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The application of oleogels in food products: Classification, preparation, and characterisation

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REVIEW PAPER

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ABSTRACT

Oleogels have been extensively investigated in the food processing in recent years, and they have become one of the healthier alternative. The possibility of constructing oleogel material in a manner similar to hydrocolloid gel has now been gradually becoming a reality. In this regard, this review provides coverage of the latest developments and applications of oleogels in terms of preparation strategies, physicochemical properties, health aspects, and potential food applications. Both solid fat content and crystallisation behaviour are discussed for oleogels fabricated by gelators and under different conditions. Oleogels could replace hydrogenated vegetable oils in food product, reduce the fatty acid content, and be used to prepare food products such as meat, ice-cream, chocolate, bread, and biscuits with desirable properties. The aims were to assess the formation mechanism, construction methods of oleogels and the advance on the application of oleogel structures in the food field, as well as the further exploration of oleogels and in complex food systems in the future.

KEYWORDS

oleogels, classification, preparation, application

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1. INTRODUCTION

The relation of dietary fat and the risk of many chronic diseases has been raising concern recently. The excessive intake of fat is linked to diseases such as obesity, heart disease, diabetes, liver and pancreatic cancer (Zhu et al., 2019). The edible oils of butter and coconut oil rich in saturated fatty acids (SFAs) are commonly used, while olive and canola oils contain high levels of unsaturated fatty acids (UFAs). Coconut oil, olive oil, or butter increase TC/HDL-C ratio in blood lipids in healthy men and women (Khaw et al., 2018). Palm oil (PO) is the most abundant vegetable oil in the world, with almost equal amounts of SFA (mainly palmitic acid 44–45%) and unsaturated fatty acids (oleic acid 39–40%; linoleic acid 10–11%). Although PO has been suggested as an alternative to partially hydrogenated fats in the food supply, it has been shown that palm oil may be an unhealthy fat due to its high saturated fatty acid content (Fattore et al., 2014). SFA in the diet is mostly found in animal fat and milk fat, which is rich in cholesterol. Increases of blood cholesterol, triacylglycerols, and low-density lipoprotein cholesterol (LDL-C) can be caused by excessive SFA consumption. Increasing intake of saturated fat is correlated with diabetes, hypertension, and obesity (Gribbin et al., 2022). Studies have shown that excessive consumption of SFAs is the cause of chronic diseases (Pigsborg et al., 2022). Although foods that are rich in saturated fatty acids are more delicious, excessive intake of total fat and saturated fat has become one of the main killers that threaten human life and health. The dietary guidelines also recommend to limit the consumption of products with high SFA content. Therefore, many researchers in the food industry focus on the development of low-fat foods. To completely or partially remove unnecessary fats from food while maintaining its original sensory and quality characteristics is a great challenge.

Oleogels have attracted widespread attention due to their low saturated fatty acid and low trans fatty acid (TFA) contents, safety, and easy production. They are commonly used in the food industry for controlled phase separation with reduced oil migration and mobility to provide solid-like properties without high levels of saturated fats or bioactive components (Barragán-Martínez et al., 2022). As a class of organogels, oleogels are viscoelastic mixtures of liquid or solid lipids, which are mainly composed of vegetable oils and small amounts of organogel agent. Organogels are semi-solid systems consisting of self-assembled, cross-linked, or entangled gel fibres, and they might be applied in chemistry, the pharmaceutical industry, cosmetics, biotechnology, and food technology. Specifically, oleogels are solid fat-like substances with a three-dimensional network structure, dissolving a small amount (typically $\leq$10%) of a single or composite agent with liquid vegetable oil after heating and lowering the temperature below the gel point. They have the benefits of solid fats (providing unique flavour and texture, especially plasticity) without altering the health and nutritional composition of the liquid vegetable oil. One of the main advantages of oleogels is that they could provide a wide variety of oils and fats to satisfy the requirements of the human body. Since the matrix of oleogels is vegetable oil, researchers have the opportunity to choose a variety of vegetable oils (such as sunflower oils and corn oil) as raw materials according to the market demand (Ferro et al., 2021; Li, L. et al., 2021). Another crucial trait is that the oleogels are essentially free of TFA during gelation without hydrogenation. In addition, the beneficial components in vegetable oils, such as sterols and vitamin E, are stable during the formation of the oleogels. But there are doubts about whether the original characteristics of the food can be preserved. Therefore, this review focuses on the application of oleogels in foods. It would provide alternatives for saturated fats and trans fats in traditional oily foods and show how oleogels could be used to maintain nutritional content and function.

