice bran wax and, viscosity and texture properties of oleogels prepared by different oil types and rice bran wax concentration (1%, 3%, 6%, and 10%). They reported that melting and crystallization temperature of rice bran wax was not affected significantly from oil type while viscosity and textural properties were highly related with oil types. Salad oil showed softer organogels than olive oil and camellia oil. These results were linked to different fatty acid composition of the selected oil. Higher viscosity values and harder olegels were obtained from oil containing higher saturated TGA and high melting fatty acid.

Lupi et al. (2012) investigated the effects of organogelator and fat source on rheological properties of olive oil-based organogels prepared by different cacao butter and olive oil ratio. They concluded that at critical Myverol concentration (2%), crystallization and gelling behavior, and rheological properties were affected from fat source. Crystallization and gelling temperature and storage modulus values increased with saturation degree level (cacao butter fraction). Above critical point, rheological properties were only influenced by gelator concentration. Effect of the oil type on rheological and microstructural properties of the oleogels was investigated by Zetzl et al. (2014). Canola, soybean, and flaxseed oil was used to evaluate oil type effect on pore diameter of oleogel. Higher pore diameter was found for canola oil and pore diameter decreased with increasing unsaturations of the fatty acids present in the oil component.

It can be highlighted that gelling ability of gelators differs based on fatty acid composition, molecular weight, and acyl chain length of the edible oils. Lower amount of gelators can be used when the preparing of oleogels with vegetable oil containing high oleic content such as olive oil, rice bran oil, and high oleic sun flower oil. Using lower amount of gelators can provide economical advantages. On the other hand, healthy aspect (high contents of polyunsaturated fatty acids) and wide availability can be considered in addition to low-cost and textural properties. From this point, soybean and canola oils can be alternative to other edible oils.

Oleogelators

During conversion of liquid oil to hard substance, it is necessary to immobilize the liquid oils, which is achieved by means of oleogelators, structuring agents, responsible for the formation of 3-dimensional network. Well known and widely used oleogelators are vegetable waxes, monodiglycerides, alcohols or esters of fatty acids, phospholipids, and phytosterols (Pérez-Monterroza et al., 2014). Concentration of organogelators lower than 0.5% (weight bases) is adequate for gelation of organic solvent (Hughes et al., 2009). However, in food applications generally higher level is preferred, depending on the expected quality characteristics of oleogels. Oleogelators should typically have the following properties: (i) the presence of lipophilic and interacting parts, (ii) surface activity, (iii) thermoreversible characteristics, (iv) natural origin, and (v) being considered as GRAS (Grob et al., 1994; Patel et al., 2013; Doan et al., 2015). Gelling behavior of oleogelators are classified into four groups (Fig. 1) as the following (Patel and Dewettinck, 2015):

i.) crystalline particles (natural waxes, MAGs, DAGs, fatty acids + fatty alcohols (policosanol, cholesterol, D-limonene + medium chain tryglicerides + dicarboxylic acids), phytosterols + monoglycerides, sorbitan esters + phospholipids),
ii.) self-assembled structures with low molecular weight compounds (SAFIN forming fatty acids (hydroxy stearic acid, ricinelaidic acid, sphingolipids), phytosterols + sterol esters (sorbitanmonostearate, sorbitantristearate, γ-oryzanol + β-sitosterol, lecithin + sorbitantristearate), phospholipids + tocopherols (α-tocopherol + lecithin)),
iii.) self-assembled structures of polymers (hydrophobic (ethyl cellulose) and hydrophibic (methyl cellulose, hydroxypropyl methylcellulose), hydrophilic (β-lactoglobulins, gelatin) and hydrophobic (zein) proteins, protein + polysaccharides, chitosan and chitin, ethylene-vinylacetate-copolymer, Isoplen PP 070 homopolymer) and
iv.) miscellaneous structures or inorganic particles (inorganic particles (fumed silica), HIPE gels (polyglycerol-polyricinoleate + locust bean gum)).

In the self-assembly system molecular-level self-organization in oil phase results in formation of oleogels (Dassanayake et al., 2011). Regarding the crystal particles, nucleation followed by crystal growth is responsible for the network formation (Dassanayake et al., 2011).

According to the findings of previous studies, it was noted that waxes were the most efficient oleogelators since they have ability to crystallize even at concentrations lower than 10% to form network, which is associated with their strong oil-holding capacity (Hwang et al., 2012; Patel et al., 2013). Crystallization behavior of waxes is originated from their low polarity, long chain length, and high melting point (Mukkamala and Weiss, 1996). Therefore, selection of wax type and wax concentration be added to the formulation of the food material is substantial for obtaining the products with desired quality. Although waxes are typically known as long-chain fatty acids they also include different substances like hydrocarbons, fatty alcohols, ketones, mono-, di-, triacyl-glycerols, and sterol esters (Kolattukudy, 1976; Toro-Vazquez et al., 2010). During oleogelation, crystallization of oleogelators is indispensable process necessary for formation of gel network depending on the interaction of crystals and crystalline aggregates, resulting in the immobilization of liquid oil into three-dimensional network (Marangoni, 2012).

Several studies have already been reported dealing with wax crystallization in liquid oil such as candelilla wax in safflower oil (Toro-Vazquez et al., 2007), rice bran wax in olive oil (Dassanayake et al., 2009), sunflower wax in milk fat (Martini et al., 2008), plant waxes, and animal waxes in sunflower oil (Soomro and Sherazi, 2013), beeswax and sunflower wax in olive oil (Yılmaz and Ögütcü, 2014a), and beeswax in hazelnut oil (Yılmaz and Ögütcü, 2014b).

As seen many different organogelators have been investigated in terms of their organogel formation ability. And the previous studies showed that organogels could be used in different food formulations in order to decrease the saturated fatty acid concentration and to improve some quality properties of foods such as decreasing or eliminating fat bloom and fat migration. Such promising characteristics of oleogels have encouraged researchers to investigate potential use of oleogels in different food materials. Therefore, when considering food products, oleogelators be used in them should be food grade. In addition, the oleogelators must be safe, effective in gel formation at lower concentrations, easy available in market and economic as well as providing desired characteristics (Marangoni, 2012). Regarding those properties, it was suggested that waxes extracted from plants and saturated mono-glycerides could be used in food applications (Dassanayake et al., 2009; Da Pieve et al., 2010; Hwang et al., 2012; Marangoni, 2012). Chemical characteristics of the waxes namely, fatty acid, fatty alcohol and hydrocarbon concentrations, determine their physicochemical properties. The predominant chain length of rice bran wax, candedilla and carnauba waxes is 24 (40.5%), 30–32 (32% and 33%), and 24 (30%), respectively, and their melting points were found to be 78–82 °C, 60–73 °C, and 80–85°C, respectively (Warth, 1956; Tulloch, 1973; Vali et al., 2005; Dassanayake et al., 2009). Doan et al. (2015) investigated that rheological, thermal, and solid fat content properties of rice bran oil-based oleogels prepared with different natural waxes (sunflower wax, beeswax, rice bran wax, candedilla wax, carnauba wax, and berry wax). They found that using different waxes in oleogel formulation significantly affected all the characteristics. Moreover, minimum concentration of the oleogelators necessary for gel network formation also depends on their type. In high oleic sunflower oil, minimum gelling concentration of sunflower wax, carnauba wax, candedilla wax, bees wax, berry wax, and fruit wax changed between 0.5 and 7.0%. As ciritical gelling concentration of sunflower was found to be 0.5%, which was 7.0% for fruit waxes (Patel, 2015).