2. OLEOGEL: STRUCTURE: CLASSIFICATION, PREPARATION AND MECHANISM

2.1. Classification of oleogels

Nowadays a variety of gelators such as waxes, ethyl cellulose (EC), hydroxypropyl methyl cellulose (HPMC), and glyceryl monostearate are often employed to form oleogels. Based on the amount of gelling agent, they are classified as one-component or composite oleogels. According to the mechanism of formation of the oleogels, they can be divided into crystalline particle network oleogels, self-assembled crystalline systems, polymer network oleogels, and indirect templated system oleogels (Fig. 1).

2.1.1. Oleogels of crystal particles network. Small molecule structurants generate a three-dimensional network of crystalline particles. The crystallization of the structurant is the primary reason for the formation of networks in crystalline particle-formed oleogels. The liquid oil is

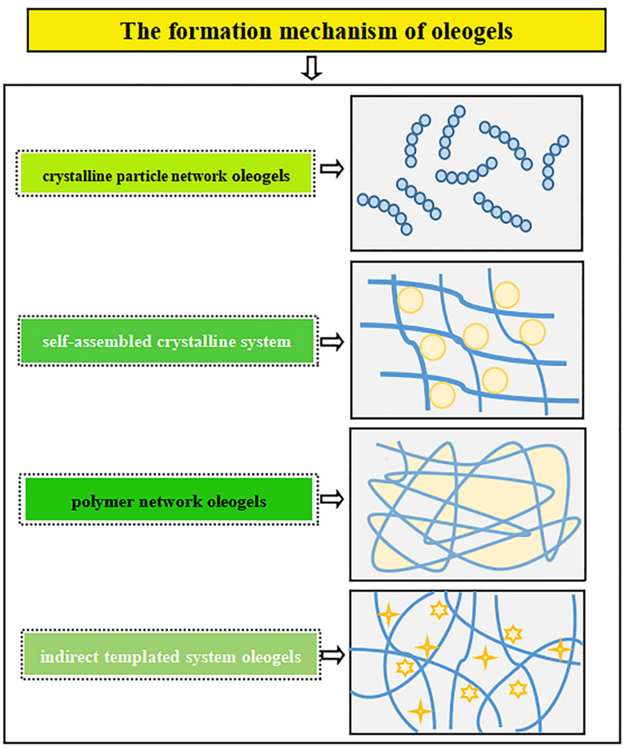

Fig. 1. The mechanism of formation of the oleogels

confined in the network structure as the crystalline particles develop to form clusters, which would create a tight crystalline network as they are cooled. Monoglycerides, diglycerides, fatty acids, fatty alcohols, biowaxes, and wax esters are the most common single component and small molecule structurants identified by the particle crystallisation principle.

One of the most common oleogels is made by wax as the structural agent, and the wax can self-assemble into a continuous crystal lattice structure that confines the liquid oil and forms an oleogel. By adding wax to the oleogel, Blake et al. (2014) discovered that the crystal chain structure became more complex and formed multiple crystal regions. Wax lipids of saturated wax ester acid with carbon chain lengths of 10–31 formed an oleogel comparable to that of fatty acid oleogels in edible oils (sunflower oil), essential oils (lavender oil), and hydrocarbons (diesel oil). As the chain length increases, the minimum gel formation concentration in liquid oils and fats decreases. At low concentrations, wax esters produce flakes or needles of crystals in edible oils, unlike saturated glycerides (Daniel and Rajasekharan, 2003). In addition, Rogers et al. (2008) investigated 12-hydroxystearic acid crystallisation in vegetable oils as well as the crystal stability of its self-assembled structure through non-isothermal nucleation and crystallisation. Phytosterols and a mixture of phytosterols and β-sitosterol are commonly employed as gelling agents. β-sitosterol, estradiol, and dihydrocholesterol are the most commonly used gelling agents. Bot et al. (2011) found that the fibrils were formed during the gelling process of oleo-hydrogels. The self-assembled tubules of phytoretinol is different from pure oleogels, it is mostly monohydric sterol crystals in the emulsion that forms fibrous structures.

2.1.2. Oleogels of self-assembly systems network. It becomes supersaturated when the organogelator-solvent melt point is below the melting point of the organogelator, leading to the self-assembly of the gelator molecules into a nucleation process. 12-hydroxystearic acid and ceramides, as self-assembling gelling agents, control the flow of liquid oil by forming spiral or twisted crystalline fibres. They can simulate the natural self-assembly and crystallisation ability of triacylglycerols controlling the hierarchical assembly of gelling agents through weak physical molecular interactions, such as hydrogen bonding, van der Waals forces, electrostatic interactions, dipole forces and hydrophobic forces (van Esch and Feringa, 2000). Bot and Agterof (2006) investigated the relationship between the gel-forming properties and the molecular structure of the oil-forming agent by using dihydrocholesterol, cholesterol, β-sitosterol, and dulcitol from sunflower oil to self-assemble into firm and transparent gels with γ-sitosterol. The gelation capacity depends mainly on the hydrogen bonding between the hydroxyl group of sitosterol and the carbonyl group of oryzanol and is also related to the degree of desaturation in the cholesterol ring structure.