Several studies were also conducted on oleogelator characteristics of the hydrophobic polymers such as ethylcellulose (EC). Application of these types of polymer as oleogelators in food science and technology has attracted attention since they are commercially available, inexpensive in comparison with most of the highly purified organogelators. In addition to these advantages, EC is used as food grade or near-food grade material (Zetzl et al., 2014). EC is a semicrystalline polymer, which goes through a thermoreversible sol-gel transition in the presence of liquid oil. This behavior resulted from the polymer's ability to associate through physical bonds. Shear and temperature, solvent and surfactants type affect these interactions (Davidovich-Pinhas et al., 2016).

Gravelle et al. (2016) studied influence of solvent quality on the mechanical strength of EC oleogels. It was reported that gel strength of the oleogels was significantly influenced by the solvent polarity. According to their results, gel strength was positively correlated to polarity of the solvent. They also reported that this correlation was attributed to the ability of the polar groups present in the oil phase to interact with the EC gel network.

The effects of temperature on the mechanical properties of ethylcellulose (EC)-based oleogels have been conducted by Davidovich-Pinhas et al. (2015a). It was reported that gel strength of the oleogels increased with increasing setting temperature. Increase in the gel strength was linked to the formation of a more efficient polymer–polymer hydrogen-bonding network. They also reported that storage modulus decreased with increasing temperature due to weakening of the intermolecular bonds. According to their findings, higher gel strength was observed when the molten gel is heated above the polymer melting temperature (T_m).

Effects of the surfactants type on gel properties of the EC oleogels was studied by Davidovich-Pinhas et al (2015b). They reported that addition of the glycerol-based surfactant caused to dramatic decrease in the sol-gel and gel-sol transition temperatures in comparison with sorbitan-based surfactants. This behavior was linked to the plasticizing properties of the small head group of glycerol compared to the larger head group of sorbitan surfactants. Therefore, orgenogelator type be used in the formulation of the corresponding product should be adjusted carefully considering desired quality of the products and economic factors.

Oleogel applications in food industry

Application of the oleogels (or organogels) in both industrial and scientific fields has gradually increased (Terech and Weiss, 1997; Dassanayake et al., 2011; Marangoni and Garti, 2011) because of the promising characteristics of them. The major aim of the using oleogels in the food industry is reduction of saturated fatty acid composition since their excessive consumption have been associated with several health problem such as obesity, cardiovascular disease, metabolic syndrome, and diabetes (Marangoni and Garti, 2011; Gravelle et al., 2014). In recent years, change in attention of consumers about the detrimental effect of some food additives on human health has led to researchers to perform a works about finding alternative applications without health-adverse effects. The solid fats or shortenings used in the food industry are the questionable for human health due to their saturated and *trans* fatty acid contents. Also, the recent nutritional guidelines by different government agencies have recommended that a decrease in dietary saturated fat and also the reduction of *trans* fatty acids with replacement from unsaturated sources (Mert and Demirkesen, 2016a). Application of oleogels in the processed foods instead of solid fats fabricated by hydrogenization, esterification and fractionation processes have increased due to adverse effect of the consumption of the saturated and trans fatty acids. However, consumability of the products also depends on the other desired quality parameters as well as health beneficial effects. Therefore, in this study the effect of oleogel on the quality characteristics of different products was also mentioned to reveal advantages and disadvantages of oleogel usage in terms of quality of the product.

The findings related with oleogel applications indicated that heat stability of the chocolates is major problem for storing in countries with warm climate and chocolates stability should be improved by new approach such as oleogel usage. Another important problem is that transportation of oils in cream-including food products may be eliminated or reduced by using oleogel, which implied that oleogel might be used to solve the fat bloom problem in chocolate or chocolate-derived products. In addition, oleogels could be used as a carrier for water insoluble bioactive substances, improving releasing properties of such substances. The usage possibility of several oleogels prepared by different oils and oleogelators has been investigated for (Jang et al., 2015; Yilmaz and Ogutcu, 2015; Mert and Demirkesen, 2016a, b), spreads (Ogutcu and Yilmaz, 2014; Patel et al., 2014a; Ogutcu et al., 2015), cakes (Patel et al., 2014 a, b), chocolate pastes (Patel et al., 2014b), chocolate (Stortz et al., 2015), compound chocolate (Stortz and Marangoni, 2013; Stortz et al., 2014), frankfurters (Zetzl et al., 2012), and ice cream (Zulim Botenga et al., 2013b). General food applications of oleogels are summarized in Table 1.

In the food industry, as mentioned above oleogel formation is a novel strategy to impart solid-fat functionality to liquid oils (Nikiforisdis and Scholten, 2015). In oleogel production, the choice of oil type is a significant parameter as well as oleogelator type, as noted previously. For instance, it is recognized that canola oil is high in healthier unsaturated fats, compared to other vegetable oils. Therefore, canola oil was chosen to prepare oleogel samples containing a high level of unsaturated fatty acids (Jang et al., 2015). Another factor taken into consideration is obtaining the product with desired quality similar to that of conventional one in terms of physical and sensory characteristics. Cooked frankfurters including oleogels prepared with gelation of canola oil with ethylcellulose have very similar textural parameters with control sample in terms of chewiness and hardness parameters (Zetzl et al., 2012).

Table 1. Food applications of oleogels.