2.1.3. Oleogels of the polymeric network. The fundamental condition in generating polymer oleogels is cross-linking or non-covalent self-assembly between polymeric structural components through chemical bonding. EC is the only known polymeric gelling agent that can be dispersed in oil directly, it can be modified in various ways for different applications. Also, as a derivative of a semi-crystalline polymer, it is allowed by physical binding forces to complete the sol-gel transition in the presence of liquid oil. The hydrophobic and semi-crystalline nature of EC makes it useful as a gelling agent for oils. When the temperature reaches the phase transition temperature, the EC polymer is melted and dispersed in the liquid grease. While during cooling, the ECs in the hydrophobic phase cross-link with each other when the temperature of the molten mixture is below the sol-gel transition temperature. As a result of physical forces like van der Waals forces and hydrogen bonds, they form a uniform and stable three-dimensional network structure, which binds the hydrophobic phase oil within. Zhang et al. (2019) investigated structure cinnamon oil oleogels by cross-linking EC molecules at high temperatures. As the EC viscosity increases, the structure of the oleogel is dense, which enhances the binding capacity and stability of oil. The team of Kai Zhang also explored EC of three viscosities for cinnamon essential oil (CEO) structure, the physicochemical properties of oleogels and its emulsion. The results showed that the network structure of the CEO-EC oleogels tightened as the EC viscosity increased, which was consistent with the above study. This unique behaviour regarding the sol-gel conversion experienced by the EC in the presence of a liquid oil is based on the ability of the polymers to bind through physical bonding. For example, EC was mixed with oleogels of monoglycerides (MG) and candlestick wax (CW) to improve the rheological properties of EC oleogels by enhancing EC solubility and forming hydrogen bonds between the hydroxyl groups of EC and MG (Rodríguez-Hernández et al., 2021). In conclusion, by interacting with polar functional groups in the oil phase, EC can change the functional characteristics of oleogels for food ingredients or oral administration of fat-soluble compounds.

2.1.4. Indirect templated system. There are shortcomings to direct oleogel reactions due to the high temperature process of preparing, leading to oxidation and mass loss of oil. Therefore, some studies have explored using indirect template methods to prepare oleogels, mainly including emulsion template methods (Erinç and Okur, 2021), foam template methods (Oh et al., 2019), or solvent exchange methods (Davidovich-Pinhas et al., 2016). In the emulsion template method, the emulsion is commonly employed as a template for the preparation of oleogels. The specific process forms an emulsion with high oil content, and the interaction between the polymer of emulsifiers in the droplet forms a network structure in the emulsion system following certain treatments. The continuous phase is composed of solid fat crystals and the aqueous phase remains in the gel network, then it is dried to remove the water content to obtain the oleogel.

However, the disadvantage of the indirect method of gel formation is that additional ingredients such as proteins are introduced during the preparation process and the oleogel sample must be dried to remove the aqueous phase. Nevertheless, due to interface destabilisation, the process of dehydration may cause aggregation of the oil phase and oxidation of unsaturated oils, which is not beneficial to the construction and stability of oleogel systems.

2.2. Preparation of oleogel

There are three main methods of constructing oleogels including direct dispersion, indirect methods, and oil adsorption (Fig. 2).

Direct dispersion is often applied in solid fats and is an extension of the traditional process of dispersing wax, fatty acids, monoglycerides, and EC. They are in a liquid oil is heated to above the melting point of the gelling agent and heat it until it is completely dissolved, leaving the liquid oil to then cooled to a lower temperature by natural cooling or shearing at certain temperatures. As a result of the cooling process, nucleation and crystallization can occur and thereby Polymerization to form self-assemblies are formed. Also, the study of Zhao et al. (2020) showed that single component oleogels hardly mimic the properties of traditional solid fats, while mixtures of oleogel agents can generate more desirable oleogels by strengthening the network.