Food	Organogelator(s)	Organogelator(s) Concentration	Oil	Substituted Food Component (SFC)	Oleogel:SFC	Quality Parameters	Reference
Cookies	Candelilla wax	3.0%and 6.0% of oil (w/w)	Canola oil	Shortenings	30:70 60:40	Dimensional characteristics, moisture content, texture	Mert and Demirkesen (2016a)
	Candelilla wax Carnuaba wax	2.5%and 5.0% of oil (w/w)	Sunflower oil	Shortenings	100:0	Dimensional characteristics, moisture content, texture, rheology	Mert and Demirkesen (2016b)
	Candelilla wax	3.0%and 6.0% of oil (w/w)	Canola oil	Shortenings	100:0	Rheology, dimensional characteristics, texture	Jang et al. (2015)
	Beeswax Sunflower wax	5.0% of oil (w/w)	Hazelnut oil	Shortening	100:0	Physicochemical, sensory and texture properties	Yilmaz and Ogutcu (2015)
Spreads (Breakfast margarine)	Carnuaba wax	3.0%, 7.0%and 10.0% of oil (w/w)	Virgin olive oil	—	—	Physical, melting, texture properties, morphology, oxidation stability	Ogutcu and Yilmaz (2014)
	Beeswax	5.0% of oil (w/w)	Virgin olive oil	—	—	Physical, melting, texture properties, morphology, oxidation stability	Ogutcu et al. (2015)
	Shellac wax Rice bran wax Sunflower wax	5.0% of spread (w/w)	Rapeseed oil	as continuous oil phase	100:0	Microstructure, rheology, droplet size	Patel et al. (2014a)
Chocolate paste	Shellac wax	1.5% of chocolate paste (w/w)	Rapeseed oil	as oil binder	27:73	Rheology, solid fat content	Patel et al. (2014a)
Cakes	Shellac wax	5.0% of cakes (w/w)	Rapeseed oil	Shortening	100:0	Texture, sensory	Patel et al. (2014a)
	*Methylcellulose **Xanthan gum	*0.67% and 1.33% of oil (w/w) **1.0% of oil (w/w)	Sunflower oil	Shortening	100:0	Texture, sensory	Patel et al. (2014b)
Chocolate (Compound)	Ethylcellulose	1.0 and 2.2% of chocolate (w/w)	Palm kernel oil, Cacao butter	as heat resistant agent	100:0	Texture, oil migration, melting and physical properties	Stortz and Marangoni (2013)
	Ethylcellulose	2.17% of chocolate (w/w)	Palm kernel oil	as heat resistant agent	100:0	Texture and microstructure properties	Stortz et al. (2014)
Chocolate (milk, dark and white)	Ethylcellulose	2.17% of chocolate (w/w)	Palm kernel oil	as heat resistant agent	100:0	Texture and microstructure properties	Stortz et al. (2015)
Confectionery fillings	Bees wax	1.5–3% of oil	Rice bran oil	Palm oil	17:83 33:67 50:50	Rheological properties, Microstructure, Crystallization and melting behavior	Doan et al. (2016)
Frankfurters	Ethylcellulose	10.0% of oil	Canola oil Soybean oil Flaxseed oil	Beef fat	100:0	Fatty acid composition, fat globule size distribution, texture and microstructure properties	Zetzl et al. (2012)
Ice cream	Rice bran wax	10.0% of oil	High oleic sunflower oil	Butter (80% fat)	100:0	Overrun, particle size distribution, meltdown stability, melting and microstructure properties	Zulim Botega et al. (2013a)
	Rice bran wax Candelilla wax Carnuaba wax	10.0% of oil	High oleic sunflower oil	Butter (80% fat)	100.0	Air cell size distribution and shape, mix microstructure, meltdown rate, over run	Zulim Botega et al. (2013b)
Cream cheese	Rice bran wax ●Ethylcellulose	10.0% of oil	Soybean oil High oleic soybean oil	Milk fat	100.0	Rheology, texture, microstructure, sensory properties, fatty acid composition	Bemer et al. (2016)
Emulsions	Shellac wax	0–6% of oil	Rapeseed oil	Rapeseed oil	100.0	Rheology, microstructure	Patel et al (2013)

For example, the difficulty in reducing fat ingestion arises mainly from the fact that saturated fats contribute to many of the important organoleptic properties of foods, including palatability, lubrication and structure (Ceballos et al., 2014). Also, the major approach to forming novel oil-based materials is to incorporate oil components that by various molecular interactions will alter the physical properties of the oil so that its fluidity will decrease and the rheological properties will be similar to those of fats (Marangoni and Garti, 2011). Such effects are striking in especially for bakery products. Therefore, when oleogels are used in the products like cookies and cake, physical, textural and sensory characteristics of the end product should also be considered as well as fatty acid composition. In study carried out be Jang et al. (2015), the oleogels prepared from canola oil with candelilla wax were utilized as a shortening replacer to prepare cookies with a high level of unsaturated fatty acids. The candelilla wax (3.0% and 6.0%, by weight) was incorporated to canola oil in order to produce the oleogels with solid-like properties. They reported that the firmness of the oleogels was lower than that of the shortening at room temperature. Steady shear measurements showed that viscosity of the oleogels decreased rapidly with increasing temperature, and increased with increasing levels of candelilla wax (Jang et al., 2015).

In products, it may be possible to apply vegetable oil to baked goods instead of shortening. However, the use of vegetable oil produces baked goods with more greasy and less crispy characteristics, and also decreases the storage stability of the products mainly due to oil oxidation. In addition, the low viscosity of the oil causes a difficulty in handling and shaping of dough (Jang et al., 2015). Also, in other recent studies, potential application of Candelilla wax (Mert and Demirkesen, 2016a) and Carnauba wax (Mert and Demirkesen, 2016b) containing oleogels for partial replacement of the shortening in cookies were investigated. When the shortening was completely replaced with oleogel softer products were obtained compared to liquid oil, but they were harder than the shortening containing products. On the other hand, partial replacement of shortening with oleogels achieved much more acceptable dough and cookie characteristics. However, texture measurements showed that Carnauba wax and Candelilla wax oleogels were not able to form short dough as good as commercial shortening with higher solid fat content and this resulted with cookies which had harder texture and higher spread ratio (Mert and Demirkesen, 2016b). Results in the related studies suggest that gradual replacement of shortening with oleogels may be a suitable approach for reduction of saturated fat in short dough products (Mert and Demirkesn, 2016a). The findings of the studies related with usage of oleogels in the bakery products implied that by partially replacing of shortening with oleogels resulted in the fabrication of the end product with lower saturated fatty acid content when compared with that of conventionally produced ones. However, by adjusting oleogelator concentration and oil type, the production of suitable formulations can be possible for bakery products. Therefore, more efforts should be expended in this area.