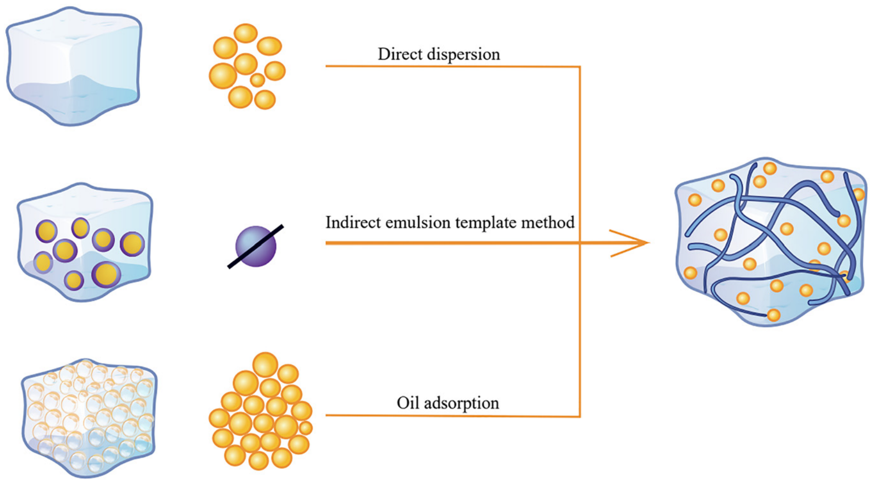

Fig. 2. The methods of constructing oleogels. The small circles represent oil; the big circles represent water; the 'traces' in the dispersed phase refer to their structure

The indirect method involves the addition of a polymer to an aqueous solvent or continuous emulsion to structure the vegetable oil into an emulsion gel, and then the water is removed by freeze-drying or heating-drying to maintain the tightness of the gel grid structure. This structure is formed to confine large quantities of liquid oil without any oil leakage even during long storage processes. This method has attracted increasing interest as the preparation of oleogels with different properties and structures by selecting colloidal particles with various characteristics and modulating the features of the template emulsion. Patel et al. (2014a) prepared emulsion templates using gelatine and xanthan gum as emulsifiers at an oil: water ratio of 6:4 and then obtained oleogels with very high oil content by oven drying or freeze-drying. Another method of gelation is the oil adsorption method using materials where the structure agent forms a porous structure in the aqueous phase or by increasing the density of the fluid near the interface to construct an oleogel. The preliminary study found that it could replace hydrogenated oil as a stabilizer for peanut butter and partially or fully replace oil for cookie frosting (Tanti et al., 2016). The report by Patel et al. (2015) summarized and conducted preliminary studies on gel fats and oleogels constructed from water-soluble food polymers by emulsion template and foam template methods. They found that MC and xanthan gum, gelatine and xanthan gum could transform liquid vegetable oils into high strength ($G' > 4{,}000$ Pa), thixotropic soft solids and gel fats and oils at the oil-water interface through interactions oleogel (with liquid oil content above 97%). The preparation technique is green and friendly without chemical modification or chemical cross-linking agent addition, and it cans replacement of traditional margarine and shortening for baking applications. In spite of its potential, this method is required to the control the oxidation of oils and fats during the drying and dehydration process of gel grease construction. But the mechanism of cross-linked networks stabilizing liquid oils between biomolecular chains is ambiguous. Essentially, the process of preparing oleogels by adsorption is to enrich material near the interface, or to increase fluid density. Porous additives and other absorbent fillers with high specific surface area are commonly included in food formulations to accumulate and confine excess water to increase consistency, flow, and texture in different products. Patel et al. (2015) successfully modelled the creation of porous cryogels using hydroxypropyl methylcellulose to form aqueous foam as a template. Because of its high porosity, the porous cryogel can absorb oil at more than 100 times its weight, and this oil-absorbing material is subsequently sheared to form an oleogel containing 98 wt% liquid oil. This method is novel and environmentally friendly, its preparation involves water foaming, freeze drying, and shearing without high temperatures, additives, harsh chemicals, or cross-linking agents.

3. APPLICATION IN FOOD

The use of oleogels in baked goods is ever-increasing due to their low saturated fatty acid, low trans fatty acid, and oxidative stability properties. Table 1 lists the applications of oleogels in meat products, margarine and shortening, ice-cream, chocolate in recent years.

Since fat is an important component of bakery products (bread, biscuits, pastries, etc.), it plays a vital role in the texture, sensory and flavour properties of the food. Fats may contribute to the development of various health problems such as obesity, cardiovascular diseases (CVDs), metabolic syndrome, and diabetes (de Souza et al., 2015). The U.S. Food and Drug Administration has determined that partially hydrogenated oil no longer qualifies as generally recognised as safe, so it must be removed from foods by June 2018. Subsequently, the World Health Organization launched a public consultation on 7 May 2018 about draft guidelines for the intake of saturated and trans fats. The WHO recommends that saturated fats provide 10% or less of calories and trans fats provide 1% or less of calories. The guidelines aim to reduce the risk of CVDs in adults and children. Therefore, the main objective of using oleogels in baked goods is to reduce the amount of saturated fatty acids in foods.

3.1. Meat products

The oleogels are applied in meat products by replacing animal fats with healthier structured oils. Panagiotopoulou et al. (2016) formulated Frankfurters using an organogel emulsion with γ-sitosterol/β-sitosterol and sensory analysis showed no significant difference between sausages prepared with 10% sunflower oil gel compared to control sausages that were rich in saturated fatty acids. EC oleogel could also be applied to reduce the cooked frankfurter sausages instead of beef fat, it showed no significant difference in chewiness and firmness compared to the control product made with beef fat (Zetzl et al., 2012). The experiment (Oh et al., 2019) showed that the use of foam-structured HPMC as an oleogel agent successfully converted rapeseed oil into a solid oleogel and that HPMC oleogel substitution for beef tallow improved the quality attributes of meat patties. In addition, the level of saturated fatty acids in meat patties containing HPMC oleogel was significantly reduced to 15% compared to beef samples (42%). The study by Martins et al. (2020) investigated a healthy meat-based spreadable product (pâté) that reduced fat content by replacing pork fat with healthier structural oils. Finally, the results showed that replacing 30% or 60% of pork fat with linseed oleogel decreased the SFA by 21% or 55%, respectively. Although differences were found in parameters of hardness and cohesiveness, these were not sufficient to limit the possibilities to apply these techniques.

Table 1. The applications of oleogels in food

Applications		Structuring Agent	Oil phase	Effect on fatty acid content	Preparation method	Authors
Meat products	Frankfurter sausages	Beeswax	Linseed oil	SFA content of 35.15 g/100 g reduced to 32.34 g/100 g	Direct dispersion	Martins et al. (2020)
	Bologna sausages	Glyceryl monostearate	Sunflower oils	The reductions in SFA were 15.8%, 30.9%, 57.4%, and 71.9%, respectively, compared to the control group.	Direct dispersion	Ferro et al. (2021)
	Bologna sausages	Pork skin	High oleic sunflower oil (HOSO)		Direct dispersion	da Silva et al. (2019)
	Meat patties	HPMC	Canola oil		Indirect methods (foam template method)	Oh et al. (2019)
Margarine and shortening	Shortenings	EC	Sunflower oil and palm stearin	33–42% reduction in SFA and 2.85% reduction in TFA	Direct dispersion	Naeli et al. (2022)
	Shortenings	EC, monoglycerides, and candelilla wax	HOSO		Indirect methods	Rodríguez-Hernández et al. (2021)
	Margarines	Monoglyceride stearate			Direct dispersion	Wang et al. (2021)
Ice-cream	Ice-cream	EC	HOSO		Direct dispersion	Munk et al. (2018)
	Ice-cream	Rice bran wax (RBW)	HOSO		Direct dispersion	Zulim Botega et al. (2013b)
Baking products	Maize tortillas	Candelilla wax	Canola oil		Direct dispersion	Vernon-Carter et al. (2020)
	Gluten-free muffins	Plant and bees waxes	Rapeseed oil	More than three times lower in fatty acids	Direct dispersion	Kupiec et al. (2021)
	Cookies	Glyceryl monostearate	Camellia oil		Indirect methods (emulsion template method)	Pan et al. (2020)
	Short-dough biscuits	Waxes (candelilla, rice bran, yellow and white beeswax)	High-oleic rapeseed oil		Direct dispersion	Onacik-Gür and Żbikowska (2020)
Chocolate	Chocolate	HPMC	Sunflower oil	39% reduction in saturated fat	Indirect methods (emulsion template method)	Espert et al. (2021)
	Chocolate	Monoglyceric stearate, β-sitosterol + lecithin, and EC	Corn oil		Direct dispersion	Li, L. et al. (2021)
	Chocolate spreads	HPMC and xanthan gum	Olive and sunflower oil		Indirect methods (emulsion template method)	Bascuas et al. (2021)

Fat substitution in meat products is a concern for the meat industry, it is necessary to meet nutritional requirements in addition to organoleptic requirements. A major negative impact on the nutritional aspects of meat products is the fat content and composition, so the reformulation of meat products must achieve a reduction in total fat content and an improvement in fatty acid distribution.