According to the results related with usage of oleogels at different levels in different food products, especially textural and rheological properties of chocolate and chocolate-derived products including oleogel were found to be acceptable levels when compared with the conventional ones and it also concluded that oleogels could be used to produce heat resistant chocolate (Stortz and Marangoni, 2013; Patel et al., 2014a; Stortz et al., 2014; Stortz et al., 2015). These previous results are very significant since shelf life of such products could be improved at different climatic conditions as well as tolerating the quality defects resulted from storing samples at improper storage and selling conditions. In addition, there are limited studies where the oleogels were applied to meat products (Zetzl et al., 2012), and ice cream (Zulim Botega et al., 2013b). There is a need to extend the use of oleogels to a wider variety of products (Jang et al., 2015).

The physical, mechanical and organoleptic characteristics of many food products are largely influenced by the functional properties of their solid fat content (Gravelle et al., 2014). When the studies about all product groups were investigated, the fact that sensorial analyses were carried out in only three studies (Patel et al., 2014a, b; Yilmaz and Ogutcu, 2015). Shortening is an edible fat that has been traditionally used to make baked products such as pastries, cakes, and cookies. Since shortening prevents the cohesion of gluten strands during mixing, it plays a critical role in the tender texture and mouth feel of the final products (Jang et al., 2015). Therefore sensory properties of the products are one of the most important factors of using shortenings in food formulations. From this reason, oleogel usage could also be investigated regarding sensory properties of the end product. Moreover, it will belong to investigate release level of the aroma-associated volatile compounds in the oleogel including media. Unusual taste or aroma can be felt in the products containing oleogel due to the wax, which could be eliminated by application of deodorizing process during fabrication of the oleogels. As seen, although the influence of oleogel use on some quality characteristics of the products was investigated, there are sensory results deficiencies. When considering the fact that sensory characteristics are one of the main factors determining desirableness of the products, more studies in this subject are required.

In the food technology, employability of the new ingredients in the existing equipments and systems or incorporating of such ingredients in the processes with minor modifications in the process becomes prominent considering economical and industry-related factors. Oleogels are shown to have great potential applications in the food industry because oleogels have specific consistency and firmness without changing their chemical compositions (Marangoni, 2012; Jang et al., 2015). Considering those characteristics, it could be stated that some oleogelators such as ethylcelulose and methyl cellulose have more advantages. In a various studies, the use of ethyl cellulose has been investigated for its application in various aspects of the food industry such as organogelator for vegetable oils (Aiache et al., 1992; Almeida and Bahia, 2006; Gravelle et al., 2012a; Zetzl et al., 2012). In the most of studied products, oleogels are used as a substitute for the traditionally used fat source (Zetzl et al., 2014). Also, ethyl cellulose based oleogels have shown promise as an oil structuring agent in many applications, which are under protection with patents such as replacement of fats in foods (US8940354 B2), heat resistance agent in chocolate (US2012183651 A1), oil binding agents in bakery

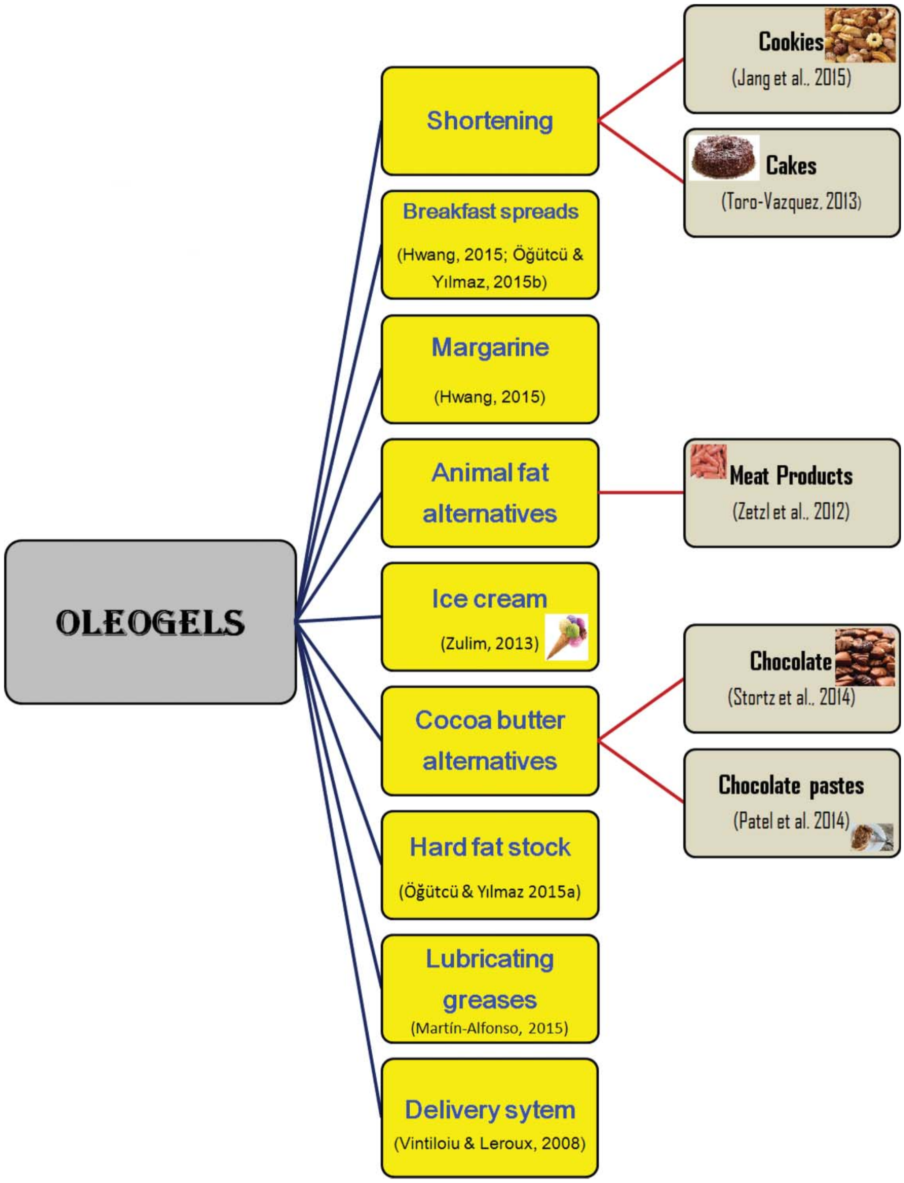

Figure 2. Usage purposes of oleogels and corresponding materials where oleogels were used in different studies.

products (US20140044839 A1) (Davidovich-Pinhas et al., 2015b). Ethyl cellulose has also advantages regarding regulations. Ethyl cellulose has listed as a food additive by the FAO with collaboration with the WHO (Zetzl et al., 2014). This preamble lists over 60 different food applications for ethylcellulose, including confectionery products, fat spreads and emulsions, cheeses, poultry and game products, processed comminuted meat and batters (Zetzl et al., 2014).