3.2. Production of margarine and shortening

Oleogels as fat phase substitutes provide the ideal functional and organoleptic properties for the production of healthier margarines and shortenings without TFA and reduced SFA. Hwang et al. (2013) prepared oleogels using rice bran wax (RBW) and soybean oil and obtained very soft margarines. Shortenings used in the bakery and confectionery products are usually with high concentrations of SFA and TFA. Oleogel constructed with sunflower oil, methyl cellulose, and xanthan gum (Patel et al., 2014b) was employed as a complete or partial substitute for shortening in the production of bakery products, and the function of oleogel provided significantly better rheological and textural properties than the oil or commercial shortenings. Yılmaz and Öğütcü (2014) compared olive oil oleogels with breakfast margarine and concluded that oleogels with 7% or less sunflower wax and 3% or less bees wax could provide similar properties with spreadable margarine. However, when measuring the melting point and crystallisation point of the oleogels, it was found that the melting temperature of the oleogels depended on the amount of wax. Both oleogels were thermally reversible gels, and phase separation might occur in the preparation process. Polarised light micrographs (PLMs) of the resulting oleogel samples are shown to demonstrate the structure of the oleogels (Fig. 3). OB3, OB7, OB10 stand for oleogels made from olive oil and beeswax (BW) added at 3%, 7% and 10%; OS3, OS7, OS10 stand for oleogels made from olive oil and sunflower wax (SW) added at 3%, 7% and 10%. It can be seen

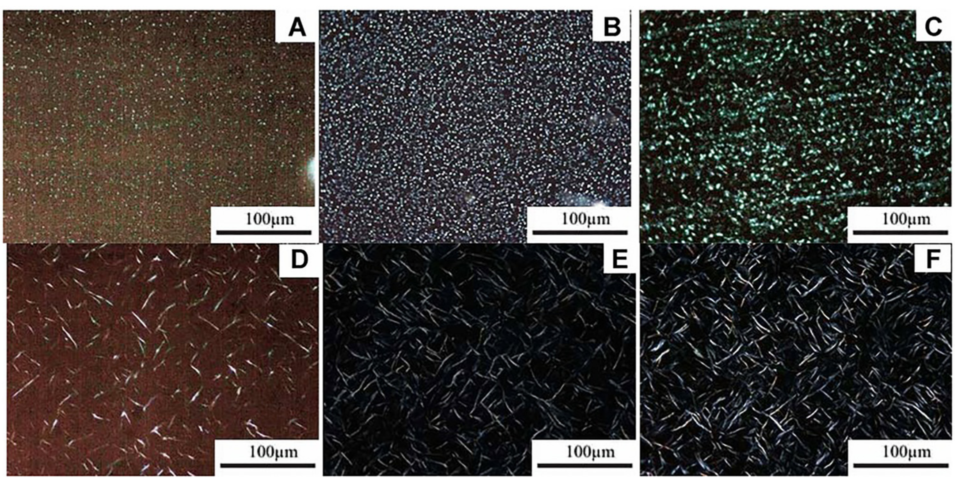

Fig. 3. Polarised light microphotographs (PLM) of the organogel samples: (A) OB3, (B) OB7, (C) OB10, (D) OS3, (E) OS7, and (F) OS10. Reproduced with permission from (Yılmaz and Öğütcü, 2014). Copyright 2014 Champaign, III. Institute of Food Technologists

that SW has longer fibrous crystals in the oleogels system compared to BW. As a result, one can expect that such fibrous, needle-like crystals would form stronger, stiffer gels with higher melting and crystallisation points. Moreover, the use of oleogels in the preparation of creams is effective in reducing fat content. The oleogel-based margarine had a similar appearance, better thermal stability, softer body, 44% less SFA, 92% less TFA, reduced total fat content, and better nutritional properties than commercially available margarines (Da Silva et al., 2018). However, these studies predominantly focused on performance characterisation and lack of the separation and melting process of friction force due to mixing during production. In conclusion, the application of oleogels rich in nutrients or active ingredients in margarine and shortenings with their green, safe, and non-polluting nature not only reduces the content of trans and saturated fatty acids, but also increases their nutritional value, extends their shelf life and provides lower fat content.

3.3. Production of ice-cream

Ice cream consists of fat spheres and air bubbles dispersed in partially frozen water. As a result of the freezing and churning process, the fat globules form a partially merged network, stabilising the merged bubbles and changing the texture and flavour of the product. The cream of ice cream is mainly composed of saturated fat to provide to the product structure. Oleogels have excellent rheological properties and are thermally reversible, so they can provide a source of stable unsaturated fatty acids for ice cream mixes. Many ice cream recipes are designed without dairy fats, so the use of oleogels is considered to build an appropriate fat structure as a substitute for non-dairy saturated fats. Moreover, the study of Zulim Botega et al. (2013a) showed that the oleogel structure in ice cream was enhanced under the right conditions by unsaturated structuring agents either rice bran wax (RBW) or glycerol monooleate (GMO) at sufficiently high oleogel concentrations in the mixture. The ultrastructure and crystallisation process of oleogel droplets in ice cream was also studied by transmission electron microscopy (Fig. 4). The addition of GMO to the mixture resulted in the formation of RBW oleogel droplets with a homogeneous shape and crystals inside. Carnauba wax is presented by long, thin crystals around the droplet interface. This may be a thin layer of submicron crystals, with visible large crystals on the oleogel droplets. Carnauba wax forms single molecule crystals, and it can contribute to the stability of the carnauba wax oleogel droplet and prevent aggregation. Ice cream made with oleogel produces small spherical gas chambers with smaller bubbles and more delicate structure than that without oleogel. But few studies have clearly clarified whether oleogel could increase the anti-thaw properties of ice cream and prolong its shelf life.

The ice creams were successfully prepared by with EC of different viscosities (cP10 and cP20) as oleogels for high oil content sunflower oil (HOSO) ice creams (Munk et al., 2018). Differences of the ice cream spheres prepared from EC cP10 and EC cP20 and their resistance to agglomeration resistance were observed by microscopy., T with the size and morphology of the EC cP20 oleogel spheres being were similar to those of ice cream made from coconut oil (Fig. 5). The micrographs clearly showed that the spherical droplets were reduced in B and C compared to A. EC hindered agglomeration because the ice cream made from of unstructured HOSO which consisted of large spherical oil droplets. Hence, the premise that oleogels currently seem to be a promising fat substitutes but it depends on is to control the concentration and type of oil used.

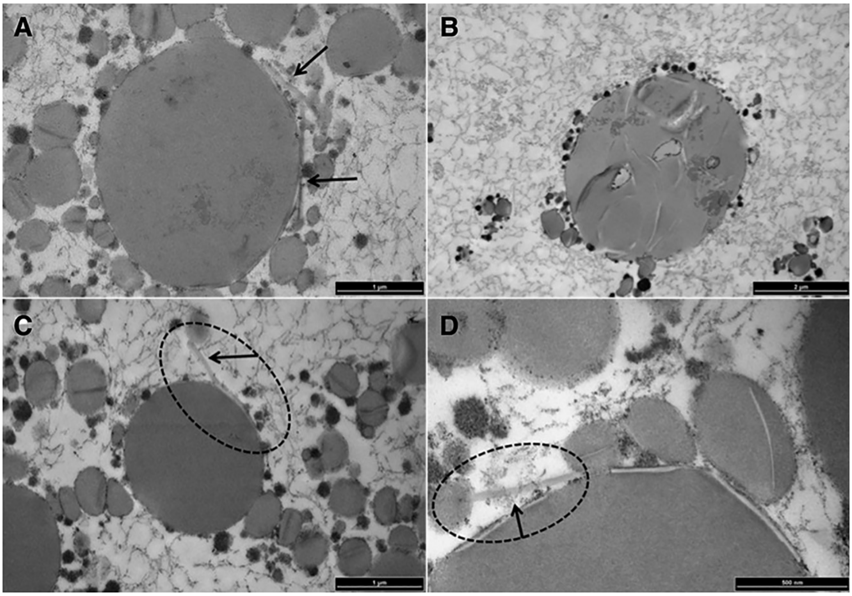

Fig. 4. Transmission electron micrographs of rice bran wax (RBW) oleogel ice cream mix prepared with glycerol monooleate (GMO). The RBW crystals can be seen projecting out of the fat droplet membrane (pointed by arrows). Scale bar = 1 μm (A and C), 2 μm (B), and 500 nm (D). Reproduced with permission from (Zulim Botega et al., 2013a) Daniele C. Zulim Botega. Copyright 2013 Champaign, III. Institute of Food Technologists

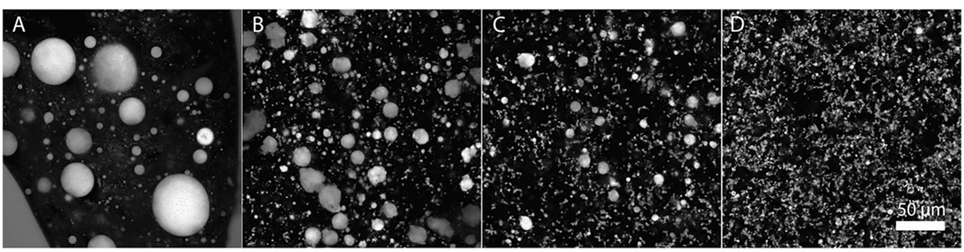

Fig. 5. Confocal laser scanning micrographs showing size, morphology and structure formation of lipid droplets/globules in molten ice cream made with (A) unstructured HOSO, (B) EC cP10 oleogel, (C) EC cP20 oleogel, and (D) coconut fat. A, B, and C is emulsified by unsaturated monoglycerides rich in oleic acid and D is emulsified by mono-diglycerides of saturated fatty acids. The lipid fraction is stained with Bodipy 493/503. The reduced size of spherical droplets in B and C, compared to A, shows that EC is retarding coalescence. Reproduced with permission from (Munk et al., 2018). Copyright 2018 Champaign, III. Institute of Food Technologists