As a summary, applications of the oleogels are summarized in Fig. 2. Traditionally, application of oleogels include the stabilization of emulsions, tailoring the rate of nutraceutical release, restriction of oil migration, and/or the direct replacement of fats rich in saturated and *trans*-fatty acids (Marangoni and Garti, 2011; Gravelle et al., 2012a; Jang et al., 2015). Oleogelation has been used for the development of delivery systems of nutraceuticals (Mert and Demirkesen, 2016a). In the work performed by Ceballos et al. (2014), they showed that ethylcellulose dissolved in medium chain triglycerides or soybean oil is able to stabilize nonaqueous emulsions of propylene glycol as dispersed phase. Propylene glycol-in-oil emulsions have interesting structural and flow properties which make them attractive to be used in food formulations, either as emulsions themselves or as potential vehicles for active ingredients (Ceballos et al., 2014). It is needed to obtain functional foods with delivering of bioactive compounds by means of oleogels and make trials in terms of effectiveness of such systems in different food matrix.

Conclusion

Health concerns of consumers forced them to consume the products with lower saturated and *trans*-fatty acid contents due to their adverse health effects in terms of cardiovascular disease, diabetes, and obesity. Therefore, one of the most important aim of the food industry and researchers to produce solid fats rich in unsaturated fatty acid contents. One of the conspicuous is production of oleogels which are structured oils prepared from oils by means of oleogelator. In this study, history of oleogels, raw materials used in oleogels, production methods and their functions in oleogel quality and applications of oleogels in different food products were reviewed. According to the results of previous studies, oleogels could be used in bakery products, chocolate products, and meat products to reduce saturated fatty acid composition without deteriorating the product quality. However, more detailed studies are needed to exactly determine their potential applications. Sensory studies are required to evaluate the sensory characteristics of the oleogel-including products and depending on the obtained results modifications in formulation or production methods such as deodorization could be needed.

Conflict of interest

There are no financial disclosures or conflict of interest for any of the authors.

ORCID

Mehmet Demirci http://orcid.org/0000-0002-4394-9852
Omer Said Toker http://orcid.org/0000-0002-7304-2071
Salih Karasu http://orcid.org/0000-0003-0597-283X
Osman Sagdic http://orcid.org/0000-0002-2063-1462

References

Abdallah, D. J., Sirchio, S. A. and Weiss, R. G. (2000). Hexatriacontane organogels. The first determination of the conformation and molecular packing of a low-molecular-mass organogelator in its gelled state. *Langmuir* **16**:7558–7561.

Aiache, J. M., Gauthier, P. and Aiache, S. (1992). New gelification method for vegetable oils I: Cosmetic application. *Int. J. Cosmetic Sci.* **14**:228–234.

Almedia, I. F. and Bahia, M. F. (2006). Evaluation of the physical stability of two oleogels. *Int. J. Pharm.* **327**:73–77.

Ambrosini, G. L. (2014). Childhood dietary patterns and later obesity: A review of the evidence. *Proc. Nutr. Soc.* **73**:137–146.

American Heart Association Statistics Committee and Stroke Statistics Subcommittee. (2012). Heart disease and stroke statistics–2012 update: A report from the American Heart Association. Circulation. *J. Am. Heart Assoc.* **125**:e2–e220.

Başoğlu, F. (2014). Yemeklik Yağ Teknolojisi. Dora Publications, 4 press, Bursa, Turkey.

Bemer, H. L. and Limbaugh, M., et al. (2016). Vegetable organogels incorporation in cream cheese products. *Food Res. Int.* **85**:67–75.

Bot, A., Veldhuizen, Y. S. J., den Adel, R. and Roijers, E. C. (2009). Non-TAG structuring of edible oils and emulsions. *Food Hydrocolloid.* **23**:1184–1189.

Bot, A., Flöter, E., Lammers, J. G. and Pelan, E. G. (2007). Understanding and Controlling the Microstructure of Complex Foods, pp. 575–599. McClements, D. J., Ed., Woodhead Publishing, Cambridge.

Botega, D. C. Z. (2012). Application of rice bran wax organogel to substitute solid fat and enhance unsaturated fat content in ice cream. Thesis, The University of Guelph, Guelph, Ontario, Canada.

Brouwer, I. A., Wanders, A. J. and Katan, M. B. (2010). Effect of animal and industrial trans fatty acids on HDL and LDL cholesterol levels in humans e a quantitative review. *Plos One* **5**(3):1–10.

Calligaris, S., Da Pieve, S., Arrighetti, G. and Barba, L. (2010). Effect of the structure of monoglyceride–oil–water gels on aroma partition. *Food Res. Int.* **43**:671–677.

Ceballos, M. R., Brailovsky, V., Bierbrauer, K. L., Cuffini, S. L., Beltramo, D. M. and Bianco, I. D. (2014). Effect of ethylcellulose on the structure and stability of non-aqueous oil based propylene glycol emulsions. *Food Res. Int.* **62**:416–423.

Co, E. D. and Marangoni, A. G. (2012). Organogels: An alternative edible oil-structuring method. *J. Am. Oil Chem. Soc.* **89**:749–780.

Dassanayake, L. S. K., Kodali, D. R., Ueno, S. and Sato, K. (2009). Physical properties of rice bran wax in bulk and organogels. *J. Am. Oil Chem. Soc.* **86**(12):1163–1173.

Dassanayake, L. S. K., Kodali, D. R. and Ueno, S. (2011). Formation of oleogels based on edible lipid materials. *Curr. Opin. Colloid In.* **16**:432–439.

Dassanayake, L. S. T., Rodali, D. R., Ueno, S. and Sato, K. (2012). Crystallization kinetics of organogels prepared by rice bran wax and vegetable oils. *J. Oleo Sci.* **61**:1–9.

Da Pieve, S., Calligaris, S., Co, E., Nicoli, M. C. and Marangoni, A. G. (2010). Shear nanostructuring of monoglyceride organogels. *Food Biophys.* **5**:211–217.

Davidovich-Pinhas, M., Barbut, S. and Marangoni, A. G. (2015). The role of surfactans on ethylcelluloseoleogel structure and mechanical properties. *Carbohyd. Polym.* **127**:355–362.

Davidovich-Pinhas, M., Gravelle, A. J., et al. (2015a). Temperature effects on the gelation of ethylcelluloseoleogels. *Food Hydrocolloids.* **46**:76–83.

Davidovich-Pinhas, M., Barbut, S. and Marangoni, A. G. (2015b). The role of surfactans on ethylcelluloseoleogel structure and mechanical properties. *Carbohyd. Polym.* **127**:355–362.

Davidovich-Pinhas, M., Barbut, S., et al. (2016). Development, characterization, and utilization of Food-grade polymer oleogels. *Annu Rev. Food Sci. Technol.* **7**(1):65–91.

Demirci, M. (2003). Beslenme, Rebel Publication, 1. Ed., Tekirdağ, Turkey.

Doan, C. D., de Walle, D. V., Dewettinck, K. and Patel, A. R. (2015). Evaluating the oil-gelling properties of natural waxes in rice bran oil: Rheological, thermal and microstructural study. *J. Am. Oil Chem. Soc.* **92**:801–811.

Doan, C. D., Patel, A. R., Tavernier, I., De Clercq, N., Van Raemdonck, K., Van de Walle, D., Delbaere, C. and Dewettinck, K. (2016). The feasibility of wax-based oleogel as a potential co-structurant with palm oil in low-saturated fat confectionery fillings. *Eur. J. Lipid Sci. Technol.* **118**:1903–1914. doi:10.1002/ejlt.201500172.

Domingues, M. A. F., Ribeiro, A. P. B., Chiu, M. C.and Gonçalves, L. A. G. (2015). Sorbitan and sucrose esters as modifiers of the solidification properties of zero trans fats. *LWT-Food Sci. Technol.* **62**(1):122–130.

Eitel, W. (1975). Silicate Structures and Dispersion System. Academic Press Inc, New York.

Gallego, R., Arteaga, J. F., Valencia, C. and Franco, J. M. (2013). Rheology and thermal degradation of isocyanate-functionalized methyl cellulose-based oleogels. *Carbohyd. Polym.* **98**(1):152–160.

Gandolfo, F. G., Bot, A. and Flöter, E. (2004). Structuring of edible oils by long-chain FA, fatty alcohols, and their mixtures. *J. Am. Oil Chem. Soc.* **81**(1):1–6.

Gravelle, A. J., Barbut, S. and Marangoni, A. G. (2012a). Ethylcelluloseoleogels: Manufacturing considerations and effects of oil oxidation. *Food Res. Int.* **48**:578–583.

Gravelle, A. J., Barbut, S., Quinton, M. and Marangoni, A. G. (2014). Towards the development of predictive model of the formulation-dependent mechanical behavior of edible oil-based ethylcelluloseoleogels. *J. Food Eng.* **143**:114–122.

Gravelle, A. J., Davidovich-Pinhas, M., Zetzl, A. K., Barbut, S., & Marangoni, A. G. (2016). Influence of solvent quality on the mechanical strength of ethylcellulose oleogels. *Carbohyd Polymers*, **135**:169–179.

Grob, K, Giuffré, A. M., Leuzzi, U. and Mincione, B. (1994). Recognition of adulterated oils by direct analysis of the minor components. *Lipid/Fett.* **96**(8):286–290.

Gronwald, O., Snip, E. and Shinkai, S. (2002). Gelators for organic liquids based on self-assembly: A new facet of supramolecular and combinatorial chemistry. *Curr. Opin. Colloid In.* **7**:148–156.

Guenet, J. M. (2008). Polymer-Solvent Molecular Compounds. Elsevier Ltd., Oxford.

Hinze, W. L., Uemasu, I., Dai, F. and Braun, J. M. (1996). Analytical and related applications of organogels. *Curr. Opin. Colloid In.* **1**:502–513.

Hughes, N. E., Marangoni, A. G., Wright, A. J., Rogers, M. A. and Rush, J. W. E. (2009). Potential applications of edible oil organogels. *Trends Food Sci. Tech.* **20**:470–480.

Hwang, H.- S., Kim, S., Singh, M., Winkler-Moser, J. K. and Liu, S. X. (2012). Organogel formation of soybean oil with waxes. *J. Am. Oil Chem. Soc.* **89**(4):639–647.

Hwang, H. S., Kim, S., Evans, K. O., Koga, C. and Lee, Y. (2015). Morphology and networks of sunflower wax crystals in soybean oil organogel. *Food Struct.* **5**:10–20.

Jang, A., Bae, W., Hwang, H. S., Lee, H. G. and Lee, S. (2015). Evaluation of canola oil oleogels with candelilla wax as an alternative to shortening in baked goods. *Food Chem.* **187**:525–529.

Kolattukudy, P. E. (1976). Chemistry and Biochemistry of Natural Waxes. Elsevier Scientific Pub. Co, Amsterdam.

Lakmali, S. K. D., Dharma, R. K. and Ueno, S. (2011). Formation of oleogels based on edible lipid materials. *Curr. Opin. Colloid In.* **16**:432–439.

Lopez-Martínez, A., Charó-Alonso, M. A., Marangoni, A. G. and Toro-Vazquez, J. F. (2015). Monoglycerideorganogels developed in vegetable oil with and without ethylcellulose. *Food Res. Int.* **72**:37–46.

Lupi, F. R., Gabriele, D. and de Cindio, B. (2012). Effect of shear rate on crystallisation phenomena in olive oil based organogels. *Food Bioprocess Tech.* **5**:2880–2888.

Lupi, F. R., Gabriele, D., Greco, V., Baldino, N., Seta, L. and De Cindio, B. (2013). A rheological characterisation of an olive oil/fatty alcohols organogel. *Food Res. Int.* **51**(2):510–517.

Marangoni, A. G. (2012). Organogels: an alternative edible oilstructuring method. *J. Am. Oil Chem. Soc.* **89**(5):749–780.

Marangoni, A. G. and Garti, N. (2011). Edible Oleogels: Structure and Health Implications. AOCS Press, Urbana, Illinois.

Marangoni, A. G. (2000). Elasticity of high-volume-fraction fractal aggregate networks: A thermodynamic approach. *Phys. Rev. B.* **62**:13951–13955.

Marangoni, A. G. (2004). Fat Crystal Networks. Marcel Dekker, New York.

Marangoni, A. G. and Rogers, M. A. (2003). Structural basis for the yield stress in plastic disperse systems. *Appl. Phys. Lett.* **82**:3239–3241.

Martín-Alfonso, J. E. and Franco, J. M. (2015). Influence of polymer reprocessing cycles on the microstructure and rheological behavior of polypropylene/mineral oil oleogels. *Polymer Test.* **45**:12–19.

Martini, S., Carelli, A. A. and Lee, J. (2008). Effect of the addition of waxes on the crystallization behavior of anhydrous milk fat. *J. Am. Oil Chem. Soc.* **85**(12):1097–1104.

Mensink, R. P., Zock, P. L., Kester, A. D. M. and Katan, M. B. (2003). Effects of dietary fatty acids and carbohydrates on the ratio of serum total to HDL cholesterol and on serum lipids and apolipoproteins: A meta-analysis of 60 controlled trials. *Am. J. Clin. Nutr.* **77**(5):1146–1155.