3.4. Baking products of bread and biscuit

Although the fat in food can provide good structure, special flavour and excellent taste, it can be unhealthy. Oleogels can be used instead of fat as a binder in baking products including cakes, biscuits, pastries, and loaves of bread. In cakes, fat increases the softness of the leavening agent and traps air cells in the creaming process. In biscuits, fats are used to lubricate and coat flour particles to prevent water absorption, as well as the development of starch and gluten to achieve fine crumbs (friable texture) and a soft, supple texture. Therefore, use of oleogels with solid-phase structures is promising in bakery products. The ratio of saturated to unsaturated fatty acids was significantly reduced from 2.81 to 0.41 when oleogels were incorporated into muffin formulation (Lim et al., 2017). Furthermore, Vernon-Carter et al. (2020) showed that oleogel incorporation improved the texture of tortillas, delayed mould growth, reduced the *in vitro* digestibility of starch, and had a beneficial effect on the texture and starch digestibility characteristics of tortillas. Different types of oleogels were prepared using hydroxypropyl methyl cellulose (HPMC), monoacylglycerol (MAG), sodium stearoyl lactate (SSL), rice bran wax (RBW), and beeswax (BW) as gelling agents and were applied to biscuits. It was concluded that the oleogels made with MAG and RBW as structure agents produced biscuits with a homogeneous porous structure, desirable colour, and crispy texture (Li, S. et al., 2021). Despite the use of oleogels in baked goods and its development potential, there is a lack of data on the complex interactions between components and changes in their properties during storage.

3.5. Chocolate

Among the solid particles in chocolate (cocoa powder, milk particles, and sugar crystals) there is a crystalline fat matrix dominated by cocoa butter (CB). When saturated fats, such as coconut oil or palm oil, are used in chocolate products, they affect the product's texture, which gives the chocolate its delicate melt-in-your-mouth softness. Despite its fantastic value, cocoa butter has high saturated fat content and can potentially cause adverse effects on human health. In addition, the oil mobility of oleogels is lower than that of raw chocolate during storage. Preventing fat mobility is a crucial part of maintaining the storage quality of these confectioneries, which can suffer a quality loss due to fat mobility.

The study of Fayaz et al. (2017) investigated the feasibility of partial replacement of the fat phase in chocolate sauce by pomegranate seed oil oleogels. The oleogels were prepared by mixing the oils with palm oil in a 1:1 ratio with monoglycerides (MG), beeswax (BW), and propolis wax (PW) as structuring agents. Fourier infrared spectroscopy (FTIR), X-ray-powder diffraction analysis XRD, and rotational rheometry were employed to investigate the strength and network formation of the palm oil-oleogel system and chocolate paste. Based on the results, MG, BW, and PW had different chemical properties, which led to different crystalline networks in palm oil-oleogel systems. The possibility of replacing solid coconut fat with an oleogel of olive and sunflower oils has been provided (Bascuas et al., 2021). Moreover, the study illustrates how the ingredients are distributed in the spreads based on confocal laser scanning microscopy. Some of the diffused components form small balls, while others are uniformly distributed and interact with the components of the matrix. During the diffusion process of 100% replacement of coconut fat with olive oleogels and sunflower oleogels, part of the fat binds to the protein, forming a continuous phase. However, the shear force used will lead to the structured and uniform diffusion of some undissolved particles, resulting in instability of early coalescence, which may have an impact on appearance, shelf life, and stability. However, the effect of emulsion processing on interface stability was not mentioned. In another study, by adding cocoa butter for to chocolate effectively reduced fat saturation, which has a positive effect on improving both physical and blooms stability of chocolate under different storage conditions (Li, L. et al., 2021). A number of questions regarding heat resistance of the chocolate remain to be addressed.

Above all, the gelation of oils and fats offers the possibility of to reducing the fat migration rate and provides a reference for the wide application of oleogels of from low saturated and trans fatty acid contents in the chocolate industry.

4. CONCLUSIONS

Oleogels are studied for structure, preparation, character and application in food product. Oleogels with texture and flavour similar to that of fats can be prepared by immobilising vegetable oils in a three-dimensional network with a gelling agent. As a result of the crystallisation of oleogels, highly anisotropic structures are formed, such as fibres, plates, strips, tubules, and filaments. There are still many issues to be resolved due to the different structures of the oleogels, first of all, whether the incorporation of oleogels has an influence on the structure of other components in the food. Moreover, the effect of oleogels on the fatty acid composition is to be explored in food products. Such information could be utilised in the manufacture of healthy foods. Although oleogels are not part of our daily diet, oleogels offer new possibilities and platforms for the ingredients of food. In conclusion, this study could contribute to the understanding and rational development of oleogels and expand the scope of oleogels in food-related applications.

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