Mert, B. and Demirkesen, I. (2016a). Reducing saturated fat with oleogel/shortening blends in baked product. *Food Chem.* **199**:809–816.

Mert, B. and Demirkesen, I. (2016b). Evaluation of highly unsaturated oleogels as shortening replacer in short dough product. *LWT–Food Sci. Tech.* **68**:477–484.

Micha, R. and Mozaffarian, D. (2009). Trans fatty acids: Effects on metabolic syndrome, heart disease and diabetes. *Nat. Rev. Endocrinol.* **5**:335–344.

Mozaffarain, D., Aro, A. and Willet, W. C. (2009). Health effects of trans-fatty acids: experimental and observational evidence. *Europ. J. Clin. Nutr.* **63**:S5–S21.

Muguerza, E., Fista, G., Ansorena, D., Astiasaran, I. and Bloukas, J. G. (2002). Effect of fat level and partial replacement of pork backfat with olive oil on processing and quality characteristics of fermented sausages. *Meat Sci.* **61**:397–404.

Mukkamala, R. and Weiss, R. G. (1996). Physical gelation of organic fluids by anthraquinone-steroid-based molecules. Structural features influencing the properties of gels. *Langmuir* **12**:1474–1482.

Narine, S. S. and Marangoni, A. G. (1999a). Fractal nature of fat crystal networks. *Phys. Rev. E.* **59**:1908–1920.

Nikiforidis, C. V. and Scholten, E. (2015). High internal phase emulsion gels (HIPE-gels) created through assembly of natural oil bodies. *Food Hydrocolloid.* **43**:283–289.

O'Brien, R. D. (2004). Fats and Oils: Formulating and Processing for Applications, 2nd edn. CRC Press, Boca Raton, FL, ABD.

O'Brien, R. D. (2003). Fats and Oils, 2nd edition, pp. 92–104. CRC Press, Boca Raton, FL.

Öğütcü, M. and Yılmaz, E. (2014). Oleogels of virgin olive oil with carnauba wax and monoglyceride as spreadable products. *Grasas y Aceites* **65**(3):e040.

Öğütcü, M. and Yılmaz, E. (2015a). Characterization of hazelnut oil oleogels prepared with sunflower and carnauba waxes. *Int. J. Food Prop.* **18**(8):1741–1755.

Öğütcü, M. and Yılmaz, E. (2015b). Comparison of the pomegranate seed oil organogels of carnauba wax and monoglyceride. *J. Appl. Polym. Sci.* **132**:41343, doi: 10.1002/app.41343.

Ogutcu, M., Arifoglu, N. and Yilmaz, E. (2015). Preparation and characterization of virgin olive oil-beeswax oleogel emulsion products. *J. Am. Oil Chem. Soc.* **92**:459–471.

Ojijo, N. K. O., Neeman, I., Eger, S. and Shimoni, E. (2004). Effects of monoglyceride content, cooling rate and shear on the rheological properties of olive oil/monoglyceride gel networks. *J. Sci. Food Agr.* **84**:1585–1593.

Patel, A. R., Schatteman, D., De Vos, W. H., Lesaffer, A. and Dewettinck, K. (2013). Preparation and rheological characterization of shellac oleogels and oleogel-based emulsions. *J. Colloid and Interf. Sci.* **411**:114–121.

Patel, A. R., Cludts, N., Sintang, M. D. B., Lesaffer, A. and Dewettinck, K. (2014a). Edible oleogels based on water soluble food polymers: preparation, characterization and potential application. *Food Function* **5**(11):2833–2841.

Patel, A. R., Rajarethinem, P. S., Gredowska, A., Turhan, O., Lesaffer, A., De Vos, W. H. and Dewettinck, K. (2014b). Edible applications of shellac oleogels: spreads, chocolate paste and cakes. *Food Function*. **5**(4):645–652.

Patel, A. R. (2015). Alternative Routes to oil Structuring. Hartel, R. W., Clark, P. J., Finley, J. W., Rodriguez-Lazaro, D., Roos, Y. and Topping, D., Eds. 1st ed., Springer International Publishing, New York.

Patel, A. R. and Dewettinck, K. (2015). Comparative evaluation of structured oil systems: Shellac oleogel, HPMC oleogel, and HIPE gel. *Eur. J. Lip. Sci. Tech.* **117**:1772–1781.

Pérez-Monterroza, E. J., Márquez-Cardozo, C. J. and Ciro-Velásquez, H. J. (2014). Rheological behavior of avocado (*Perseaamericana Mill, cv. Hass*) oleogels considering the combined effect of structuring agents. *LWT-Food Sci. Tech.* **59**:673–679.

Pernetti, M., van Malssen, K. F., Floter, E. and Bot, A. (2007a). Structuring of edible oils by alternatives to crystalline fat. *Curr. Opin. Coll. In.* **12**:221–231.

Pernetti, M., van Malssen, K. F., Floter, E. and Bot, A. (2007b). Structuring edible oils by alternatives to crystalline fat. *Curr. Opin. Coll. In.* **84**:989–1000.

Riccardi, G., Giacco, R. and Rivellese, A. A. (2004). Dietary fat, insulin sensitivity and the metabolic syndrome. *Clin. Nutr.* **23**:447–456.

Rogers, M. A., Wright, A. J. and Marangoni, A. G. (2008). Engineering the oil binding capacity and crystallinity of self-assembled fibrillar networks of 12-hydroxystearic acid in edible oils. *Soft Matter.* **4**(7):1483–1490.

Rogers, M. A., Wright, A. J. and Marangoni, A. G. (2009). Oil organogels: the fat of the future. *Soft Matter.* **5**:1594–1596.

Sahoo, S., Kumar, N., Bhattacharya, C., Sagiri, S. S., Jain, K., Pal, K. and Nayak, B. (2011). Organogels: properties and applications in drug delivery. *Des. Monomers Polym.* **14**(2):95–108.

Sánchez, R., Franco, J. M., Delgado, M. A., Valencia, C. and Gallegos, C. (2008). Effect of thermo-mechanical processing on the rheology of oleogels potentially applicable as biodegradable lubricating greases. *Chem. Eng. Res. Des.* **86**(10):1073–1082.

Sánchez, R., Stringari, G. B., Franco, J. M., Valencia, C. and Gallegos, C. (2011). Use of chitin, chitosan and acylated derivatives as thickener agents of vegetable oils for bio-lubricant applications. *Carbohyd. Polym.* **85**(3):705–714.

Sagiri, S. S., Singh, V. K., Pal, K., Banerjee, I. and Basak, P. (2015). Stearic acid based oleogels: A study on the molecular, thermal and mechanical properties. *Mater. Sci. Eng. C.* **48**:688–699.

Soomro, R. K. and Sherazi, S. T. H. (2013). Extraction and characterization of seed oil waxes by using chromatographic techniques. *Int. J. Ind. Chem.* **4**(1):1–7.

Stauffer, C. E. (1996). Fats and Oils Practical Guide for the Food Industry. An Eagan Press Handbook AACC Inc, Minnesota, ABD.

Stortz, T. A., De Moura, D. C., Laredo, T. and Marangoni, A. G. (2014). Molecular interactions of ethylcellulose with sucrose particles. *RSC Advances* **4**(98):55048–55061.

Stortz, T. A. and Marangoni, A. G. (2013). Ethylcellulose solvent substitution method of preparing heat resistant chocolate. *Food Res. Int.* **51**(2):797–803.

Stortz, T. A., Zetzl, A. K., Barbut, S., Cattaruzza, A. and Marangoni, A. G. (2012). Edible oleogels in food products to help maximize health benefits and improve nutritional profiles. *Lipid Tech*. **24**(7):151–154.

Stortz, T. A., Laredo, T. and Marangoni, A. G. (2015). The role of lecithin and solvent addition in ethylcellulose-stabilized heat resistant chocolate. *Food Biophys*. **10**:253–263.

Tanti, R. (2015). Hydroxypropyl-methylcellulose and methylcellulose structured oils as an alternative low saturated fat stabilizer and shortening replacement for food applications, Thesis, The University of Guelph, Guelph, Ontario, Canada.

Terech, P. and Weiss, R. G. (1997). Low molecular mass gelators of organic liquids and the properties of their gels. *Chem. Rev.* **97**:3133–3159.

Toro-Vazquez, J. F., Morales-Rueda, J., Mallia, V. A. and Weiss, R. G. (2010). Relationship between molecular structure and thermo-mechanical properties of candelilla wax and amides derived from (R)12-hydroxystearic acid as gelators of safflower oil. *Food Biophys.* **5**(3):193–202.

Toro-Vazquez, J. F., Morales-Rueda, J. A., Dibildox-Alvarado, E., Charó-Alonso, M., Alonzo-Macias, M. and González-Chávez, M. (2007). Thermal and textural properties of organogels developed by candelilla wax in safflower oil. *J. Am. Oil Chem. Soc.* **84**(11):989–1000.

Tulloch, A. P. (1973). Comparison of some commercial waxes by gas liquid chromatography. *J. Am. Oil Chem. Soc.* **50**(9):367–371.

Uauy, R., Aro, A., Clarke, R., Ghafoorunissa, R., L'Abbe, M., Mozaffarian, D., Skeaff, M., Stender, S. and Tavella, M. (2009). WHO update on trans fatty acids: summary and conclusions. *Eur. J. Clin. Nutr.* **63**:S68–S75.

Vaclavik, V. A. and Christian, E. W. (2014). Essentials of Food Science. Heldman, D.R., Ed., 4th edition, Springer, New York.

Vali, S. R., Ju, Y. H., Kaimal, T. N. and Chern, Y. T. (2005). A process for the preparation of food grade rice bran wax and the determination of its composition. *J. Am. Oil Chem. Soc.* **82**(1):57–64.

Van Esch, J. H. and Feringa, B. L. (2000). New functional materials based on selfassembling organogels: from serendipity towards design. *Angew. Chem. Int. Edit.* **39**:2263–2266.

Vandana, D., Neelam, G., Kulveer Singh, A. and Bhupender, S. K. (2011). Trans fats-sources, health risks and alternative approach—a review. *J. Food Sci. Tech.* **48**:534–541.

Vintiloiu, A. and Leroux, J. C. (2008). Organogels and their use in drug delivery—a review. *J. Control. Release* **125**(3):179–192.

W.H.O. (2002). The World Health Report, 2002: Reducing Risks, Promoting Healthy Life. World Health Organization, Geneva.

Warth, A. H. (1956). The Chemistry and Technology of Waxes, pp. 940. Reinhold Publishing Corporation, New York (NY).

Weiss, R. G. (2006). Terech, P. Introduction. **In**: Molecular Gels Materials with Self-Assembled Fibrillar Networks, pp. 1–13. Weiss, R. G. and Terech, P., Eds., Springer, Dordrecht, The Netherlands.

Wright, A. and Marangoni, A. (2006). Formation, structure, and rheological properties of ricinelaidic acid-vegetable oil organogels. *J. Am. Oil Chem. Soc.* **83**(6):497–503.

Wright, A. J., Marangoni, A. G. and Garti, N. (2011). Vegetable oil-based ricinelaidic acid organogels—phase behavior, microstructure and rheology. Edible Oleogels: Structure and Health Implications, pp. 81–99. AOCS Press, Urbana.

Yılmaz, E. and Ogutcu, M. (2015). The texture, sensory properties and stability of cookies prepared with wax oleogels. *Food Funct.* **6**(4):1194–1204.

Yılmaz, E. and Öğütcü, M. (2014a). Comparative analysis of olive oil organogels containing beeswax and sunflower wax with breakfast margarine. *J. Food Sci.* **79**(9):E1732–E1738.

Yılmaz, E. and Öğütcü, M. (2014b). Properties and stability of hazelnut oil organogels with beeswax and monoglyceride. *J. Am. Oil Chem. Soc.* **91**(6):1007–1017.

Zetzl, A. K., Marangoni, A. G. and Barbut, S. (2012). Mechanical properties of ethylcelluloseoleogels and their potential for saturated fat reduction in frankfurters. *Food Funct.* **3**(3):327–337.

Zetzl, A. K., Gravelle, A. J., Kurylowicz, M., Dutcher, J., Barbut, S. and Marangoni, A. G. (2014). Microstructure of ethylcelluloseoleogels and its relationship to mechanical properties. *Food Struct*. **2**:27–40.

Zahi, M. R., Liang, H. and Yuan, Q. (2015). Improving the antimicrobial activity of D-limonene using a novel organogel-based nanoemulsion. *Food Control*. **50**:554–559.

Zulim, B. D. C., Marangoni, A. G., Smith, A. K. and Goff, H. D. (2013a). The potential application of rice bran wax oleogel to replace solid fat and enhance unsaturated fat content in ice cream. *J. Food Sci.* **78**(9):C1334–C1339.

Zulim Botega, D. C., Marangoni, A. G., Smith, A. K. and Goff, H. D. (2013b). Development of formulations and processes to incorporate wax oleogels in ice cream. *J. Food Sci.* **78**:C1845–C1851.